INTERPARTICLE FORCES IN FLUIDISED
BED FILTRATION OF HOT GASES

by

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A dissertation submitted for the Degree of Doctor of Philosophy
in the University of Surrey

February, 1987
"So many hundred hands in this mill, so many hundred horse steam power. It is known, to the force of a single pound weight, what the engine will do; but not all the calculators of the National Debt can tell me the capacity for good or evil, for love or hatred, for the decomposition of virtue into vice, or the reverse."

Charles Dickens

of Bounderby's Mill in

"Coketown"

To my wife, for putting up with it.
Summary

The lack of a suitable system for hot gas cleaning is the greatest obstacle to the development of gasification combined-cycle power generation. In this work, a shallow 0.15 m diameter fluidised bed of 1 mm sand particles was used as a filter for 0.5-10 μm gasifier char particles redispersed in nitrogen at 700-900°C and 1 bar a. Earlier work has suggested that, for a correctly-designed low free-area distributor, initial collection efficiencies greater than 99% at 2 μm could be realised but that poor retention limits the overall filtration efficiency. As an aid to retention, heavy fuel oil was sprayed continuously into the bed using a concentric-tube atomiser. With this arrangement, theoretically-predicted filtration efficiencies were approached for particles below 7 μm in size, but the overall performance was seriously marred by secondary aerosol originating from the addition of the oil itself. The results suggested that two modes of filter operation are possible: at lower gas and retention-aid flowrates the challenging aerosol is retained on the collector; at higher gas and retention-aid flowrates the challenging aerosol is agglomerated and re-emitted from the bed in a distribution with a larger mean size.

The operation of various instruments for particle size measurement below 10 μm has been reviewed, and their capabilities have been compared by experiment. The method selected (collection in a liquid impinger, followed by off-line analysis by "Coulter Counter") is described in detail.

The addition of a liquid retention-aid to a fluidised bed can cause modification of its fluidisation behaviour, leading eventually to catastrophic defluidisation. Theoretical and experimental aspect of these effects are discussed, leading to the development of an experimental method for direct measurement of interparticle forces, and an expression for the transition condition between Geldart’s (1973) groups A and B.
ACKNOWLEDGEMENTS

This work could not have been completed without the cooperation, advice and encouragement of a large number of people and organisations:

the Coal Research establishment, Stoke Orchard, who financially supported both me and this work - personal thanks are due particularly to Brian Robson, Graham Reed, John Whitehead, Mike Arnold and Chris Bower, all staff of the Establishment;

the technical staff of the Department of Chemical and Process Engineering - in particular, Ken George, Austen Harrup and Peter Pennington were of great assistance in designing and building my experimental equipment;

the staff of the Microstructural Studies Unit and the Audio Visual Aids Unit, for their painstaking electron microscopy and photographic work;

the secretarial staff of the Department - Babs Armstrong for typing the initial part of this thesis, and, in particular, Beryl Dolloway, who achieved miracles in putting it into its final form in a remarkably short time (and didn't complain about my writing);

my fellow graduate students (a fine body of men) - Barry Tan, Laurie Scandrett, Wing Cheung and, above all, José Coury, with whom I shared many argumentative, happy and illuminating hours;

Kelly Thamba Muthu, who advised me to do a Ph.D.;

my academic colleagues in the Department - in particular, Mojtaba Ghadiri (who designed and built much of the equipment I have gone on to use), Ugur Tüzün and Ron Schulz (for much good advice);

and lastly, Roland Clift, my supervisor, colleague, and friend, without whom I would never have started it.
PREFACE

The work described in this dissertation was carried out in the Department of Chemical and Process Engineering of the University of Surrey, between October 1981 and February 1987. It is the original and independent work of the author, except where otherwise stated in the text. No part of this dissertation has been submitted for a degree at any other University.

J P K Seville
February 1987
Interparticle Forces in Fluidised Bed Filtration of Hot Gases
J P K Seville

Addendum

p. 53 The overall collection efficiencies reported by Nienow and Killick (1983), who collected potassium permanganate aerosols in beds of bronze shot, are considerably lower than those which would be predicted using Thambimuthu's (1980) correlation for collection in the jet region. Three possible explanations for the disagreement are presented here:

(i) the possibility of "rebound" or bounce of the solid aerosol particles from the collector;
(ii) a different mode of gas entry to the bed, i.e. bubbling rather than jetting (Thambimuthu's correlation is based on measurements with particles of lower density, for which jetting would have been more likely);
(iii) reduced entrainment of collector particles into the jet due to the formation of interparticle liquid bridges.

Although Nienow and Killick (1983) investigated the effects of water addition to the bed (primarily to reduce static electrification), continuous water addition was not employed in the experiments which are reviewed in this thesis (Nienow, 1987). Thus, possibility (iii) above is eliminated. Moreover, it is clear from work reported after the writing of this thesis (Nienow and Killick, 1987) that the filtration efficiencies reported earlier by Nienow and Killick (1983) were subject to considerable reduction due to bounce. Using a high free-area distributor such as the one employed in the work reported in Chapter 6, Nienow and Killick (1987) were able to improve the overall filtration efficiency from about 70% to 98% by continuous addition of a suitable retention aid (polyethylene glycol). The latter figure is in good agreement with the predictions of the Thambimuthu (1980) correlation. It therefore appears that suggestion (i) above is the most likely explanation for the discrepancy between Nienow and Killick's (1983) results and those of Thambimuthu and others.
The term in square brackets on the right hand side of the particle momentum balance (4.24) represents the total interaction between the particles and the fluid, i.e. the sum of the buoyancy reaction and the hydrodynamic drag contributions. The two terms following the square bracket represent the gravitational force and the interparticle force contribution, respectively. Anderson and Jackson (1967) pointed out that the momentum balances for the dispersed and continuous phases should include the terms $V_R g$ and $V R g$, respectively, where

$$
\bar{R}_g = \rho_g \left[ (1-\varepsilon) v' v' \right]
$$

and

$$
\bar{R}_g = \rho_g \left[ \varepsilon u' u' \right]
$$

$v'$ and $u'$ are the turbulent fluctuating velocity components for the particle and fluid velocities, respectively. Thus, $\bar{R}_g$ and $\bar{R}_g$ are effectively Reynolds stresses. Anderson and Jackson argue that since their effects are the same as those of the stress tensors $\bar{e}_g$ and $\bar{e}_g$, then they can be included by defining new stress tensors incorporating both the $\bar{e}$ and the $\bar{R}$ terms. (Note that $\bar{R}_g$ must be zero if there is no interaction between the particles, which is obviously not the case here.) Rietema and van den Akker (1983) show that it is not formally permissible to eliminate $\bar{R}_g$ in this way, but do not suggest an alternative way of accommodating it within the momentum balance. Both Reynolds stress terms have been omitted from the arguments which follow because their contributions will be small (and thus not likely to alter the form of the stability analysis), because there is no agreed form for either term, and because explicit inclusion of these terms increases complication without improvement in clarity.

References (additional to those on p. 326 et seq.)


J. P. K. Seville
1st June 1987

1Note, however, that the two terms within the bracket do not simply represent the individual buoyancy and drag terms (see, for example, Clift et al., 1987).
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<td>--------</td>
<td>------------</td>
<td></td>
</tr>
<tr>
<td>A</td>
<td>projected or cross-sectional area ($m^2$), Hamaker constant (J)</td>
<td></td>
</tr>
<tr>
<td>a</td>
<td>particle separation distance (m)</td>
<td></td>
</tr>
<tr>
<td>b</td>
<td>bubble frequency (Hz)</td>
<td></td>
</tr>
<tr>
<td>c</td>
<td>Cunningham &quot;slip correction factor&quot; (-), number concentration of aerosol particles ($m^{-3}$)</td>
<td></td>
</tr>
<tr>
<td>c</td>
<td>particle hydrodynamic resistance (m), cohesion constant ($N m^{-2}$)</td>
<td></td>
</tr>
<tr>
<td>D</td>
<td>particle diffusivity ($m^2/s$)</td>
<td></td>
</tr>
<tr>
<td>d</td>
<td>particle diameter (m)</td>
<td></td>
</tr>
<tr>
<td>E</td>
<td>single-particle collection efficiency (-), particle aspect ratio (-), elasticity of expanded bed ($N m^{-2}$), energy (J)</td>
<td></td>
</tr>
<tr>
<td>e</td>
<td>coefficient of restitution (-)</td>
<td></td>
</tr>
<tr>
<td>F</td>
<td>interparticle force (N), drag force per unit volume ($N m^{-3}$)</td>
<td></td>
</tr>
<tr>
<td>F_H</td>
<td>force exerted at single contact, irrespective of direction (N)</td>
<td></td>
</tr>
<tr>
<td>F_T</td>
<td>tensile force transmitted per particle (N)</td>
<td></td>
</tr>
<tr>
<td>f</td>
<td>aerosol penetration ($l = 1 = \eta$) (-)</td>
<td></td>
</tr>
<tr>
<td>G_p</td>
<td>particle weight ($= \pi d^3 (\rho_p - \rho_g) g/6$) (N)</td>
<td></td>
</tr>
<tr>
<td>g</td>
<td>gravitational acceleration ($ms^{-2}$)</td>
<td></td>
</tr>
<tr>
<td>h, H</td>
<td>height (m)</td>
<td></td>
</tr>
<tr>
<td>h_j</td>
<td>height of jet region (m)</td>
<td></td>
</tr>
<tr>
<td>h_w</td>
<td>Lifshitz constant ($= 4/3 \pi \lambda$) (J)</td>
<td></td>
</tr>
<tr>
<td>l</td>
<td>length (m)</td>
<td></td>
</tr>
<tr>
<td>m</td>
<td>particle refractive index ($= n_1 - n_2 i$) (-)</td>
<td></td>
</tr>
<tr>
<td>N_or</td>
<td>orifice number ($= S_{or} N_{UC}^{1/3}$) (-)</td>
<td></td>
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</table>
\( \text{Nuc} \) dimensionless collector particle terminal velocity
\( \left( = \frac{3\rho_g u_{tc}^3}{4 (\rho_c - \rho_g)} \right) \)

\( n \) number of distributor orifices (\( - \)), number of particles per unit area in failure plane (\( \text{m}^{-2} \))

\( P, P \) pressure (\( \text{Nm}^{-2} \))

\( Q \) gas volumetric flowrate (\( \text{m}^3\text{s}^{-1} \))

\( Q_b \) volumetric bubble flowrate (\( \text{m}^3\text{s}^{-1} \))

\( q_a \) charge on aerosol particle (\( \text{C} \))

\( R \) particle radius (\( \text{m} \))

\( Re \) Reynolds number (\( = \frac{\rho g Ud}{\mu} \)) (\( - \))

\( R_v \) aerosol collection rate per unit bed volume (\( \text{s}^{-1}\text{m}^{-3} \))

\( r \) radius of curvature of liquid bridge (\( \text{m} \))

\( S \) surface area (\( \text{m}^2 \))

\( St \) Stokes number (\( = \frac{C \rho_d a^2 U}{9 \mu d_c} \)) (\( - \))

\( St_{or} \) Stokes number for jet collection (\( = \frac{C \rho_d a^2 U_{or}}{9 \mu d_c} \)) (\( - \))

\( T \) temperature (\( ^\circ\text{C} \) or \( \text{K} \)), limiting tensile stress (\( \text{Nm}^{-2} \))

\( T_s \) sintering temperature (\( ^\circ\text{C} \) or \( \text{K} \))

\( U \) superficial gas velocity (\( \text{ms}^{-1} \))

\( u \) gas velocity, slip velocity (\( \text{ms}^{-1} \))

\( u_t \) terminal particle velocity (\( \text{ms}^{-1} \))

\( V \) volume (\( \text{m}^3 \))

\( v \) particle velocity (\( \text{ms}^{-1} \))

\( W \) interaction potential (\( \text{J} \))

\( X \) "cross-flow factor", number of transfer units, defined in Appendix Al (\( - \))

\( Y \) number of transfer units per layer of collector, defined in Appendix Al (\( - \)), Young's modulus (\( \text{Nm}^{-2} \))

\( z \) distance (\( \text{m} \))
<table>
<thead>
<tr>
<th>SYMBOL</th>
<th>DESCRIPTION</th>
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</thead>
<tbody>
<tr>
<td>ALPHA</td>
<td>$\alpha$ particle size parameter ($=\eta d/\lambda$)(-)</td>
</tr>
<tr>
<td>BETA</td>
<td>$\beta$ liquid bridge half-angle (°), fraction of gas within the bubble phase (-)</td>
</tr>
<tr>
<td>GAMMA</td>
<td>$\gamma$ surface tension (N/m)</td>
</tr>
<tr>
<td>DELTA</td>
<td>$\delta$ thickness of raining layer (m), elastic deformation (m)</td>
</tr>
<tr>
<td>EPSILON</td>
<td>$\epsilon$ voidage fraction (-)</td>
</tr>
<tr>
<td>ETA</td>
<td>$\eta$ efficiency</td>
</tr>
<tr>
<td></td>
<td>$\eta_1$ grade efficiency (-)</td>
</tr>
<tr>
<td>THETA</td>
<td>$\theta$ light-scattering angle (°)</td>
</tr>
<tr>
<td>KAPPA</td>
<td>$\kappa$ rate constant for filtration (m⁻¹)</td>
</tr>
<tr>
<td>LAMBD A</td>
<td>$\lambda$ molecular mean free path (m), wavelength of light (m)</td>
</tr>
<tr>
<td>MU</td>
<td>$\mu$ gas viscosity (kg m⁻¹s⁻¹)</td>
</tr>
<tr>
<td>NU</td>
<td>$\nu$ Poisson's ratio (-)</td>
</tr>
<tr>
<td>RHO</td>
<td>$\rho$ density (kg m⁻³)</td>
</tr>
<tr>
<td>SIGMA</td>
<td>$\sigma$ normal stress (Nm⁻²)</td>
</tr>
<tr>
<td>TAU</td>
<td>$\tau_{ij}$ stress (normal stress if $i=j$)(Nm⁻²)</td>
</tr>
<tr>
<td>PHI</td>
<td>$\varphi$ angle of internal friction (°), $\varphi_s$ particle sphericity ((-))</td>
</tr>
<tr>
<td>SUBSCRIPTS</td>
<td></td>
</tr>
<tr>
<td>------------</td>
<td>---------------------------------</td>
</tr>
<tr>
<td>A</td>
<td>projected-area-equivalent</td>
</tr>
<tr>
<td>MTT</td>
<td>attrition product</td>
</tr>
<tr>
<td>a</td>
<td>aerosol particle</td>
</tr>
<tr>
<td>B</td>
<td>bubble phase, bulk (density)</td>
</tr>
<tr>
<td>b</td>
<td>bubbling bed</td>
</tr>
<tr>
<td>C</td>
<td>capillary</td>
</tr>
<tr>
<td>c</td>
<td>collector particle</td>
</tr>
<tr>
<td>D</td>
<td>diffusion</td>
</tr>
<tr>
<td>DI</td>
<td>direct interception</td>
</tr>
<tr>
<td>e</td>
<td>equilibrium</td>
</tr>
<tr>
<td>G</td>
<td>gravity</td>
</tr>
<tr>
<td>g</td>
<td>gas</td>
</tr>
<tr>
<td>H</td>
<td>at height H</td>
</tr>
<tr>
<td>I</td>
<td>impaction</td>
</tr>
<tr>
<td>IN</td>
<td>entering filter</td>
</tr>
<tr>
<td>j</td>
<td>jet or jet region</td>
</tr>
<tr>
<td>MB</td>
<td>minimum bubbling</td>
</tr>
<tr>
<td>MF</td>
<td>minimum fluidisation</td>
</tr>
<tr>
<td>OUT</td>
<td>leaving filter</td>
</tr>
<tr>
<td>or</td>
<td>orifice</td>
</tr>
<tr>
<td>p</td>
<td>particle</td>
</tr>
<tr>
<td>S</td>
<td>Stokes, solid</td>
</tr>
<tr>
<td>SO</td>
<td>Aerodynamic</td>
</tr>
<tr>
<td>t</td>
<td>terminal conditions</td>
</tr>
<tr>
<td>V</td>
<td>volume-equivalent</td>
</tr>
<tr>
<td>W</td>
<td>van der Waals</td>
</tr>
<tr>
<td>50</td>
<td>at 50% point</td>
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Chapter 1

Introduction

1.1 Power from Coal

Coal is the foundation upon which our modern industrial society is based. Before the widespread use of coal as a fuel, manufacturing industry existed only in small isolated areas, where local conditions were favourable. The iron industry in Sussex, for example, depended on water power and a supply of local wood, which were both in strictly limited supply. The late 18th and early 19th centuries saw a profound shift from the technology of wood, wind and water to the technology of "carboniferous capitalism" (Mumford, 1934). The development of the coal-fired steam engine meant that power became freely available; industries were no longer tied to a few suitable locations, and expansion was rapid. In turn, steam power revolutionised coal mining and stimulated the transport revolution which transformed Britain economically and socially into the country we know today. Yet the development of coal as the major source of power brought problems which, in different forms, remain with us: "on every side and as far as the eye could see into the heavy distance, tall chimneys, crowding on each other, poured out their plague of smoke" (Charles Dickens, "The Old Curiosity Shop").

The 20th century has seen a rapid increase in the use of oil and gas for power. These fuels are cheaper, easier to recover, and more convenient to use than coal, but reserves are relatively small and their widespread use is likely to be, in historical terms, no more than a brief diversion. The expansion in the use of nuclear power, on the
other hand, is almost unlimited by raw material resource constraints, but presents technical and political problems which have yet to be solved. Apart from the risk of major accident, the construction of nuclear power stations also raises economic problems; the cost of building even one station has a major effect on the economy of a country as small as the U.K. However, the choice between nuclear and coal-fired power stations in the UK is not straightforward even on economic grounds, especially since it is replacement rather than expansion of capacity which is being proposed. For example, Fothergill et al (1983) have calculated that the lower electricity prices which the U.K. Central Electricity Generating Board (CEGB) hopes will result from the operation of the Sizewell "B" Pressurised Water Reactor could be produced by a reduction in the cost of coal, rather than investment in new generating capacity. Sizewell "B" may cost £2 billion or more, including interest during construction; capital investment in the coal industry is currently only £750 million per year. Fothergill et al maintain that even the largest reduction in electricity prices made possible by Sizewell "B" could instead be achieved by a 3% reduction in the cost of coal to the CEGB. Such a reduction in coal prices, by higher investment and productivity, might be attained at less cost than the construction of a new power station. (The "cost" considered by Fothergill et al is the total cost to the community, which takes into account the cost of unemployment.)

The next generation of advanced coal-fired power stations will operate on combined steam-turbine and gas-turbine cycles, of which many variants have been proposed. The conversion of coal to fuel gas which could then be used to run a gas turbine was pioneered by Ludwig Mond in the late 19th century (Squires, 1972), but was not then economic because of the very low conversion efficiencies of gas turbines. The
development of gas turbines for aircraft led to an increase in the tolerable turbine inlet temperature (and hence in the conversion efficiency) due to a combination of advances in metallurgy and in blade design. Turbine inlet temperatures in excess of 1300°C are conceivable for the 1990s (Gluckman and Louks, 1982).

Two basic types of combined-cycle power generation have been considered:

(i) the pressurised fluidised bed combustion (PFBC) process with gas-turbine/steam-turbine combined-cycle; and

(ii) the coal gasifier with gas-turbine/steam-turbine combined-cycle.

In PFBC, coal is burned in a pressurised fluidised bed combustor into which heat-transfer surfaces are inserted. Water is supplied to the surfaces and steam is raised in a conventional steam cycle, thus generating electrical power in the conventional way. A further, smaller contribution to the electrical output is made by a gas turbine which is driven by the hot compressed combustion gases. The life of the turbine depends critically on prevention of corrosion and erosion of the blades, which requires a high efficiency of removal of particulate material and aggressive chemical species from the entering gas. Turbine entry conditions are typically 10 - 20 bar and 850 to 900°C. The PFBC cycle operated by the CEGB/NCB at Grimethorpe in Yorkshire is shown schematically in Figure 1.1 (Lawton et al, 1985). A major advantage of this technology over conventional coal-fired power stations is the ability to retain sulphur in the combustor by addition of pulverised limestone or dolomite. Over 90% of the sulphur in the fuel can be removed by reaction to calcium sulphate. The reduced level
of excess oxygen and lower bed temperature, by comparison with conventional technologies, means that the production of NO\textsubscript{x} is also greatly reduced in the PFBC.

The alternative to PFBC, which is even more promising economically, is the gasification combined-cycle (Robson, 1977), in which coal is completely or partially gasified, to produce a gas of low to medium calorific value. Residual char from the gasifier may be fed to a pressurised fluidised combustor in which steam is raised as in PFBC,
while the off-gases from the gasifier are used to fire a gas turbine, which generates most of the power. Figure 1.2 shows the particular design proposed by the National Coal Board (Arnold, 1981), in which the excess oxygen in the combustor flue gas, with the addition of a small amount of steam, is sufficient to operate the gasifier. In the gasification combined-cycle there is considerable economic incentive to operate the gasifier at higher temperatures (1000°C and above), so that gas cleaning requirements are even more stringent. The process retains the advantage of low NOx emission, and sulphur emission is reduced by operating with dolomite addition to the gasifier.

Problems which remain, but will not be discussed here, are the emissions of alkali metal compounds (predominantly sulphates in combustion and chlorides in gasification; Scandrett and Clift, 1984) and of hydrogen chloride, which is a particular problem in combustion of British coals.

Davidson and Moore (1979) compared the relative thermal efficiencies of advanced coal-burning systems, and their conclusions are presented in Figure 1.3. Their reference plant is a 500 MW unit with feed preheating and single interstage re-heat. Based on current steam-cycle practice, this gives a typical thermal efficiency of 38.7%. Simple flue-gas scrubbing to remove 90% of the sulphur and re-heating of the flue gas to 120°C reduces efficiency by about 3.5 percentage points. PFBC enables the thermal efficiency to be at least maintained while still reducing the sulphur emission by 90%. However, gasification combined-cycle clearly shows a considerable economic advantage, which will increase as turbine entry temperatures increase. The single most critical area of uncertainty in the development of the gasification combined-cycle is the clean-up of the fuel gas prior to passage through
Figure 1.2 British Coal Gasification Combined-Cycle (Arnold, 1981)

<table>
<thead>
<tr>
<th>Type of Plant</th>
<th>Overall Efficiency (%)</th>
<th>Turbine Entry Temp.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gasification Combined Cycle</td>
<td></td>
<td>-1600°C</td>
</tr>
<tr>
<td>P.F.B.C.</td>
<td></td>
<td>1500</td>
</tr>
<tr>
<td>Modern Conventional</td>
<td></td>
<td>1400</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1300</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1200</td>
</tr>
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<td></td>
<td></td>
<td>1100</td>
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<tr>
<td></td>
<td></td>
<td>1000</td>
</tr>
<tr>
<td></td>
<td></td>
<td>900</td>
</tr>
</tbody>
</table>

- 35 37 39 41 43 45

- Design
- with flue-gas scrubbing and re-heat to 120°C

Figure 1.3 Overall Efficiencies of UK Coal-Fired Power-Plant (Davidson & Moore, 1979)
the gas turbine, and it is one method for doing this which is investigated in this thesis. Davidson and Moore (1977) considered two possibilities:

(i) reduction of the gas temperature to 120°C, sulphur removal using the Selexol process, followed by particulate scrubbing and gas reheat;

(ii) cleaning of the hot gas without temperature reduction, combined with the use of dolomite to retain the sulphur in the gasifier.

There are obvious thermodynamic advantages in hot gas cleaning, as illustrated in Figure 1.3. These advantages are discussed by Robson (1981), who considered hot gas cleaning using a panel bed filter (see Section 1.3.4.2) and Reed (1983), who considered hot gas cleaning using ceramic candle filters (see Section 1.3.4.1). In both studies, the economics of the hot gas cleaning process were compared with those of a process in which the gas was first cooled, then cleaned using conventional water scrubbers, and subsequently re-heated. Thermal cycle efficiencies for the cases considered by Robson (1981) and Reed (1983) are presented in Table 1.1. In each case the study was carried out for a 1200 MW power station and gas conditions of 15.5 bar g and 1000°C and showed not only that the overall operating cost of the hot gas cleaning systems considered is less than that of the cold system, but that the capital costs are comparable, primarily because of the extra cost of the re-heat exchangers in the cold case. The temperature of the gas which is re-heated after cold cleaning is limited to about 400°C, which accounts for the inferior thermal efficiency of the cold system.
Table 1.1 Thermal Cycle Efficiencies for Gasification

Combined-Cycle Plant (Robson, 1981; Reed, 1983)

Basis: 1200 MW plant.

Turbine entry conditions 15.5 bar g & 1000°C

<table>
<thead>
<tr>
<th>Methods of Gas Cleaning</th>
<th>Cycle Efficiency %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cold (wet scrubber)</td>
<td>40.7</td>
</tr>
<tr>
<td>Hot (panel bed)</td>
<td>42.7</td>
</tr>
<tr>
<td>Hot (ceramic candle)</td>
<td>44.0 - 44.4</td>
</tr>
</tbody>
</table>

In summary, therefore, gasification combined-cycle represents the current most attractive proposition for power generation from coal, and gas cleaning, preferably at turbine inlet temperatures, represents the single greatest obstacle to its development.

1.2 Hot Gas Cleaning Requirements

On the basis of National Coal Board experience, Reed (1983) gives the particle loading leaving the gasifier as about $400 \ g/\ m^3$ (18% wt. < 10 μm). The degree of certainty with which gas turbine manufacturers have been able to define tolerable particulate loadings and size distributions is limited by the modest amount of experience with coal-fired turbines. In addition, tolerance standards will alter as turbine design improves and as entry temperatures increase. Reed (1983) gives a tolerable entry loading of $4 \ mg/m^2$ with 99% < 6 μm and none > 10 μm. Pillai (1983) suggests that environmental requirements impose an even tighter restraint on particulate levels, effectively limiting emissions to sub-micron levels.
The work which follows is concerned only with the removal of devolatilised carbon char, "gasifier fines", from the off-gases of coal gasifiers. It is worth noting, however, that other processes besides coal gasification and combustion for power generation may require hot gas cleaning. These include industrial coal gasification for Substitute Natural Gas production, cement manufacture (Kerton, 1983) and capture of aerosols ejected from nuclear incidents.

1.3 Methods for Hot Gas Cleaning

Four main classes of gas cleaning device have been considered for use at elevated temperature:

(i) inertial separators;
(ii) electrostatic precipitators;
(iii) wet scrubbers;
(iv) filters.

The principles and limitations of these devices are considered briefly below.

1.3.1 Inertial Separators

Inertial separation relies on the difference in density which usually exists between the particle and the fluid. The most successful and widely-used inertial separator is the cyclone, in which a centrifugal motion is induced in the fluid and the particles migrate to the wall,
where they are collected. Since inertial separation requires the particles to move relative to the fluid the drag force is of prime interest. The aerosol particle Reynolds number, $Re_a$, is defined as:

$$Re_a = \frac{\rho_g u d_a}{\mu} \quad (1.1)$$

where $d_a$ is the aerosol particle diameter, $\rho_g$ and $\mu$ are the gas density and viscosity, and $u$ is the velocity of the particle relative to the gas. In most gas cleaning devices, $Re_a$ is small, and the drag force, $F_d$, can then be estimated from Stokes' law:

$$F_d = 3\pi \mu C u d_a \quad (1.2)$$

where $C$ is the "Cunningham Slip Correction Factor", which accounts for slip effects when the particle diameter is comparable with the mean free path of the gas molecules, $\lambda$. $C$ can be estimated from Davies' (1945) modification of a form proposed by Knudsen and Weber (1911):

$$C = 1 + Kn [2.514 + 0.8 \exp (-0.55/Kn)] \quad (1.3)$$

where $Kn$ is the Knudsen number:

$$Kn = \frac{\lambda}{d_a} \quad (1.4)$$

The mean free path, $\lambda$, can be obtained from an expression due to Beard (1976):

$$\lambda = 3200 \mu \left[ \frac{\pi}{2 \rho_g \mu^2} \right]^{1/2} \quad (1.5)$$

where $\lambda$ is in $\mu$m, $\mu$ in Ns m$^{-2}$ and $P$ in bars.
For a particle settling freely under its own weight in a gas, the drag force counterbalances the immersed weight of the particle. For low values of \( \text{Re}_a \), therefore, the particle terminal velocity \( u_t \) is given by:

\[
3\pi \mu C u_t d_a = \frac{\pi d_a^3}{6} (\rho_a - \rho_g) g
\]  

(1.6)

Normally, \( \rho_a >> \rho_g \) so that equation (1.6) can be written:

\[
\frac{\rho_a \text{d}a^2}{u_t} = \frac{18}{\mu}
\]  

(1.7)

Hence, in the absence of slip effects, \( u_t \propto \mu^{-1} \). From elementary kinetic theory (Hinds, 1982), gas viscosity is approximately proportional to the square root of absolute temperature, and independent of pressure. Hence, \( u_t \propto 1/\sqrt{T} \) and increase in temperature increases fluid-particle drag, reduces settling velocity and makes fluid-particle separation more difficult.

Since the pioneering design and test work of Stairmand (1951), many cyclone designs have been proposed, including some with electrostatic enhancement (Saxena et al, 1986). In practice none has consistently achieved a significantly better separation performance than Stairmand’s high efficiency design (Figure 1.4). A recent development has been the so-called Cardiff enhanced cyclone (Biffin et al, 1984; Syred et al, 1986) developed at the University of Cardiff. This incorporates additional cylindrical collection pockets attached to the circumference of the main cyclone and is claimed to give superior performance. However, independent performance comparison with the Stairmand high-efficiency design (Clift et al, 1984) casts doubt on this claim.
Gas cleaning devices can be crudely compared (with caution) in terms of their value of \( d_{50} \), the diameter of particle for which the efficiency of collection is 50%. Current cyclone technology cannot, in practice, achieve \( d_{50} \) values of better than 2 \( \mu m \) at ambient temperature, which is likely to rise to about 4 \( \mu m \) at 1000°C, for the reasons given above.

Wheeldon et al (1986) report \( d_{50} \) values of 3.5 \( \mu m \) and 5.1 \( \mu m \), respectively, for primary and secondary cyclones approximating to the Stairmand high-efficiency design, in tests on the Grimethorpe PPBC facility, the data being obtained at between 640 and 910°C and between 6 and 12 bar a. The high primary cyclone efficiency is attributed to the very high dust loadings (up to 140 g/actual m\(^3\)) leaving the combustor. However, other experience at the Grimethorpe PPBC plant (Snow, 1983) and at Westinghouse (Ghadiri, 1986) suggests that in practice, frequent cyclone upsets occur, arising mainly from blockages in the cyclone dip-legs, and leading to "excursions" in particle loading which would have a catastrophic effect on an operating turbine. It is clear, therefore, that cyclones alone are unlikely ever to satisfy even the gas turbine entry requirements for a combined-cycle
plant, and will certainly not meet the environmental limits. The present thinking is that a tertiary gas cleaning stage with a much higher cleaning efficiency for micron size particles is required.

1.3.2 Electrostatic Precipitators

Electrostatic precipitators are devices in which the particles to be removed are first charged by an electrical corona (generally negative) and then caused to migrate to an oppositely charged electrode, where they are collected and removed periodically by "rapping" the collection plate. For a given volumetric gas flow, electrostatic precipitators are usually more expensive than any of the other devices considered here, but they have the potential to remove particles of micron size and below. Since they depend on inducing particle movement relative to the fluid their operation is subject to the same deleterious temperature effects as were noted in the previous section. However, these may be mitigated or overshadowed by the effect of temperature and pressure on the corona spark-over voltage, i.e. the minimum potential gradient causing ionic breakdown of the gas, which generally decreases with increasing temperature, but increases with pressure (Riepe and Wiggers, 1986). If the potential gradient can be increased beyond that possible under ambient conditions then this may compensate for the increase in fluid-particle drag.

A more serious objection to the use of electrostatic precipitators, which has limited their use at elevated temperatures, is the difficulty in finding insulating materials which will retain their properties as the temperature is increased (Mayer-Schwinning, 1984). This fundamental physical problem (Chirlian, 1971) may set an absolute temperature limit to the use of precipitators. Operation on gasifier
gases is, at yet, unproven and it is possible that additional problems will be caused by the inability of the relatively cohesionless gasifier char to form a stable cake on the collector plates.

1.3.3 Wet Scrubbers

A wet scrubber is a device in which particles are removed from a gas by contact with a liquid. The scrubbing liquid must be fluid at the process conditions but must not react chemically with the gas or the containment material and must not be highly volatile. Laboratory scale tests have been carried out at the University of Essen (Weber, 1977; Weber et al, 1981) using molten salts and tin as the scrubbing liquids, but in general this option is the least likely to be adopted for coal-fired power-plant, due to the severe difficulties involved in handling the scrubbing liquids.

1.3.4 Filters

Filters are devices in which the gas passes through a permeable medium which collects and retains the particles. There are two basic types: membrane filters, in which the passages through the medium are comparable to the particle size and particles are collected as a "cake" on the upstream surface, and granular bed filters, in which the passages through the medium are large compared with the particle size and particles are (usually) collected in the bulk of the medium ("depth filtration"). The division between the two types is somewhat blurred as cake filtration is still possible in a granular bed filter if the conditions are favourable (see later). For both types, the efficiency of cleaning is a major consideration, and, as this depends on
particle-collector adhesion and particle-particle cohesion it is system-specific and may be very much affected by changes in temperature.

1.3.4.1 Membrane Filters

Many types of fabric are in use in ambient temperature membrane filters such as the familiar "bag filter". In a conventional bag filter, after an initial cake formation period, filtration occurs entirely on the surface and the bag is cleaned by shaking, reverse flow or pressure pulsing when the pressure drop across it rises to a certain level. For high temperature use, the main problem is to find a material which will withstand the mechanical (and possibly thermal) stresses due to cleaning (Chang et al, 1986). Those which have been used or proposed include sintered porous metal or ceramics, and woven, felted or compressed metal or ceramic fibres (Schulz and Weber, 1986). All are capable of achieving very high particle removal efficiency but their mechanical durability remains largely unproven, particularly in the case of metal and ceramic-fibre filters (Ciliberti and Lippert, 1984; Weber and Schulz, 1984). The most promising type appears to be the rigid sintered ceramic candle investigated by Westinghouse (Ciliberti and Lippert, 1984; Lippert et al, 1986), the University of Essen (Weber and Schulz, 1984), and the Coal Research Establishment of British Coal (Reed, 1985). The tests carried out at the Coal Research Establishment were on gasifier gases and are therefore particularly relevant to this study. In tests involving a composite candle of silicon carbide grains at 950-970°C and atmospheric pressure, a steady pressure-drop of 7.5 kPa at 50 mm/s face velocity was achieved with no loss of mechanical strength detected over 220 hours. The candle was cleaned on-line by a reverse pulse of nitrogen at 4 to 10 bar whenever
the pressure drop rose 1-2.5 kPa above the residual value for the cleaned medium. A recent development which has appeared very attractive in initial tests on both combustion and gasification dusts is the ceramic cross-flow filter (Ciliberti and Lippert, 1986). In this type of absolute filter, filtration occurs at the walls of channels within a ceramic monolith, rather similar to a ceramic heat exchanger. The filter is cleaned by reverse pulse. A major advantage of this device is said to be its very low pressure drop; typically 1.0 kPa at a filtration velocity of 33 mm/s.

1.3.4.2 Granular Bed Filters

As noted earlier, granular bed filtration differs from membrane filtration in that the passages through the medium are large compared with the dimensions of the particles to be filtered. Granular bed filtration is therefore not "absolute" in the sense that membrane filtration can be, and cleaning efficiency will often be a strong function of aerosol size, which is not usually the case with membrane filtration.

Granular filters consist of beds of unbonded filter elements, which are typically particles several hundred microns in size of some durable material such as quartz sand. They fall broadly into three categories:

(i) **Fixed Beds.** The medium remains stationary and filtration occurs in the bulk of the bed ("depth filtration") and possibly, after some time, on the upstream surface ("cake filtration"). The filter may be used once, to saturation, or cleaned in situ by back pulsing the gas or fluidising the whole medium.
(ii) Moving Beds. The medium flows, either continuously or intermittently, and usually under the influence of gravity, filtration occurring in the bulk. The filter can be cleaned by removing all or just the surface layer of medium from the filtration zone and regenerating it elsewhere by fluidisation, pneumatic conveying or combustion.

(iii) Fluidised Beds. The medium is continuously fluidised so that although most filtration occurs close to the distributor, collector particle circulation is rapid and all the medium is equally effective in collecting the aerosol. The dirty medium can be withdrawn and replaced continuously and dumped, re-generated, or used for other purposes. The possibility exists to use this type of bed as a filter/agglomerator, in which aerosol agglomerates grow on seed particles which are added to the bed or formed by breakage of existing agglomerates.

In principle, granular bed filters present considerable economic advantages over the other types of gas-cleaning devices listed here, as demonstrated for fluidised beds at ambient temperature by Doganoglu et al (1978). This is primarily because the face velocity can be much higher than for electrostatic precipitators, wet scrubbers and membrane filters, so that the volume of the equipment is much less. Indeed, if the particles to be collected are above the most penetrating size (see Chapter 2) it is advantageous for the face velocity to be high because this improves collection by inertial impaction.

It is necessary to distinguish between three primary processes occurring in granular bed filtration:
(i) collection of dust particles by the collector particles;
(ii) retention of captured dust by the collector particles;
(iii) modification of the filter structure due to the build-up of collected dust (with its effect on the subsequent filtration efficiency).

In "stationary filtration", collected particles either rebound instantaneously or are permanently retained, and collection occurs on the surface of the collector particles and not on the new structure formed by the collected aerosol. Filtration efficiency therefore remains constant with time. The initial stages of filtration in fixed beds, and filtration in moving and fluidised beds at low aerosol concentration, can be considered stationary. If the filter performance changes with time, it is said to be "non-stationary". This is the case in cake filtration, where a cake forms on the upstream face of the filter, and the filter efficiency and pressure drop increase. Non-stationary filtration could also occur in fluidised bed filters, if agglomerates of collected aerosols were detached from the collector granules and entrained from the bed.

Table 1.2 lists the types of granular bed filter which have been tested at high temperatures and Figures 1.5 to 1.10 illustrate examples of the types.
Table 1.2 Types of Granular Bed Filter Which Have Been Tested at High Temperature.

<table>
<thead>
<tr>
<th>Fig. No.</th>
<th>Name</th>
<th>Type</th>
<th>Medium</th>
<th>How Cleaned</th>
<th>Reference</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.6</td>
<td>Panel bed</td>
<td>Fixed</td>
<td>Sand</td>
<td>Intermittent reverse pulse removes upstream layer for disposal or regeneration elsewhere</td>
<td>Arnold (1981)</td>
<td>Cake formation essential. Little work at high temperature. Problems with re-entrainment.</td>
</tr>
<tr>
<td>1.7</td>
<td>Moving bed</td>
<td>Moving</td>
<td>Sand</td>
<td>Continuous removal for disposal or regeneration, usually pneumatically</td>
<td>Moreseco and Cooper (1981)</td>
<td>Frequently electrostatically-enhanced. No cake formation. A number of units in industrial use in USA, not all successfully.</td>
</tr>
<tr>
<td>1.9</td>
<td>Dry plate scrubber</td>
<td>Moving</td>
<td>Zirconia</td>
<td>Continuous removal for regeneration elsewhere</td>
<td>Calvert et al (1977)</td>
<td>Similar to fluidised beds in theory and performance.</td>
</tr>
</tbody>
</table>
In the "Ducon" filter (Figure 1.5), dusty gas passes through vertical screens and is directed downwards through a fixed bed of collection medium. Filtration can either be in the bulk of the medium or primarily at the surface, depending on the gas velocity, and the collector and aerosol properties. The bed is regenerated periodically in situ by back-flow or fluidisation. The problems in high temperature tests have concerned blocking of the retaining screens and poor cleaning efficiency. In the "Panel Bed" (Figure 1.6), the medium is retained in a tall, narrow "panel" between two vertical walls resembling Venetian blinds. Gas passes through the solid more or less horizontally. The medium is displaced periodically by a sharp reverse pressure pulse, which removes the first few layers of filter granules from the upstream face, which are then collected for disposal or regeneration elsewhere. If the dust is relatively loosely bound on the collector, regeneration can occur simply by pneumatically conveying the medium back up to the top of the unit. In the panel bed it is essential that a cake is formed on the upstream face of the filter, and it is here that problems have arisen in its use at high temperature. Panel beds have been tested at high temperature by British Coal (Arnold, 1981; Reed, 1983). Both plant and laboratory tests have shown that gasifier fines will not form a cake, while fly ash of about the same particle size will do so. Coury (1983) attributed this to the fact that the fly ash was electrically charged and therefore cohesive, while gasifier fines are not. He also showed that fly ash cake formation will not occur if the collector particle size is too large (typically about 500 μm) or the face velocity is too high. The operation of a panel bed filter is therefore highly system-specific and filter performance using a given dust cannot be predicted a priori.
Figure 1.5 "Ducon" Fixed Bed Filter with Fluidised Expansion Cleaning
Inlet velocity 15.2 m/s, solid density 2000 kg/m³ in air
at 20°C. Cyclone diameter, D = 0.2 m.
Figure 1.6 Panel Bed Filter
Figures 1.7 a and b illustrate two types of moving bed filter, in which the collection medium is in continuous downward motion and the gas flows counter-currently or in the cross-flow direction through it. In this type of filter, as in the others considered here, it is clearly important for the collected dust to be strongly retained on the collector until regeneration occurs in the pneumatic riser, sometimes assisted by impacting the collector particles onto a target to detach the collected dust. This type of filter has been electrostatically enhanced (Saxena et al., 1986), particularly to improve the collection efficiency for submicron aerosols (for which other collection mechanisms are rather ineffective; see Chapter 2). However, electrostatic augmentation is subject to the same operating difficulties at high temperatures as those discussed earlier in connection with electrostatic precipitators.

Figure 1.8 illustrates the magnetically-stabilised expanded-bed filter developed by Exxon and arranged in a vertical cylinder in a similar way to the panel bed. The collection medium is magnetic cobalt maintained in a state of uniform voidage by an electric current passing through a solenoid wrapped round the outside, so that very high face velocities (up to 6 m/s) can be employed without displacing the medium. The use of the magnetic field also means that it is unnecessary to use a retaining screen on the downstream side of the filter, which avoids the problems of screen blockage encountered with other granular bed filters. It remains to be seen whether the advantages of the Exxon filter outweigh the cost of the special filter medium and of maintaining the magnetic field.
collected particulate

to bag filter

fluidised
bed

fluidising air

dirty gas

clean gas

annular moving bed

injector air

transport air

collector medium transport

screens

Figure 1.7a Cross-Flow Moving-Bed Filter
(Combustion Power Co.)
Figure 1.7b Screenless Counter-Current Moving-Bed Filter
(Combustion Power Co.)
Figure 1.8 Magnetically-Stabilised Granular-Bed Filter (Exxon)
The "dry plate scrubber" is shown schematically in Figure 1.9. It consists of a number of shallow beds a few centimetres deep, through which the collector particles are caused to flow continuously by gravity. Dusty gas enters the beds through 1-2 mm diameter orifices at high gas velocity (up to 40 m/s) in a similar way to the fluidised bed filter to be described later. Collection occurs mainly close to the orifices, where inertial effects are strong because of the high relative velocity between the gas and the collector particles. Zirconia beads are recommended as collectors in place of sand, because their high density enables higher gas velocities to be used. The concept and performance of the dry plate scrubber are very similar to those of the fluidised bed filter.

The fluidised bed filter is illustrated schematically in Figure 1.10. This device, which is the subject of the remainder of this thesis, represents the simplest design of moving granular bed filter and is particularly well-suited to applications in which it is desired to remove spent medium and introduce fresh medium on a continuous basis. It is usually most efficient to use a low-free-area gas distributor in which the gas enters the bed at high velocity through an array of orifices. In this case, the region immediately above each orifice experiences very much higher gas and particle velocities and a very much lower solids concentration than in the rest of the bed. The fluctuating flame-like region thus produced is sometimes known as a "jet", by analogy with a conventional turbulent jet (Abramovich, 1963), but the detailed mechanistic analysis of particle and fluid behaviour in this region is still a matter of controversy (see, for example, Clift, 1983). From the point of view of fluidised bed filtration, what is important about this gas-entry or "jet" zone is that the collector particles which are entrained into it experience a high slip velocity
relative to the gas. The efficiency of aerosol collection in this region is therefore greatly enhanced compared with its value in the bubbling bed region (because inertial collection processes are more efficient at higher velocities) and in practical filters the bed then simply represents a reservoir of collection medium and need be only a few centimetres deeper than the jet length. It is therefore necessary to consider two distinct collection regions: the entry or jet region, and the bubbling bed region. There is now a large body of experimental and theoretical work on filtration in fluidised beds, although relatively little is concerned primarily with the all-important entry region. The next chapter considers the relevant experimental work and describes how theoretical collection efficiencies can be calculated.

Although the fluidised bed filter has the advantages of ease of operation and high collection efficiency, it suffers from the same disadvantage as all other types of granular bed filter, namely, poor retention with certain types of dust. In Section 2.1.2 of the next chapter, retention is considered from a theoretical viewpoint, indicating that an improvement can be made by making the collector more adhesive, by addition of a liquid "retention aid", for example. Section 2.2 concerns the experimental work in which a retention aid was used by previous workers, at either ambient or high temperatures.
Figure 1.9 Dry-Plate Scrubber (A.P.T.)
Figure 1.10 Fluidised-Bed Filter
1.4 Other Aspects to be Examined

Particle size measurements in the sub-micron to 10 µm range and at high temperatures are notoriously difficult to make, and it would be desirable to employ sophisticated on-line techniques to measure particle size distribution and concentration. A variety of techniques has been used in previous experimental work, and it is not always apparent how the measurements of one group of workers relate to those of another group using different techniques. Chapter 3, therefore, concerns the practical problems of obtaining particle size and concentration at high temperatures. The operation of the various available instruments is reviewed and their capabilities are compared by experiment using the aerosol particles of interest here. Finally, the method chosen for this investigation is described in detail.

An unfortunate consequence of the addition of liquid retention-aid to a fluidised bed is the resulting modification of fluidisation behaviour, which may lead eventually to complete defluidisation. Since this is potentially the cause of catastrophic filter failure, it is the major objection to the use of this technique in combined-cycle power generation. Interest in the effects of increase in interparticle forces on fluidisation behaviour is not limited to filtration applications; understanding of these effects is necessary in several other applications, such as fluidised bed combustion and agglomeration. Theoretical and experimental aspects of fluidisation behaviour in the presence of interparticle forces are considered in detail in Chapters 4 and 5.
In Chapter 6, the choice of retention-aid and practical methods for introducing it to the bed at high temperature are discussed. Finally, experiments are described in which the chosen retention-aid was used to enhance high-temperature filtration performance. Results of these experiments are analysed, and a novel method of filter operation is proposed.
Chapter 2

Fluidised Bed Filtration

2.1 Filtration Efficiency

In all types of granular bed filtration, as noted in Chapter 1, it is necessary to distinguish between the collection efficiency and the retention efficiency. The net filtration efficiency is the product of the two:

\[ \eta_{\text{filtration}} = \eta_{\text{collection}} \times \eta_{\text{retention}} \]  

The factors which govern collection and retention are now considered in turn.

2.1.1 Collection

Consider a single spherical collector particle of diameter \( d_c \), in a gas stream of superficial gas velocity \( U \) as shown in Figure 2.1. The "single (collector) particle collection efficiency", \( E \), is defined as:

\[ E = \frac{\text{number of dust particles collected}}{\text{number of dust particles in approach volume}} \]  

(2.2)

\( E \) is a function of dust and collector size and electrostatic properties, dust density, gas properties and bed voidage, \( \varepsilon \).

The volume of gas swept out by the collector in unit time = \( \frac{\pi d_c^2}{4} \cdot U \)
TRAJECOTRY OF PARTICLE WITH INERTIA

APPROACH VOLUME GAS STREAMLINES

Figure 2.1 Single Particle Collection.

(some authors use $U/e$ in place of $U$, which leads to numerically different values for $E$). If the number concentration of particles in the gas approaching the collector is $C$, then the number of particles collected per second is $CUC(πd_c^2/4)$.

Now 1 m$^3$ of bed contains $6(1−e)/πd_c^3$ collector particles, so the collection rate per unit bed volume, $R_V$, is given by

$$R_V = K_vC$$

(2.3)

where $K_v = 3EU(1−e)/2d_c$ and is the collection rate constant per unit bed volume. Collection is a First Order process, i.e. the rate of filtration is proportional to the dust concentration. For non-spherical particles, particle shape will modify this result, but the effect will be slight provided that no one collector dimension is very
much greater than the others. If the collector consists of a range of particle sizes, the correct value for \( d_c \) is the surface-volume or Sauter mean.

It remains to calculate \( E \). The four purely mechanical processes which can cause collection are illustrated in Figure 2.2 and the most reliable correlations for these are listed in Table 2.1. All of the expressions given have been extensively tested against experimental data obtained from experiments with liquid aerosols, for which complete retention can be assumed. The expression chosen for the collection efficiency due to inertial impaction deserves more comment, however, since D'Ottavio and Goren (1983) have recently proposed a more sophisticated alternative incorporating a dependence on collector particle Reynolds number \( (U_d \rho_d/\mu) \). Coury et al (1987), reviewing all the available data, conclude that there is little justification for the inclusion of a Reynolds number dependence and that, within the range of interest, the D'Ottavio and Goren correlation predicts very similar results to those obtained from the earlier Thambimuthu (1980) correlation. Since the Thambimuthu correlation has the additional merit of being simpler, it is used here in preference to the rather complex D'Ottavio and Goren formulation.

It is normally sufficiently accurate to obtain \( E \) by addition of the individual terms, provided that all terms are small or one dominates (Dorman, 1960). Figure 2.3 shows individual and overall single particle collection efficiencies for typical values of bed and aerosol properties at ambient temperature plotted as functions of aerosol size.
Figure 2.2 Mechanical Mechanisms of Collection.
Table 2.1

Efficiency of Dust Capture in Granular Bed Filters by Mechanical Processes

<table>
<thead>
<tr>
<th>Mechanism</th>
<th>Collection Efficiency, E</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>a. Diffusional collection</td>
<td>$4.36 \frac{D^{2/3}}{e} \left[\frac{D}{Ud_a}\right]^{2/3}$</td>
<td>Wilson and Geankoplis (1966)</td>
</tr>
</tbody>
</table>

where

- $U =$ superficial gas velocity through filter (m/s)
- $e =$ void fraction in filter (-)
- $D =$ $\frac{kg \cdot C}{3 \pi \mu d_a}$, particle diffusivity (m$^2$/s)
- $K_B =$ Boltzmann's constant (J K$^{-1}$)
- $T =$ absolute temperature (K)
- $\mu =$ gas viscosity (kg/ms)
- $C =$ Cunningham slip correction factor (-)
- $C = 1 + Kn \left[2.514 + 0.8 \exp (-0.55/Kn)\right]$ for air
- $Kn =$ Knudsen number, $\lambda/d_a$ (-)
- $\lambda =$ mean free path (m)
- $\rho =$ pressure (bars)
- $P =$ pressure (bars)

b. Inertial impaction

$$\left[\frac{St}{St - 0.062e}\right]^3$$

Thambimuthu (1980)

where

$$St = C \frac{\rho_a d_a^2 U}{9 \mu d_C}$$

c. Direct interception

$$6.36e^{-2.4} \left[\frac{d_a}{d_c}\right]^2$$

Paretzky (1972)

d. Gravitational settling:

- Upflow: $0.0375(ut_a / U)^{0.5}$
- Downflow: $0.0375(ut_a / U)^{0.5} + (ut_a / U)^{0.78}$

where $ut_a$ is the terminal velocity of the aerosol particle.
Figure 2.3 Typical Single Particle Collection Efficiencies.
Figure 2.4 Effect of Temperature on Overall Single Particle Collection Efficiency.

\[ \begin{align*}
U &= 0.08 \text{ m/s} \\
\text{d}_c &= 4.61 \mu \text{m} \\
\varepsilon &= 0.41 \\
\rho_a &= 1800 \text{ kg/m}^3 \\
nitrogen \quad 1 \text{ bar a.}
\end{align*} \]
For any filter operating conditions, there is a "most penetrating particle size" at which the collection efficiency is a minimum. For aerosol particles below this size, diffusional collection dominates, while, for larger aerosols, inertial impaction usually dominates. The Brownian diffusivity, $D$, can be evaluated by means of the Stokes-Einstein equation (Clift et al, 1981):

$$D = \frac{Ck_BT}{3\pi\mu d_c} \quad (2.4)$$

where $k_B$ is Boltzmann's constant, $1.38 \times 10^{-23}$ JK$^{-1}$.

From Section 1.3.1, gas viscosity is approximately proportional to the square root of absolute temperature, $T$. Hence,

$$D \propto \sqrt{T} \quad (2.5)$$

so that $D$, and the single particle collection efficiency due to diffusion (Table 2.1 a) both increase with increase in temperature. The influence of pressure arises only through the Cunningham slip correction factor, $C$, and is much weaker.

Increase in temperature reduces the collection of aerosols due to inertial impaction (Table 2.1 b) because of the increase in gas viscosity. The combined effect is to increase the most penetrating particle size at higher temperatures, as shown in Figure 2.4. In addition to these purely mechanical processes, electrophoretic collection may be significant in some circumstances. The main electrical collection mechanisms are reviewed by Ranz and Wong (1952)
and Clift (1983), and consist of:

(i) Coloumbic attraction between charged aerosol and oppositely charged collector;
(ii) attraction between charged aerosol and dipole induced in the collector;
(iii) attraction between charged collector and dipole induced in the aerosol particle;
(iv) space-charge repulsion, i.e. repulsion of aerosol particles of like charge;
(v) attraction between charged aerosol and earthed collector carrying opposite charge induced by space-charge effects from surrounding aerosols.

Of these, (i) and (ii) have been used to enhance filter efficiency. (i) effectively uses the bed as the collection region of an electrostatic precipitator, but this is costly and difficult to achieve. (ii), the charged-induced dipole mechanism, may occur in any case if the aerosol to be collected is "naturally" charged. Whether this is the case in many industrially operational granular bed filters remains unknown because of the difficulty in measuring charge distributions of aerosols in situ. The single particle collection efficiency for this mechanism is given approximately (Coury, 1983) by:

\[
E = 9 \left[ \frac{C \gamma_c d_a^2}{3 \pi^2 \epsilon_0 \mu d_d d_c^2} \right]^{0.5}
\]  

(2.6)
where \( q_a \) is the charge on the aerosol particle (C)

\( C \) is the Cunningham slip correction factor (-)

\( \varepsilon_0 \) is the permittivity of free space \( (C^2 s^{-2} m^{-1}) \)

\( d_a \) is the diameter of the aerosol particle (m)

\( \gamma_c \) is the collector polarisation coefficient, (-)

\[ \gamma_c = \frac{(\varepsilon_c - \varepsilon_g) / (\varepsilon_c + 2\varepsilon_g)}{\varepsilon_0} \]

where \( \varepsilon_c \) is the dielectric constant of the collector and \( \varepsilon_g \) is the dielectric constant of the gas. In most practical cases, \( \varepsilon_c > \varepsilon_g \) so that \( \gamma_c \approx 1 \). For charge levels typical of corona discharge, electrophoretic collection dominates for \( d_a \) around the most penetrating particle size.

Court (1983) showed that the electrostatic charge level on gasifier fines dispersed as in this work was very low, which is not surprising considering that the dispersion method (see Chapter 3) is relatively "gentle" and that the aerosol particles are highly conducting. Electrophoretic collection has therefore been ignored in this work.

2.1.2. Retention

In order to remove an aerosol particle from a gas stream it is insufficient for it merely to be collected (by the mechanisms listed above); it must also be retained, i.e. it must not rebound or be subsequently dislodged. There is ample experimental evidence (see, for example, D'Ottavio and Goren, 1983) that poor retention limits the filtration efficiency of granular filters for larger particles and at higher gas velocities, i.e. in the region in which inertial collection is most effective. For a particle which adheres on impact, the adhesion force is normally sufficiently strong for fluid drag alone not
to cause re-entrainment, as shown for fibrous filters by Stenhouse and Freshwater (1976), so that the problem is generally one of instantaneous rebound. However, in fluidised and moving bed filters subsequent attrition of collected particles may also occur.

A theory of rebound has been developed by Coury (1983) and Clift (1983), from the work of Dahneke (1971) and others. If the kinetic energy of an aerosol particle approaching the collector is \( E_i \) and the "depth of the potential well" (Dahneke, 1971) due to the short range van der Waals forces is \( E_v \), then the aerosol particle energy at contact (approaching) is \( E_i + E_v \) and at contact (receding) is \( e^2(E_i + E_v) \) where \( e \) is the coefficient of restitution for the contact. For rebound to occur

\[
e^2(E_i + E_v) > E_d
\]  

(2.7)

where \( E_d \) is the new depth of the potential well from which the particle must climb in order to escape, i.e. the remaining energy must exceed the detachment energy. \( E_d \) is the sum of the van der Waals interaction energy and any terms due to electrostatic attraction, or, say, liquid bridges. Since \( E_i \) is equal to \( 1/2(mv^2) \) where \( m \) is the aerosol particle mass, inequality (2.7) can be rewritten as

\[
d_a \frac{3v^2}{2} > \frac{12}{\pi} \frac{d_a}{
e^2} \left[ E_d - E_v \right]
\]  

(2.8)

This inequality suggests that for a given particle size there is a critical impact velocity above which rebound will occur. In general, however, \( E_d, E_v \) and \( e \) are not known with accuracy, so that for a real filter there is no substitute for experiment. In summary, although collection efficiency due to inertial impaction increases with increase
in face velocity (for aerosol particles above the most penetrating size), the probability of rebound also increases, and this may eventually dominate. Rebound will be substantially reduced if either the aerosol or the collector is soft under process conditions, or if the collector can be covered with a layer of sticky "retention aid". Both of these have the effect of increasing $F_d$ and so making rebound less likely.

2.1.3 Prediction of Filtration Efficiency

When a gas enters a fluidised bed through an array of orifices in a perforated plate, there exists a region of intense gas/solid contacting immediately above the distributor, as described in Chapter 1. It is well-known (see, for example, Massimilla, 1985) that in this entry region reaction (if present), and heat and mass transfer can be much more intense than elsewhere in the bed. It is not surprising, therefore, that those workers who have used a perforated plate distributor for fluidised bed filtration should also have observed anomalous entry region effects. Figure 2.5, for example, shows some of the penetration data of Tan (1982), plotted on a logarithmic scale against $H_{mf}$, the bed height at minimum fluidisation. Analysis of collection in the bubbling bed (Appendix A.1) predicts that the penetration, $f_f$, will vary exponentially with bed height, and this is observed, but in shallow beds it is usual to observe an abrupt change in slope or a discontinuity in the data. Penetration is very high for beds shallower than this critical depth, $h_j$. Tan (1982) attributed the discontinuity to the jets breaking through the surface of the bed. This interpretation was consistent with visual observations and he found that the bed depth at which the discontinuity occurred correlated well with independent estimates of jet penetration length (see below).
Figure 2.5 Variation of Penetration with Bed Height
(Tan, 1982; capture of di-ethyl hexyl sebacate aerosol in beds of glass ballotini)
It is clear from Figure 2.5 that, for this type of low free-area distributor, most collection occurs in the jet region, and there is little to be gained by using a deep bed. In a well-designed filter, the bed depth need only be a few centimetres deeper than the jet length; the bed then simply represents a reservoir of collection medium for entrainment into the jet.

The question now arises as to how to treat this region of enhanced collection in calculations of overall collection efficiency. The method proposed by Clift et al (1981), and followed here, is to treat the bubbling bed and the jet region separately. Thus the net filtration efficiency, $\eta$, is given by:

$$1 - \eta = (1-\eta_j)(1-\eta_b)$$

where $\eta_j$ and $\eta_b$ are the filtration efficiencies attributed to the jet region and the bubbling bed respectively. In practice there is always a small contribution to the overall efficiency from the distributor itself. Ghadiri (1980) showed that, for a distributor of the type used here, this contribution is usually small enough to be ignored.

An alternative model for fluidised bed filtration based on the "bubble assembly" approach was developed by Peters et al (1981, 1982) and later by Ishiki and Tien (1984). This model considers three phases (the bubble, the "cloud", and the emulsion or particulate phase). It has been shown that the model predictions can be made to agree with a limited amount of the data in the literature. However, the model is open to a number of serious objections:
(i) its reliance on the existence of "clouds" under conditions where they have been shown not to exist as distinct regions (Clift and Grace, 1985),

(ii) its inability to allow for anomalous collection close to the distributor,

(iii) the fact that it predicts a fall in collection efficiency with increasing gas velocity, in contradiction to much of the available data, and

(iv) that the model relies heavily on prediction of interphase transfer rates under conditions where Tan (1982), for example, has shown that collection is not limited by interphase transfer.

For all these reasons, Peters' model is not considered further.

The next two sections concern the prediction of the separate collection efficiencies of the jet region and the bubbling bed.

2.1.3.1 Collection in the Jet Region.

The precise nature of gas and solids motion in the jet region is still a matter of academic debate. Rowe et al (1979) used X-rays to observe gas discharge from orifices into a fluidised bed, and suggested that a distinction should be made between "jetting", meaning the establishment of a more-or-less permanent cavity above each orifice, and the bubble-chain mode of gas entry, in which volumes of gas form and detach periodically from each orifice, which is covered by fluidised particles between successive events. They also suggested that jetting was the less-common form of gas entry, and occurred only when the bed was inadequately fluidised or when surfaces were present to hinder the
flow of particles towards the orifice. However, it is also known that jetting is more common with larger and less-dense particles, and that jet steadiness depends on gas density, density ratio and orifice gas velocity (Massimilla, 1985). Clift (1983) has suggested that, for many purposes, the distinction between modes of gas entry may be superfluous; since Rowe et al. (1979) observed a bubble frequency of about 8 Hz, the distinction may only be apparent for processes having a time constant of less than about 0.1 s. In this work, the same philosophy is followed and the "jet" region is taken as synonymous with the "entry" region, whether or not permanent voids exist within it.

Mechanistic models of the jet region are summarised by Massimilla (1985). At present, only one attempt has been made to predict the collection efficiency of the jet from the calculated single-particle collection efficiency. This is the model of Tan (1982) and Tan and Seville (1982), based on the axi-symmetric turbulent jet model of Donsi et al. (1980). This shows qualitative agreement with experimental data on the variation of penetration with aerosol and collector size and with orifice gas velocity, but needs further development before being suitable for quantitative use. It is therefore necessary to fall back on empirical correlations derived from experiments at ambient temperature. Thambimuthu (1980) correlated his own data and that of Doganoglu (1975) and Knettig and Beeckmans (1974) in terms of a modified Stokes number:

\[ N_{or} = St_{or} \left( \frac{N_{uc}}{U_{or}} \right)^{1/3} \quad (2.10) \]

where \( St_{or} \) is the Stokes number based on the superficial velocity of gas entering the bed through the distributor orifices, \( U_{or} \):
\[ St_{or} = \frac{C_p d^2 u_{or}}{a^4 u_{or}^{3/2}} \]  

(2.11)

and \( N_{uc}^{1/3} \) is a dimensionless terminal velocity for the collector particles.

\[ \frac{1}{3} \left( \frac{3 \rho_d}{4(\rho_c - \rho_d)} \right)^{1/3} u_{tc} \]  

(2.12)

where the collector particles have density \( \rho_c \) and terminal velocity \( u_{tc} \). The term \( N_{uc}^{1/3} \) is intended to take account of the variation in entrainment rate of collector particles into the jet. The penetration of dust particles through the jet region, \( f_j \), is then correlated (Thambimuthu, 1980; Clift et al, 1981) by:

\[ N_{or} < 0.4 : f_j = 1 - 3.1 N_{or}^3 \]  

(2.13)

\[ 0.4 < N_{or} < 0.8 : f_j = 1.749 - 2.62 N_{or} + 0.345 N_{or}^2 + 0.715 N_{or}^3 \]  

(2.14)

\[ 0.8 < N_{or} : f_j = 1 - \left( \frac{N_{or}}{N_{or} + 0.078} \right)^3 \]  

(2.15)

Figure 2.6 shows the available data on jet collection in beds of rounded collector, with equations (2.13)-(2.15) plotted for comparison. Thambimuthu's (1980) correlation refers only to data obtained with beds of glass ballotini. For beds of angular collector, such as sand, Tan
Figure 2.6 Empirical Correlation of Jet Collection in Beds of Spherical Collector (after Clift et al.; 1981)

<table>
<thead>
<tr>
<th>Reference</th>
<th>Symbol</th>
<th>( d_c ) (( \mu m ))</th>
<th>( d_{or} ) (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thambimuthu (1980)</td>
<td>□</td>
<td>532</td>
<td>1.2</td>
</tr>
<tr>
<td>Doganoglu (1975)</td>
<td>○</td>
<td>108</td>
<td>1.22</td>
</tr>
<tr>
<td>Knetting and Beeckmans (1974)</td>
<td>△</td>
<td>425</td>
<td>0.79</td>
</tr>
</tbody>
</table>
Figure 2.7  Jet Collection of Di-Ethyl Hexyl Sebacate Aerosols in Beds of Glass Ballotini and Silica Sand (after Tan, 1982)

\[ d_C = 461 \, \mu m \quad ; \quad d_{Or} = 1.5 \, mm. \]
(1980) and Ghadiri (1980) showed that the jet collection was considerably reduced, as illustrated in Figure 2.7. The reason for this is that the rate of entrainment of angular particles into the jet is less than for rounded particles, which is reflected in a lower time-averaged collector particle density in the jet (Seville et al., 1987). This shape effect is clearly not accounted for in Thambimuthu's correlation. Any change which reduces the rate of entrainment of collector particles into the jet, such as increased interparticle forces due to liquid bridges, for example, will decrease the collection in the jet region (Tan and Seville, 1982). Ghadiri (1980) correlated Tan's (1980) results for jet collection in beds of sand as follows:

\[ f_j = \begin{cases} \frac{3.163}{N_{or}} & \text{if } N_{or} > 3.163 \\ 1 & \text{if } N_{or} \leq 3.163 \end{cases} \]

Equations (2.16) and (2.17) were used by Ghadiri (1980) and found to be in good agreement with the data he obtained on the overall collection efficiency of boric oxide aerosols at 800°C. (Boric oxide is a liquid at that temperature so that retention would have been complete). Thus, equations (2.16) and (2.17) have been used to predict collection efficiencies in the jet region throughout this work.

There are many correlations and several different definitions for jet penetration length (Massimilla, 1985), but the one which has been found most reliable in work with small beds and simple low-free-area perforated-plate distributors (Clift, 1986) is that due to Merry (1975):

\[ \frac{L}{d_{or}} = 5.2 \left[ \frac{\rho_l d_{or}}{\rho_c d_c} \right]^{0.3} \left[ 1.9 \left( \frac{U_{or}}{g d_{or}} \right)^{0.2} - 1 \right] \]

(2.18)
where \( L \) is the jet length, \( d_\text{or} \) is the diameter of the orifice and \( U_\text{or} \) is the superficial velocity at the orifice. Tan (1982) found good agreement between \( L \) and \( h_j \) estimated from plots of penetration data such as Figure 2.5, and Ghadiri and Clift (1980) showed that \( L \) corresponded to the length of erosion marks formed on the walls of the same bed which was used for the present work, using a similar distributor at 800°C. \( \gamma \)-ray tomographic measurements using the same distributor as used in this work (Seville et al., 1986), but at ambient temperature, again suggested that Merry's (1975) correlation was the most applicable of those available.

Nienow and Killick (1983) measured the collection of potassium permanganate aerosols of count mean diameter 0.7 \( \mu \)m in shallow fluidised beds of bronze shot. An analysis of their results shows that the orifice number, \( N_{\text{or}} \), would have been above 10 in most of their experiments, so that the jet penetration, \( f_j \), should have been less than 3% in all cases, if Thambimuthu's (1980) correlation is applicable. However, the measured jet penetration values vary from 33 to 74%. There are three possible reasons for the discrepancy:

(i) Firstly since a solid aerosol was used, and impaction velocities were high, it is possible that the results were influenced by rebound or "bounce" of the aerosol particles after impact on the collector, as discussed in Section 2.1.2. This is considered unlikely, however, since water was sprayed into the bed to eliminate static charges. In separate experiments, Nienow and Killick showed that the filtration efficiency increased slightly as the rate of water addition was increased, before levelling off at a constant value. They also showed that efficiency increased with increase in gas velocity. Both of these experiments suggest that rebound is the least likely explanation for their relatively poor collection efficiencies.

See addendum.
(ii) The mode of gas entry may have been different from that in experiments with glass ballotini, i.e. bubbling, rather than jetting. Massimilla (1985) remarks that jet formation is more likely in beds of particles of lower density, e.g. sand or glass ballotini.

(iii) The mode of gas entry may have been jetting but the rate of entrainment into the jet may have been affected by factors not accounted for in Thambimuthu's (1980) correlation.

Nienow and Killick were able to estimate values for $h_j$ from plots similar to Figure 2.5. These values are much higher than the values of $L$ predicted from Merry's (1975) equation but they are, in all cases, approximately equal to $2.5d_{Bo}$, where $d_{Bo}$ is the initial diameter of a bubble formed at the orifice in the bubbling mode of gas entry:

$$d_{Bo} = \left( \frac{n}{b} \cdot \frac{Q_B}{bn} \right)^{1/3}$$

where $Q_B$ is the volumetric bubble gas flowrate at the orifice (assumed to be equal to the total gas flowrate, $Q$, minus that necessary for fluidisation, $Q_{mf}$), $b$ is the bubble frequency (8 Hz; Rowe et al, 1979), and $n$ is the number of distributor orifices. This Nienow and Killick offer as evidence of a bubbling mode of gas entry, since they observe equal values of $h_j$ for distributors with equal values of $n$. Alternatively, higher than expected values of both $h_j$ and $f_j$ would occur if the rate of entrainment of collector particles into the jet were restricted in some way, which may have occurred in these experiments since water was sprayed into the bed and interparticle forces due to the formation of liquid bridges may have been
significant. However, the fact that Nienow and Killick observed an increase in the filtration efficiency with increase in rate of water addition up to wet quenching makes this possibility also unlikely.

Comparatively little data exists on filtration efficiencies with low free-area distributors using dense particles. Jog (1975), subsequently reported in Doganoglu et al (1978), obtained very high collection efficiencies for 1.35 μm dioctyl phthalate aerosols in a bed of 142-274 μm copper shot at gas velocities in excess of 6Umf, but his data are insufficient to enable the estimation of fj for comparison with Nienow and Killick's values. The most likely explanation for their comparatively low efficiencies would therefore seem to be that their experiments were carried out in a bubbling rather than a jetting regime, and that their results reflect a genuine difference in contacting mechanism between the two types of flow pattern. If this is the case, then it is clear that it is advantageous to operate a fluidised bed filter in the jetting regime.

In summary, the jet collection correlation of Thambimuthu (1980) appears to be reliable for glass ballotini but does not account for changes in particle shape or, possibly, density. The experimental conditions of Ghadiri's (1980) experiments are closest to those for the work reported here; in Ghadiri's (1980) work, good agreement was found with equation (2.18) for the jet region height and equations (2.16) and (2.17) for jet penetration.
2.1.3.2 Collection in the Bubbling Bed

In the bubbling bed, collection only occurs in the particulate dense phase between the voids or "bubbles" which characterise gas-fluidised beds. Therefore it is necessary to account both for transfer of dust from bubbles to particulate phase and for collection within the particulate phase. However, interphase transfer is normally much more rapid than collection, so that bubble and interstitial gas can be assumed to carry the same dust concentration. The collection efficiency of the bubbling bed, is then approximated (Clift et al, 1981) by:

\[ \eta = 1 - e^{-k} \] (2.20)

for plug flow in the particulate phase, and

\[ \eta = 1 - \frac{1}{1+K} \] (2.21)

for fully mixed particulate phase,

where \( k = 3E (1 - \epsilon_d)(H - h_j)/2d_c \), \( \epsilon_d \) is the void fraction in the particulate phase of the bed, \( H \) is the overall depth of the fluidised bed, and \( h_j \) the length of the gas entry jets (if present). Shallow fluidised beds at high velocities normally operate closer to complete mixing (van Deemter, 1985) so that equation (2.21) is more generally applicable. The single granule collection efficiency, \( E \), is evaluated at mean interstitial conditions. Clift et al (1984) showed that it can be evaluated from the correlations summarised in Section 2.1.1 assuming an effective superficial velocity:

\[ U = U_{mf} \left[ 1 + 1.5 \epsilon_B^{2/3} \right] \] (2.22)

where \( \epsilon_B \) is the fraction of bed occupied by the bubble phase.
Equation (2.22) accounts for the increase in interstitial velocity due to gas passing between bubbles.

Equations (2.20) and (2.21) are approximations for the case of infinitely rapid interphase transfer. The rigorous theory proposed by Clift et al. (1981) for calculating collection in the bubbling bed is summarised in Appendix A1, together with the listing of a computer program used to calculate collection efficiencies for comparison with experimental data. Typical calculated efficiency results for the distributor used in this work are presented in Figure 2.8, showing the relatively small contribution made by the bubbling bed region. In the experiments reported in Chapter 6, an atomising nozzle was used to disperse a retention aid into the fluidised filter. It is possible that the atomising gas flow, by disrupting the flow in the bubbling bed, may affect (and probably reduce) collection in this region. However, the extent of any such effect is not easily quantified and its effect on the calculated overall efficiency will be small, so it has been ignored here.

![Figure 2.8 Predicted Grade Efficiencies](image)
Experimental studies of fluidised bed filtration have been recently reviewed by Clift et al. (1981) and by Nienow and Killick (1983). Such studies are numerous and consideration will be given here only to those in which a retention aid was used, at either ambient or high temperatures.

As noted earlier, it is retention rather than collection which is the limiting factor in the development of granular bed filters for filtering solid dusts. Retention might conceivably be improved by coating the collector with a liquid layer, raising the temperature of the system until either the collector or the aerosol becomes soft, or by causing species in the carrier gas to condense on the aerosol. In practice it is easier to modify the collector rather than the aerosol. The process then becomes essentially the same as fluidised bed agglomeration and the collected aerosol is agglomerated, layer-wise, onto the surface of the collector or seed particles. These principles were employed on a laboratory scale by Pilney and Erickson (1968a, 1968b), who collected fly ash with a mass median diameter of 8 μm by self-agglomeration in humid air, on particles coated with oil, and by agglomeration on wax particles, rendered sticky by increasing their temperature to 60°C. In the self-agglomeration process, beds of silica sand of 200-850 μm in diameter, were used as "seed" particles. A 100 mm diameter bed of 38 mm depth was used and the bed was fluidised with humid air at 0.49 m/s. Overall removal efficiencies of between 83 and 92% by weight were reported for initial fly ash loadings of up to 9 g/m², but grade efficiencies were not obtained. It is of interest that much of the fly ash in the penetrating gas was observed to be in agglomerated form, with agglomerate diameters of up to 80 μm, so that a
cyclone located downstream of the bed would have resulted in an even better overall efficiency. No deterioration in performance was observed with time. Using 13 mm deep beds of 400 μm pre-agglomerated ash particles fluidised at 0.15 m/s resulted in 98.5% removal by weight for an air relative humidity of 45%, and 100% removal when saturated air was used. Experiments were also carried out using porous aluminium silicate particles as collectors, rendered adhesive by introduction of oil. Results with 1000 μm particles at 0.64 m/s showed an improvement in removal efficiency of fly ash from 85% without oil to 95% with oil. 125 μm collector particles could not be fluidised with oil addition.

Enhanced collection was also achieved in beds of wax particles heated to 60°C to make them sticky, but no comparison with lower temperature performance is possible because the wax agglomerated to a new collector size at the higher temperature. In all the experiments reported, the penetrating aerosol was sampled onto a micropore filter and overall efficiencies obtained by weight only. Results would suggest that retention was complete in most cases, but comparison with available theory is not possible because insufficient data is given on the distributor type. This is said to consist of several layers of wire mesh, covered by a polypropylene fabric with "50-60 openings per inch". The pressure drop across the distributor was said to be negligible. Thus, collection in the jet region would have been small, but it is difficult to imagine that substantial collection on the distributor itself could have been avoided.

Doganoglu et al (1978) showed that non-porous particles could be conveniently rendered adhesive by using them to collect a liquid retention aid dispersed as an aerosol in the fluidising gas. Collection of solid methylene blue particles was improved by the pre-coating.
Ghadiri (1980), subsequently reported in Ghadiri et al (1985, 1986), studied the collection of fly ash and gasifier fines, of mass median diameters 2.2 μm and 3.6 μm respectively, in fluidised beds of sand at 800°C. Initial experiments with liquid boric oxide aerosol showed that the filtration efficiency was well predicted by the theory summarised in Section 2.1 and Appendix A1. However, filtration efficiency for solid dusts was again limited by poor retention. Two approaches were adopted to improve retention: for fly ash, the filter was operated at elevated temperature, above the softening point of the ash, and for both aerosols experiments were carried out using boric oxide as a retention aid, this approach being considered more suitable for dusts such as gasifier fines, which do not soften within the operating temperature range. Figure 2.9 shows the measured grade efficiency for gasifier fines at 800°C, filtered in a 45 mm deep bed of sand at

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**Figure 2.9** Grade Efficiency for 922 μm Silica Sand Filtering Gasifier Fines at 800°C and Superficial Gas Velocity 0.4 m/s. (after Ghadiri et al, 1985) $d_s \approx 0.89d_v$

- ○ = Coated Sand (0.0091 wt.% Boric Oxide)
- □ = Clean Sand
0.4 m/s, with and without retention aid addition. The theoretical grade efficiency, calculated according to the theory in Appendix A1, is plotted for comparison. A dramatic improvement in filtration efficiency is apparent, the change in sign of the gradient indicating a switch from rebound-dominated behaviour without retention aid, to inertial impaction-dominated behaviour with retention aid addition.

Figure 2.10 shows the effect of initial boric oxide loading on the overall penetration of gasifier fines through the filter, for two collector sand sizes, plotted against “surface cover parameter”. The surface cover parameter is defined as the product of the mass percentage retention aid in the bed and the mean bed particle diameter, and is proportional to the extent of coverage of the collector surface, or to the thickness of the film if there is complete coverage. The performance of the two sands is seen to be the same when plotted on this basis. The performance of the filter is dramatically improved by the presence of the retention aid. However, it was also observed that defluidisation occurred at relatively high levels of boric oxide loading on the bed particles, the onset being dependent on particle size. Large particles (922 μm) could carry much higher levels of boric oxide than small particles (461 μm) without causing defluidisation. The surface cover parameter does not, therefore, correlate the loading of retention aid at which fluidisation is lost. The problem of defluidisation is discussed further in Chapter 4.
Figure 2.10 Variation of Penetration with Surface Cover Parameter
(after Ghadiri et al, 1985) (Surface cover parameter defined as retention aid loading, as percentage of bed weight, multiplied by collector particle diameter in mm).
- □ 461 μm sand (U = 0.25 m/s)
- ✰ 922 μm sand (U = 0.40 m/s)

In the high temperature experiments with fly ash, Ghadiri et al (1986), show that the overall efficiency of filtration is very much better (87.5%) at 1000°C without retention aid than at 800°C with or without retention aid and at an equivalent gas velocity (the filtration efficiencies are 70.0% and 58.1% respectively). They concluded that retention is complete at 1000°C, due to the softening of the ash.

Patents by Clift et al (1982) and Cooke et al (1983) propose application of fluidised bed filtration to gas cleaning at high temperatures in industrial coal-fired power plant. However, no such applications have yet been realised. Clift et al (1981) described an application at moderate temperature in which a shallow fluidised bed of alumina was used to collect particulate fluorides in the "ALCOA 398" aluminium smelting process (Cook et al, 1971). The bed operated at
135°C. This approach was subsequently developed as the "ALCOA 446" process (Wei, 1975) for collecting a mixture of hydrocarbon vapours, coke dust and fluorides.

In more recent work at Warren Spring Laboratories, which is the subject of a patent application by Brodigan and Dawson (1982), a fluidised bed filter was used on a pilot scale to recover metallic values from metalliferous fume. In this development, the fume, consisting largely of submicron particles at high concentration (up to 7 g/m^3), was collected at 70 - 100°C in a shallow (typically 50 mm deep) bed of 1-2 mm diameter limestone fluorospar sand or dolomite particles, using a heavy fuel oil retention aid. The superficial gas velocity was typically 2 m/s, and a perforated or slotted plate distributor of high free-area (12.6%) was used. Particle removal efficiencies in excess of 90% are claimed. This work differs from the work discussed above in that the collection efficiency of submicron particles due to the mechanical collection mechanisms described in Section 2.1.1 would be low even with a low free-area distributor. For this reason, the Warren Spring filter incorporates a saw-tooth corona discharge plate below the distributor, and two grids within the bed, between which an electric field can be applied. However, it appears that in operation the corona charging also causes considerable and undesirable dust deposition on the upstream side of the distributor (Holt, 1984), which would cause operating difficulties in an industrial application.

The bed has two distinct modes of action. At low binder flowrates it will operate as a pre-agglomerator to a second bed or to a high efficiency cyclone as illustrated in Figure 2.1. At higher binder flow rates (around 5% of the mass flowrate of dust to the bed) the bed itself acts as agglomerator and collector. The resulting bed material
Figure 2.11  Warren Spring Fluidised Bed Filter - Schematic

is illustrated in Plate 2.1 and consists of a core of the original bed material surrounded by a coherent outer deposit with an unusual spiky shape, indicating that viscous bonds between the collector particles are continuously being made and broken within the bed. It is intended that this material should be used as a feed for a subsequent metal extraction and recovery process.

In a further application, developed at the University of Naples, the fluidised bed again acts as a submicron aerosol pre-agglomerator to a cyclone (Campanile et al., 1982; Barone et al., 1983). In this case, the filter medium (typically 500 μm sand) is not removed and replaced; instead, fine particles are captured by the bed material and removed by attrition in the form of much larger agglomerates (typically 1-40 μm).
Plate 2.1  Agglomerate Formed in the Warren Spring Fluidised Bed Filter.
The agglomerates are then entrained by the gas leaving the filter bed and collected in conventional cyclones (Figure 2.12). Campanile et al (1982) have developed this idea for collecting soot particles from diesel engine exhaust fumes, intending the filter to be sufficiently compact to be mounted on diesel-driven vehicles. Gutfinger et al (1977) had earlier proposed using a fluidised bed filter with renewed medium for cleaning diesel exhaust, but the filter developed by Campanile et al appears to be simpler in design and operation. The distributor is designed to give high collection efficiency and to ensure that the bed material cannot flow into the engine assembly. Provided that the bed particles are sufficiently large to be fluidised without significant entrainment from the bed, typically in the range 300 μm to 1 mm, then particles carried out by gases leaving the bed consist solely of soot and no renewal or recycle of bed material is necessary. About 70% of the soot particles are agglomerated from an initial size around 0.1 μm up to sizes which can be collected by a single cyclone, while the carbon content of the bed remains around or below 1% (Barone et al, 1983). It is also possible to provide heat exchange tubes within the fluidised bed for waste heat recovery, and this is attractive in the application of stationary diesel engines to co-generation of mechanical and thermal energy (Barone et al, 1983). A prototype of the Naples filter has now undergone 3000 hours of operation (Massimilla, 1986).
Figure 2.12 Naples Fluidised Bed Filter
2.3 Design Considerations.

As noted by Reed (1983), the capital cost of the tertiary filtration system selected for combined-cycle power generation depends critically on the number and size of filter vessels required and, therefore, on the filtration velocity. The use of a fluidised bed filter is attractive in this respect since superficial filtration velocities in excess of 1 m/s are possible, compared with 0.01 - 0.1 m/s for ceramic bag and candle filters. Furthermore, the volume required for an electrostatic precipitator is typically more than an order of magnitude larger than that required for a fluidised filter (Doganoglu et al., 1978). This advantage is balanced by a pressure drop penalty incurred in keeping the distributor orifice gas velocity high enough for efficient inertial collection. The distributor pressure drop will be approximately 0.5 bar at 15.5 bar system pressure.

Two alternative arrangements for hot gas cleaning in gasification combined-cycle could be envisaged. In both cases it is assumed that the primary and secondary cyclones are contained within the gasifier vessel. In the first design, the fluidised bed filter would also be incorporated within the gasifier vessel and would operate at approximately the same superficial gas velocity as the gasifier; in the alternative design the filter would be accommodated in a separate vessel. It is interesting to compare the size of such a vessel with those required for the ceramic candle filters considered by Reed (1983). In Reed's study, the total gas flow per gasifier was approximately 30 actual m³/s at 1000°C and 15.5 bar g. A 1200 MW power plant would require 5 such gasifiers. In the best case, the ceramic candles were assumed to operate at a face velocity of 0.033 m/s and were accommodated in 4 vessels per gasifier, each of 3.7 m internal
diameter. Each vessel contained 793 candles, giving a total filter area of approximately 900 m\(^2\) per gasifier. The pressure drop across each filter vessel was 7.4 kPa.

Assuming a superficial gas velocity of 0.5 m/s in the case of the fluidised bed filter, the distributor area required would be 50 m\(^2\), equivalent to 4 vessels per gasifier, each of diameter 4.4 m. However, the height required per bed is not more than 1.0 m, including distributor, which would make some form of tiered arrangement attractive. It might then be possible to accommodate all the beds within a single vessel not much larger than one of the ceramic candle vessels.

The mechanical complexity of the fluidised bed filter is considerably less than that required for the ceramic candle filter vessel, which requires a cooled tube sheet with ingenious fixings for the candle ends, and an elaborate network of high pressure reverse-pulse gas-distribution tubing. It is envisaged that the distributor of the fluidised bed would be dished or conical so as to compensate for differential expansion, to promote mixing and coating of the bed material, and to aid the removal of agglomerates from the bed. The complexity lies in the provision of a practical means of introducing the retention aid into the filter vessel, and this is considered in detail in Chapter 6. The atomising gas might be obtained either from the steam supply to the gasifier or by recompressing some of the clean product gas. In a practical design, provision would also have to be made for addition of collector material, via a lock hopper, for example, and discharge of the oversize agglomerates via a rotary valve.

The most convenient method of spent collector disposal might be by
addition (via a second lock hopper) to the combustion char transfer line. Combustion of the collected material would thus modestly augment the overall cycle efficiency.

2.4 Remaining Uncertainties

The work summarised in this chapter shows that fluidised bed filtration has the potential to form the tertiary gas-cleaning stage in a combined-cycle power plant. Enough is now known to predict collection efficiency with sufficient confidence to design a filter for a specific duty, provided that the twin problems of retention and de-fluidisation can be overcome, and it is these problems which are considered in the rest of this thesis.

The major gaps in our fundamental understanding of fluidised bed filtration, which need not however restrict the commercial development of the technique, relate to aerosol capture in the jet region and the phenomenon of aerosol rebound from the collector. Mechanistic models for both are in their infancy. The development of a model for collection in the jet region is of wider interest because it would assist in understanding particle and gas motion in the jet region and might help to resolve the current debate on the nature of this motion.
Chapter 3

Aerosol Dispersion, Sampling and Characterisation.

3.1 Aerosol Dispersion

The gasifier fines aerosol used for the experiments described in Chapter 6 was provided by the Coal Research Establishment of the National Coal Board and consisted of devolatilised carbon char collected from the secondary cyclone of a 0.3 m diameter atmospheric-pressure coal gasifier. A scanning electron micrograph of the particles collected on a 0.2 \( \mu \text{m} \) micropore filter is shown in Plate 3.1. The particles have a density of 1800 kg/m\(^3\) and contain approximately 80% coal char and 20% ash (Ghadiri, 1980).

The dust is not cohesive and is therefore suitable for dispersion using a fluidised bed feeder. The feeder used in this work was constructed by Ghadiri (1980), and is shown in Figure 3.1. The design is very similar to that proposed by Guichard (1976). In the arrangement used by Ghadiri, a pre-mixed 5% by weight mixture of gasifier fines with 150 \( \mu \text{m} \) glass ballotini was placed in the cylindrical feed hopper. The screw feeder (a modified spirally-wound test-tube brush) was set to rotate at 4.9 r.p.m. while the gas flow to the bed was maintained at 20 l/min (STP). This gives a superficial velocity in the bed of 47 mm/s, which corresponds to the terminal velocity of a particle of Stokes diameter 29 \( \mu \text{m} \). As the hopper gradually empties, ballotini and some coarser gasifier fines overflow into the overflow container. (The overflow was enlarged from that used by Ghadiri in order to relieve choking.) The size distribution of the aerosol before dispersion, measured by image analysis of microscope photographs (see Section
3.2.3.2), is given in Figure 3.2. For this work, the size range between 0.3 and 10 μm is of primary interest, representative of the size distribution of the aerosol present after passage through two stages of cyclones. In the initial experiments, therefore, the aerosol, dispersed in the primary dispersion gas, was passed through a settling chamber, to remove particles larger than 10 μm, before mixing with the secondary dispersion gas in a concentric eductor. The size distribution of the aerosol produced in this way is shown in Figure 3.3. Later in the experimental programme it became necessary to increase the concentration of the aerosol in the gas. This was done by feeding gasifier fines alone (i.e. not premixed with ballotini) from the feed hopper while retaining the ballotini in the bed. At the same time, the settling chamber was removed and the primary aerosol passed straight to the eductor. The gas flowrate to the bed was unchanged. In order to prevent particle deposition on the walls of the tube leading to the eductor, and to break up any agglomerates that may have been removed from the bed, a high velocity air jet was mounted at right angles to the bed exit pipe as shown in Figure 3.4. The size distribution of the aerosol resulting from the modification to the dispersion system is also shown in Figure 3.3. For both the original dispersion system and the modified system, careful sampling of the dispersed aerosol, by methods which are described later in this chapter, confirmed that the aerosol was fully dispersed and that the size distribution was stable and reproducible.
Plate 3.1  Redispersed Gasifier Fines Test Aerosol Collected on the Surface of a 0.2 μm Micropore Filter (Scanning electron microscope).
Figure 3.1 Gasifier Fines Feeder (after Ghadiri, 1980)
Figure 3.2  Size distribution of Gasifier Fines As Received (after Ghadiri, 1980)
Figure 3.3 Redispersed Gasifier Fines Aerosol.
3.2 Aerosol Sampling and Characterisation

3.2.1 Measures of Particle Diameter

For non-spherical particles, such as the gasifier fines used here, the definition of particle diameter must be specified. The following diameters have been used in this work. The volume-equivalent diameter, \( d_v \), is defined as the diameter of a sphere of volume equal to that of the particle:

\[
d_v = (6V/\pi)^{1/3}
\]

The volume-equivalent diameter, \( d_v \), is defined as the diameter of a sphere of volume equal to that of the particle:

\[
d_v = (6V/\pi)^{1/3}
\]

The projected-area-equivalent diameter, \( d_A \), is the diameter of a circle with area equal to the projected area of the particle:

\[
d_A = \sqrt{4A/\pi}
\]
In general, \( d_A \) may refer to a preferred particle orientation, or to random orientation. In the latter case, and for convex particles only, the average projected area, \( \bar{A} \), is given (Cauchy, 1908; Vouk, 1948) by:

\[
\bar{A} = \frac{S}{4} \tag{3.3}
\]

where \( S \) is the particle surface area. Hence

\[
d_A = \sqrt{\frac{S}{\pi}} \tag{3.4}
\]

The Stokes diameter, \( d_S \), is defined as the diameter of a sphere with the same density and settling velocity as the particle, assuming Stokes' Law to be applicable to the sphere. For a particle of density \( \rho_a \) settling in a gas of viscosity \( \mu \):

\[
d_s = \left[ \frac{18 \mu u_t}{C_s g (\rho_a - \rho_g)} \right]^{1/2} \tag{3.5}
\]

where \( u_t \) is the particle terminal velocity and \( C_s \) is the Cunningham 'Slip Correction' Factor. It can be shown (Ghadiri et al., 1986) that if settling is actually in the Stokes' Law range:

\[
d_s = \left[ \frac{3 \pi \rho_a d^3}{C} \right]^{1/2} = \left[ \frac{18 \nu}{C} \right]^{1/2} \tag{3.6}
\]

where \( C \) is the hydrodynamic resistance of the particle, \( 3\pi d \) for a sphere. Values of \( C \) can be calculated for a number of particle shapes in both random and preferred orientations (Clift et al., 1978). Finally, the aerodynamic diameter, \( d_{SO} \), is defined as the diameter of a sphere of density \( \rho_o = 10^3 \text{ kg/m}^3 \) which has the same settling velocity as the particle. Within the range of Stokes Law:

\[
d_{SO} = \left[ \frac{18 \mu u_t}{C_{SO} g (\rho_o - \rho_g)} \right]^{1/2} = d_s \left[ \frac{C_{so}(\rho_a - \rho_g)}{C_{SO}(\rho_o - \rho_g)} \right]^{1/2} \tag{3.7}
\]

where \( C_{SO} \) is the Cunningham factor for the sphere of diameter \( d_{SO} \). In
most practical cases, \( C_{50} \approx C_s \), and \( \rho_g \ll \rho_o \) and \( \rho_a \), so that

\[
d_{50} \approx d_s \left[ \frac{\rho_a}{\rho_o} \right]^{1/2}
\]  

(3.8)

For non-spherical particles, it is sometimes helpful to approximate the shape to that of a spheroid (Ghadiri et al, 1986). In this case, the shape is described in terms of the ratio of polar to equatorial diameter, known as the "aspect ratio", \( E \).

3.2.2 Measurement of Volume-Equivalent Diameter.

3.2.2.1 The "Coulter Counter"

Determination of filter efficiency relies on the measurement of the concentration and size distribution of particles in the gases entering and leaving the filter. In previous work on the efficiency of fluidised bed filters at high temperatures, Ghadiri (1980) isokinetically sampled the hot gases and passed them through micropore membrane filters of cellulose acetate, with a 0.2 \( \mu m \) nominal pore size. A short length of sample pipe outside the bed housing was sufficient to cool the gas sample to a temperature at which it caused no damage to the filter. He weighed the deposit on the filter to obtain overall efficiency and then ultrasonically redispersed it into dilute, particle-free sodium chloride solution (available commercially as "Isoton II") for size analysis using the "Coulter Counter" (Coulter Electronics Ltd.) In this instrument, which is described in detail by, for example, Allen (1981), an accurately known volume of the suspension of particles in electrolyte is sucked through a small, accurately-formed orifice, having an immersed electrode on either side, as shown in Figure 3.5. The size of the orifice (10-400 \( \mu m \)) depends on the
desired range of measurement. In a sufficiently dilute suspension, particles pass through the orifice one at a time. As each particle passes through, it displaces its own volume of electrolyte, thus changing the electrical resistance across the orifice. Since the current is kept constant, the change in resistance of the orifice due to the passage of a particle generates a voltage pulse whose amplitude has been shown (Scarlett, 1979; Harfield et al, 1984) to be proportional to the particle volume, provided that the particle diameter determined in this way does not exceed about 80% of the orifice diameter. (Harfield et al recommend 60% as a practical working limit.) The voltage pulses resulting from the passage through the orifice of a known volume of suspension are electronically scaled and counted, from which a particle size distribution can be derived. If the total mass of the sample is unknown, this can be obtained, provided the true density is known, by summing the total volume of the particles.

Figure 3.5 The Coulter Counter - Schematic
(after Allen, 1981)
counted in a known volume of electrolyte. Calibration is simply achieved by passing monodisperse spherical latex of known particle size through the instrument. The Coulter model "ZM" was used throughout the work reported here.

3.2.2.2 Selection of Electrolyte.

The Coulter Counter was used routinely by Batch (1964) for the size analysis of redispersed carbon particles, and by Ghadiri (1980) for sizing gasifier fines. However, during sizing runs using the same gasifier fines as used by Ghadiri (1980) anomalous electrical pulses were observed on the instrument monitor, which did not conform to the generally accepted pattern of pulses produced by particles passing through the orifice. The conditions used for these experiments are listed in Table 3.1 and the resulting pulse pattern sketched in Figure 3.6. It was at first thought that these pulses were due to external interference, but this possibility was rejected since:

(i) screening of the sampling stand did not remove them;
(ii) they were not observed when sizing fly ash or calibration materials.

The other possibility was that the effect might be due to conduction of part of the orifice current through the carbon particles. It may at first appear that the Coulter principle, which depends on measuring the increase in resistance through an orifice as a particle passes through, would not work at all for a highly conducting particle such as carbon. However, the principle has been successfully employed for conducting particles by a number of workers, such as van der Plaats and Herps
and Harfield (1982), and its use has been extended even to metal particles. The apparent non-conduction of particles of otherwise high conductivity has been variously ascribed to the formation of an oxide layer or an electrical double-layer around each particle. However, the successful size measurement of noble metal particles shows that these effects cannot provide a full explanation for the success of the Coulter technique. Harfield (1982) explains the effect by consideration of surface barrier potentials which must be overcome before conduction through the particle can occur. His investigations revealed three types of conductor:

(i) particles which produce a wholly negative (resistive) voltage pulse as they pass through the orifice;
(ii) particles which initially conduct and then become resistive (part positive, part negative pulse);
(iii) particles which produce a wholly positive (conductive) pulse.

Table 3.1 Coulter Counter Settings - Gasifier Fines Measurement

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Coulter Model ZM</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electrolyte</td>
<td>&quot;Isoton II&quot; (equivalent to 0.9% Na Cl in H2O)</td>
</tr>
<tr>
<td>Dispersant</td>
<td>Nonidet P40</td>
</tr>
<tr>
<td>Preset Amplifier Gain</td>
<td>5 k</td>
</tr>
<tr>
<td>Aperture Current</td>
<td>1 mA (except for extremes of distribution)</td>
</tr>
<tr>
<td>Aperture Size</td>
<td>50 μm.</td>
</tr>
</tbody>
</table>
In Harfield's work, errors in sizing due to particle conduction were eliminated by ensuring that the effective particle conductivity was less than twice that of the electrolyte, either by use of some surface active agent to increase the barrier potential or by increasing the electrolyte conductivity.

In order to investigate the possible influence of particle conductivity on measurements of the size of gasifier fines, two experiments were carried out:

(1) Using the standard Coulter electrolyte, Isoton II, the instrument was calibrated and the size distribution of a typical sample was measured three times, at constant orifice currents of 1 mA, 0.2 mA and 0.06 mA; the amplifier gain was adjusted appropriately to compensate for current settings. The results, shown in Figure...
3.7, indicate no significant dependence of apparent particle size on orifice current over this range, which represents almost the whole range available with the Coulter model ZM employed here. The results show, if anything, a slight decrease in measured mean particle size with decrease in orifice current. This is the reverse of the trend observed for carbon particles over a much wider range of orifice currents by van der Plaats and Herps (1983), but the reason for the discrepancy is not clear.

It was also observed that the frequency of anomalous pulses decreased with decreasing orifice current, while the frequency increased sharply on raising the orifice current momentarily to the maximum available value of 4.99 mA. It therefore appears preferable to use low orifice currents for relatively conductive materials.

(ii) Further determinations were carried out using a non-standard electrolyte of much higher conductivity, 10% NaCl solution. The Coulter counter was recalibrated with latex particles, and the calibration was found to be unaffected by the change in electrolyte. A further size distribution was obtained for the gasifier fines, using this stronger electrolyte with an orifice current of 1 mA. Results are shown in Figure 3-8. The new distribution has a slightly larger mean size than that obtained for the dispersion in Isoton; this effect of electrolyte conductivity on apparent size for conductive particles is in agreement with the findings of van der Plaats and Herps (1983) and Harfield (1982). The results of Batch (1964), reproduced here as Figure 3-9, also show that the mean size of pulverised coal indicated by Coulter measurement is larger for an electrolyte of
higher conductivity (i.e. lower resistivity). In the present work, no anomalous pulses were observed with the strong electrolyte.

It is therefore concluded that the relatively high conductivity of gasifier fines has a small but measurable effect on sizing by the Coulter method and that, in order to reduce errors from this effect, a high conductivity electrolyte should be employed. In the remainder of the work reported here 10% NaCl solution, prefiltered through a 0.2 μm micropore filter, was used as the Coulter electrolyte.

3.2.2.3 Advantages and Limitations of the Coulter Counter.

The Coulter counter measures the volume-equivalent diameter, $d_y$, as defined in equation (3.1). Since this is unambiguous in the sense that it does not depend on particle orientation, it is arguably the most useful measure of particle diameter. An added advantage of this representation is that, for oblate particles with aspect ratios down to about 0.1, and for prolate particles with aspect ratios up to about 60, $d_y$ never exceeds the Stokes diameter, $d_s$, by more than 4% (Ghadiri et al, 1986). For many practical purposes the difference between $d_y$ and $d_s$ can, therefore, be neglected, which is fortunate, since $d_s$ is the diameter which best describes the behaviour of particles in inertial collection and separation processes but it is much more difficult to measure directly than is $d_y$. 
Figure 3.7  Typical Gasifier Fines Size Distribution
(Coulter Counter) - Effect of Orifice Current

\[
\begin{align*}
& \text{Gain} & \text{Current (mA)} \\
& 5K & 1.0 \\
& 20K & 0.2 \\
& 80K & 0.06
\end{align*}
\]

Figure 3.8  Typical Gasifier Fines Size Distribution
(Coulter Counter) - Effect of Electrolyte Conductivity.

\[
\begin{align*}
& \text{Electrolyte} & \text{Conductivity (m}^{-1}\text{m}^{-1}) \\
& \text{Isoton II (0.9\% Na Cl)} & 1.82 \\
& 10.1\% \text{ Na Cl} & 13.3
\end{align*}
\]
Although the volume-equivalent diameter is in many ways the most useful measure of particle diameter, it is nevertheless time-consuming to measure it in the way described above. On-line measurement by this method is impractical, making it impossible to follow rapid changes in filter performance with time. The ultimate particle sizing requirement is for a portable on-line instrument, having a fast response and preferably producing the size distribution of the Stokes diameter, or, less desirably, of a diameter which can be related to the Stokes diameter. The instrument should be flexible in size and concentration ranges and insensitive to variation in temperature and pressure of the carrier gas. In the section which follows, the gasifier fines aerosol, dispersed at ambient temperature by the method described in Section 3.1, has been used to test a number of instruments which are claimed to have some of these advantages: several single-particle light-scattering
counters, a Fraunhofer-diffraction size analyser and an aerodynamic particle sizer. Precautions were taken in order to keep the aerosol concentration low so as to avoid "coincidence" losses (the simultaneous presence of more than one particle in the sensing volume). For each measuring method, the relationships between the measured diameters have been derived and compared with theoretical predictions.

3.2.3 Other Aerosol Characterisation Methods.

3.2.3.1 Single-Particle Light-Scattering Counters

Two single-particle light-scattering counters were investigated, with characteristics summarised in Table 3.2. Both operate in a similar way: the gas to be sampled is drawn through a small view-volume where it is illuminated by light from an incandescent light source. The forward-scattered light is collected by an elliptical mirror or, in some instruments, a lens, and refocussed onto a photomultiplier where it is converted into a pulse of electrical current, which is in turn converted into a voltage pulse. Successive voltage pulses are classified by peak height and counted to provide information on particle number in a certain size range and so, over a pre-selected period, a size distribution by number is built up. Different instruments differ in sampling rate, size of view-volume and the internal angles of the optics, as shown in Figure 3.10a.

The theory behind the light scattering technique is as follows. Consider a particle, of refractive index \( m \), in a plane formed by the incident light beam, of wavelength \( \lambda \), and the direction of observation (Figure 3.10b). The incident light energy may be deflected or
absorbed. Deflection is considered to arise from reflection, refraction, and diffraction on the particle cross-sectional area; the deflection process is referred to as "scattering". The scattered light-intensity, \( I(\theta) \), may be defined as the amount of electromagnetic energy which crosses unit area perpendicular to the flow per unit time.

### Table 3.2

**Characteristics of Single-Particle Light-Scattering Counters.**

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Bausch &amp; Lomb*3</th>
<th>Climet CI-208*4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particle Size Range (( \mu m ))</td>
<td>&gt;0.5 - &gt;10</td>
<td>&gt;0.5 - &gt;10</td>
</tr>
<tr>
<td>No. of Size Channels</td>
<td>6*5</td>
<td>6*5</td>
</tr>
<tr>
<td>Optical Angles *(°)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( \beta )</td>
<td>53</td>
<td>90</td>
</tr>
<tr>
<td>( \gamma )</td>
<td>13</td>
<td>16</td>
</tr>
<tr>
<td>( \delta )</td>
<td>33</td>
<td>36</td>
</tr>
<tr>
<td>(cm(^3)/min)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>View volume (mm(^3))</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td>Concentration *2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>giving a 5% coincidence error (no./cm(^3))</td>
<td>100</td>
<td>100</td>
</tr>
</tbody>
</table>

*Notes *

1. Defined as in Figure 3.10a.
2. Calculated according to Hinds (1982)
3. No longer manufactured.
4. Now superceded by CI-208C.
5. Fixed size thresholds.
(W/m²). The incident illumination is considered parallel, laterally coherent and unpolarised and account is taken of the scattered flux regardless of polarisation. The general theory of light scattering by spheres is that derived by Mie and is a complete solution to Maxwell's equations for the incident light wave, the wave inside the particle, and the scattered wave, subject to a set of boundary conditions at the particle surface. (Kerker, 1969, derives and explains the theory). Two simplifications of Mie theory are possible. For small particles (πd/λ < 0.3, d < 0.05 μm in visible light) the Rayleigh scattering theory can be used, in which the relative angular distribution of scattered light, I(θ), is the same for all particle sizes, shapes and refractive indices. I(θ) is then proportional to d⁶/λ⁴. For large particles (πd/λ > 3/(m-1), d > ~ 2 μm in visible light) the forward scattered light intensity (θ < 90°) can be approximated by Fraunhofer diffraction theory (see Section 3.2.3.3). However, for the range of particle diameters of interest here, 0.3 - 10 μm, Mie theory should be used.

At a distance R in the direction θ from a spherical particle illuminated with unpolarised light of intensity I₀, Hinds (1982) gives the scattered intensity as:

\[ I(θ) = I₀λ² (i₁ + i₂)/8π² R² \]

where \( i₁ \) and \( i₂ \) are the Mie intensity parameters for scattered light polarised in the perpendicular and parallel directions; \( i₁ \) and \( i₂ \) are complicated functions of refractive index, m, size parameter \( α = \frac{πd}{λ} \), and scattering angle θ. Two general observations are of importance:
(i) As the scattering angle, $\theta$, increases towards $90^\circ$, the scattered light intensity becomes more sensitive to changes in refractive index, $m$, particularly whether or not it is absorbing (Hodkinson, 1966), and to departures from sphericity;

(ii) As the size parameter, $\alpha$, increases, the angular scattering pattern becomes more complicated. A plot of Mie intensity parameters, $i_1$ and $i_2$, against $\theta$ will show variations in intensity of several orders of magnitude over a few degrees of $\theta$ for $\alpha > 10$ (Hinds, 1982).

For these two reasons most commercial counters integrate the scattered light over the forward direction ($\theta < 90^\circ$), ensuring the least dependence of response on particle refractive index and shape. Integration over a number of incident wavelengths (i.e. by using an incandescent white light source) also serves to damp out oscillations in the Mie response curves. Cooke and Kerker (1975) obtained the theoretical responses of a number of counters by integrating the Mie intensities over the collecting aperture and showed that, for those integrating the response in the forward direction, there is comparatively little dependence on non-absorbing (real) particle refractive index for $d > 1$ $\mu$m. For absorbing (complex) refractive indices, however, the responses are very different, being in general much lower than for a real refractive index with the same (real) part. Instruments with less convergent illumination (larger $\gamma$; Figure 3.10a) and smaller collection angles (smaller $\beta$-8) show a multivalued response curve, the principal uncertainty being in the range between indicated particle diameters of 0.5 and 1 $\mu$m. The effect of refractive index on experimental response curves is considered further by Whitby and Vomela (1967), Liu et al (1974), and Makynen (1983), among others.
Figure 3.10a Single-Particle Light-Scattering Counter – Optical Geometry.

- $\beta$ = collecting aperture half angle
- $\gamma$ = illuminating cone half angle
- $\delta$ = light-trap half-angle

Limiting scattering angles: upper bound $\beta + \gamma$
lower bound $\delta - \gamma$

Figure 3.10b Light-Scattering by a Single Particle.
For aerosols of industrial interest, which are in general both irregular and absorbing (and also frequently of inhomogeneous refractive index), theoretical light scattering predictions are too complicated to be used to predict the response of a particular single-particle counter, especially when the vagaries of a particular instrument (such as non-linearity of the photomultiplier) are taken into account. Even for spherical aerosols of known refractive index, theoretical response prediction is tedious, so it is usual practice to factory-calibrate each instrument using mono-sized polystyrene latex spheres (refractive index = 1.59) and to incorporate a calibration device, usually a "light chopper", into the instrument. If the instrument is to be used on an aerosol which is non-spherical and/or of a different refractive index, it is essential for it to be re-calibrated for the aerosol of interest, which is experimentally difficult. However, it is useful to be able to predict qualitatively what the response of the counter will be to non-spherical absorbing particles, in order to relate the "light-scattering diameter" to other particle diameters of interest, such as those listed in Section 3.2.1.

Predictions of scattering intensities for non-spherical particles are generally inexact and incomplete. In the range $0.3 < \alpha < 20$, particles with no great inequality between their different dimensions have a scattering pattern similar to that of spheres of equal volume (Hodkinson, 1966), although the upper size limit for which this generalisation can be made depends on the nature of the particle. Pollack and Cuzzi (1979), in their semi-empirical theory for scattering by non-spherical particles comparable in dimensions to the incident wavelength, use this limit as an adjustable parameter, a typical value for plate-type particles being $\alpha=3$ ($d \approx 0.5\mu m$). For a randomly-oriented non-spherical particle which is large compared with the wavelength of
the incident light, the forward diffraction lobe of the scattering pattern depends largely on projected area and is not significantly affected by particle shape (Hodkinson, 1966). Outside the forward diffraction lobe the main deviation in the scattering behaviour of a large irregular particle arises from internally transmitted and refracted light (Pollack and Cuzzi, 1979), whose behaviour is very sensitive to shape. The effect is to increase the scattering intensity at large scattering angles well above that for a sphere of equivalent projected area (Pollack and Cuzzi, 1979; Zerull et al, 1979). The scattering pattern for large opaque particles should deviate less from the spherical case since in this case there is scattering by diffraction and external reflection only (Hodkinson, 1966). Thus the response of a single particle counter is sensitive to both refractive index and shape of the particles sampled. For example, changing the refractive index from 1.54 (non-absorbing) to 1.54 - 0.5i (strongly absorbing) causes the Bausch and Lomb 40-1A to undersize by a factor of 3 to 4 (Cooke and Kerker, 1975). In addition to this effect a correction factor for particle shape must be applied, which will be near unity for small particles but larger than one for larger particles. Larger absorbing particles are therefore undersized less than equivalent spheres of the same refractive index.

It should be noted that single particle counters as a group are also subject to count losses due to "coincidence", the inability of the electronics to process more than one pulse simultaneously, and "spreading" of the observed distribution, whereby not all particles of the same size are counted in the same channel. These problems are considered in detail by Pisani and Thompson (1971), van der Meulen et al (1980) and Makynen et al (1982), among others.
3.2.3.2 Optical and Scanning Electron Microscopy

The techniques for optical and scanning electron microscopy, followed by size analysis using an automatic image analysing instrument, are well established. Those used in this work are described in detail by Ghadiri (1980). In brief, a small number of particles were directly sampled onto a micropore filter or redispersed in liquid and allowed to settle onto a filter. In either case, care has to be taken to ensure that the deposited particles do not touch or overlap one another because this makes subsequent image analysis more complex. (In practice this condition is achieved by keeping the sample time short.) The collection surface is then viewed either optically or, for particles as small as those of interest here, using the scanning electron microscope. Micrographs are taken under conditions of high contrast and these are then placed under the macro-viewer of an image analysis computer, such as the "Quantimet" (Cambridge Instruments Ltd.). This device digitises the image, so that areas corresponding to a particular grey-level (in this case, the particles) can be identified, counted, and chosen properties, such as area and perimeter, recorded. Derived properties, such as the diameter of the circle of equivalent area, can then be calculated and tabulated or plotted. If proper precautions are taken when depositing the particles, the diameter distribution obtained represents the maximum projected area for each particle.

3.2.3.3 Field Scattering Instruments

A number of instruments have been developed which convert the angular distribution of forward scattered light intensity from a multi-particle field measurement into a size distribution. The example used in this
work was the Malvern 2600 (Malvern Instruments Ltd.) with which it is possible to measure the scattered light from both gas-borne and liquid-borne samples. The instrument is fully described by Swithenbank et al (1976) and Faraday (1983), and the principles of the method are critically reviewed by Bayvel and Jones (1981). The aerosol (or hydrosol) is illuminated in a parallel beam of monochromatic light ($\lambda = 0.6328 \, \mu m$) light from a low-power He/Ne laser, as shown in Figure 3.11. A lens collects the scattered light and focusses it onto an annular photosensitive detector positioned at the focal plane of the lens. The proportion of light falling on each of the 30 rings is converted to a particle volume distribution, assuming the scattering pattern to obey Fraunhofer diffraction theory. In theory it is possible to obtain the particle volume distribution for spherical particles directly from the measured light energy distribution. In
practice the system of equations which must be solved is unstable (Leschonski, 1983) and this leads to oscillating size distribution functions for small errors in measured light energy. In all but the most recent instruments, therefore, alternative methods are therefore employed: either a model for the particle size distribution (normal, log-normal or Rosin-Rammler) is assumed and fitted in a least squares sense or a set of 16 weight bands is set up and an iterative heuristic technique used to obtain a satisfactory fit (the "model independent" method). Use of interchangeable collection lenses enables a theoretical size range of 1.2 µm to 1800 µm to be covered. In this work a 63 mm focal length lens was used, giving a nominal range of 1.2-118 µm. In order to reduce multiple scattering effects, it is essential not to have too many particles in the illuminated zone; in practice this condition is easily met, even for fuel sprays. There are two theoretical difficulties with the technique: the range of applicability of the Fraunhofer diffraction assumption, and so-called "anomolous diffraction". Fraunhofer diffraction is a good approximation to Mie theory for particles which are much larger than the wavelength. Jones (1977) has investigated the error in particle sizing associated with the Fraunhofer assumption and concludes that for an error on particle diameter of less than 20%, the size parameter, $\alpha (= \pi d/\lambda)$, must be greater than about 20 if $m_1(\text{real}) > 1.3$ (for non-absorbing particles) and $\alpha$ must be greater than about 8 for $m_1(\text{real}) > 1.5$ (for strongly absorbing particles). Jones' error contour plots, giving the range of applicability of Fraunhofer theory, are shown in Figure 3.12. For He/Ne laser light Jones' limits correspond to 4.0 µm and 1.6 µm respectively; the limits are increased as the relative refractive index becomes closer to unity. Under these conditions, the reflection terms
Figure 3.12 Error contour charts comparing the Mie theory with diffraction. In the shaded areas errors are greater than 20%. In the cross-hatched areas they are negative. (after Jones, 1977) $\alpha = \pi d / \lambda$; $\alpha'$ from diffraction theory only.
in the full Mie formulae go to zero and only the terms which describe the interference between the relatively undeflected refracted rays and the diffracted component are retained. The result is the so-called "anomalous diffraction" theory, which the more recent Malvern software uses to obtain particle sizes close to the wavelength of light. However, Tüzün and Farhadpour (1985) show that, not surprisingly, the "anomalous diffraction" theory is only a good approximation if the relative refractive index is close to unity, e.g. in liquid suspension. In this work, only Fraunhofer diffraction theory was used, so that the Malvern would be expected to produce a volume distribution derived from the number distribution of spheres of equivalent diffraction pattern (Leschonski, 1983). For many practical applications this will approximate to the volume distribution of projected area diameter, in either random or preferred orientation, depending on the hydrodynamic conditions. Brown and Felton (1985) have shown that this approximation gives satisfactory results for large (~ 100 μm) particles of various model shapes, but there is, as yet, no experimental proof that it is reliable for particles of diameters nearer the wavelength of light.

3.2.3.4 Aerodynamic Particle Sizer

A recently developed instrument for particle size measurement is the Aerodynamic Particle Sizer (TSI Inc., Agarwal and Remiarz, 1981), which measures aerodynamic size directly. Particles are drawn singly through a high gas-velocity nozzle where they are accelerated at a rate which depends directly on their aerodynamic size. Their exit velocities are measured by "time of flight" between two laser beams separated by approximately 120 μm and individual measurements are used to build up a
particle size distribution. The theory of the instrument is summarised by Wilson and Liu (1980). Since, in principle, it measures a balance between Stokes drag and particle inertia, it should measure the aerodynamic diameter $d_{50}$, from which the Stokes diameter, $d_s$, can be inferred using equation (3.8) if the particle density is known. Whether the indicated value for $d_{50}$ corresponds to random or preferred orientation depends on whether the particles align on passage through the nozzle. In order to eliminate the possibility of concentration effects, measurements were carried out both with and without a TSI-manufactured 100:1 diluter.

3.2.4 Experimental Results

The test aerosol was dispersed in ambient temperature air as outlined in Section 3.1. The concentrated stream was then mixed with dilution air in a concentric tube diluter, before passing to a 0.15 m diameter glass column from which it was isokinetically sampled into each of the sampling instruments (i.e. the single-particle light-scattering counters and the Aerodynamic Particle Sizer). Periodically the aerosol stream was also isokinetically sampled onto 0.2 μm micropore filters for analysis by Coulter counter. The liquid-phase Malvern samples were also obtained in this way. The gas-phase Malvern is not a sampling instrument and so the technique used here was to project a thin stream of undiluted aerosol in free air through the incident laser beam within the focal length of the lens, collecting some of the aerosol isokinetically on the other side of the beam for subsequent Coulter analysis. Coulter counter measurements for the two test aerosols are presented on a cumulative number basis in Figure 3.13. From the
volume-equivalent diameter measured by Coulter counter and the
distribution of the maximum projected-area-equivalent diameter,
obtained from optical and scanning electron microscopy, it is possible
to obtain information about particle shape (Ghadiri et al, 1986). The
aspect ratio, $E$, of the equivalent oblate spheroid (see Section 3.2.1)
was found to be 0.191 for gasifier fines, uncorrelated with particle
size. Using this estimate of $E$, values have been calculated for the
ratios $dy/dg$ and $dy/d_{A}$, in preferred and random orientations, for
comparison with experimental results. Also presented in Figure 3.13
are the cumulative number distributions for the same dispersion
measured using the single-particle light-scattering counters.
Calibration curves were simply obtained by comparison between
volume-equivalent diameter and indicated diameter at the same value of
cumulative percentage. These are presented for the single-particle
light-scattering counters in Figure 3.14. Volume distributions from
the Malvern measurements are compared with volume distributions derived
from the Coulter measurements in Figure 3.15, and the corresponding
calibration curves are presented in Figure 3.16. Number distributions
from the Aerodynamic Particle Sizer are compared with number
distributions of Stokes diameter (derived as outlined in Section 3.1)
in Figure 3.17, and the corresponding calibration curves are presented
in Figure 3.18.

3.2.5 Discussion

Direct comparison between the measured distributions for polydisperse
non-spherical particles such as these is difficult because the effect
of shape is not only to shift the mean size of the measured
distribution but also to broaden it (Liu et al, 1974; van der Meulen,
Measured Diameter [μm]

Figure 3.13 Cumulative Number Distributions of Test Aerosol
- Coulter and Single-Particle Light-Scattering Counters.

Cumulative number percent undersize

Equivalent volume sphere diameter [μm]

Figure 3.14 Calibration Curves for Single-Particle Light-Scattering Counters (from number distributions)

<table>
<thead>
<tr>
<th>Experimental</th>
<th>Theoretical</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Climet</td>
<td>A. B &amp; L response for spheres;</td>
</tr>
<tr>
<td>2. B &amp; L Fines</td>
<td>m = 1.54 - 0.5i</td>
</tr>
<tr>
<td></td>
<td>(Cooke &amp; Kerker, 1975)</td>
</tr>
</tbody>
</table>

 ● B & L - India ink
   (Whitby & Vomela, 1967)
Figure 3.15 Cumulative Volume Distributions of Test Aerosol - Coulter & Malvern (Gas-Borne & Liquid-Borne)

Approximate Fraunhofer 20% error limits (Jones, 1977)

Absorbing
- \( m_1 > 1.5 \)

Non-absorbing
- \( m_1 > 1.3 \)
- \( m_1 = 1.1 \)

Figure 3.16 Calibration Curve for Malvern Diffraction Sizer (from volume distributions)

Experimental

1. Gasifier fines (gas-borne)
2. Gasifier fines (liquid-borne)

Theoretical

A. Gasifier fines (diameter in random orientation).
Figure 3.17 Cumulative Number Distributions of Test Aerosol - Stokes Diameters Derived from Coulter, SEM and Aerodynamic Particle Sizer Measurements.

**Experimental**

(Converting from aerodynamic diameter to Stokes diameter using equation (3.8))

1. Gasifier fines

**Theoretical**

(Calculated Stokes diameter from Coulter and microscopy)

A. Random orientation

B. Preferred orientation

Figure 3.18 Calibration Curve for Aerodynamic Particle Sizer (from number distributions of Stokes diameter; see Figure 3.17 for key to curves).
This is generally true for all the instruments considered here, with the exception of the Coulter counter. However, observation of the distributions in Figures 3.13, 3.15 and 3.17 reveals no pronounced spreading effect. Therefore, in this work, in which the comparisons are confined to the central portions of the distributions, "spreading" was neglected.

3.2.5.1 Single-Particle Light-Scattering Counters

Figures 3.13 and 3.14 illustrate the effect on light-scattering counters of changes in particle refractive index and shape. The responses of the two instruments are qualitatively similar, as one would expect of instruments which all integrate the forward scattered light. The effect of particle shape is overwhelmed by the effect of a highly absorptive refractive index (1.59 - 0.66i for carbon, λ = 0.49 μm (Hinds, 1982)); particles are therefore undersized by the Bausch & Lomb and the Climet by 40-60%. The results of experiments by Whitby and Vomela (1967) who counted monosized India ink, a rough-surfaced, highly absorptive, near-spherical aerosol, using the Bausch and Lomb 40-1A, show similar undersizing. Figure 3.14 also shows a theoretical line for the sizing of spheres of refractive index 1.54 -0.5i in the Bausch and Lomb 40-1A, derived by Cooke and Kerker (1975) from Mie calculations. The difference between this line and the experimental results again suggests the effect of particle shape which tends to reduce the undersizing resulting from absorption. The effect
of particle shape is greater than can be accounted for by simply assuming that the instrument is measuring the projected area in random orientation.

3.2.5.2 Malvern Diffraction Sizer

For the strongly absorbing gasifier fines, Fraunhofer theory applies down to about 1 μm. It would be expected, therefore, that the experimental results for gasifier fines would be in good agreement with theory. However the results for gas-borne and liquid-borne systems do not agree closely but bracket the predicted line for projected area diameter in random orientation, which probably indicates failure of the approximations in the Malvern software as the particle size approaches the wavelength of the incident light.

3.2.5.3 Aerodynamic Particle Sizer

Figures 3.17 and 3.18 show the results of measurements of the Stokes diameter of gasifier fines using the Aerodynamic Particle Sizer (APS). The aerodynamic diameters measured by the APS have been converted here to Stokes diameter using equation (3.8). For comparison, the Stokes diameters calculated from equation (3.6) for random and preferred orientations are also included. It may be noted that the calculated Stokes diameters in random orientation agree closely with measurements by sedimentation techniques (Ghadiri et al, 1986). The preferred orientation is taken as that with the plane of maximum projected area normal to the direction of flow, which corresponds to maximum drag (Clift et al, 1978) and hence maximum discrepancy between volume-equivalent and Stokes diameters.
The measured Stokes diameter falls 30 - 60% below the calculated value, the difference varying with particle size. Obviously, the effect of particle orientation is not sufficient to explain the discrepancy. However, it may be possible to attribute the difference to the sharp edges of the particles. The predicted values of Stokes diameter have been calculated from the hydrodynamic resistance of the particle in Stokes flow. In this case, sharp edges on the particles do not have a significant effect on the drag (Clift et al, 1978). In contrast, the conditions in the APS are such that the presence of sharp edges may be significant. The geometry and flow conditions of the nozzle are optimized for maximum resolution of small particles and accuracy of sizing for large particles (Wilson and Liu, 1980). Consequently, large particles may experience a Reynolds number substantially larger than 0.1 during their acceleration in the nozzle. This does not affect spherical particles since the drag can still be calculated accurately. For non-spherical particles, the situation is complicated. The sharp edges determine the position of boundary layer separation and force it to occur at lower Reynolds numbers, thus affecting the drag significantly; for example, separation from a disc occurs at Reynolds numbers an order of magnitude lower than for a sphere (Clift et al, 1978). Furthermore, particles would tend to re-orientate themselves when the Reynolds number is larger than about 1, resulting in rotation and tumbling. The Stokes diameter measured in this range of Reynolds numbers may therefore be different from the diameter which would be measured for steady motion in the Stokes regime. It is of interest that significant undersizing of plate-like talc particles by the APS has also been observed by Rood and Griffiths (1984).
3.2.6 Conclusions of Comparative Study.

The results presented above suggest that:

(i) Single-particle light-scattering counters seriously undersize gasifier fines; this is primarily due to the particle refractive index, for which the shape effect may partially compensate.

(ii) For particles in the range of interest here, Fraunhofer diffraction is a reasonably good approximation, although the fact that results for gas- and liquid-borne particles disagree indicates that there is some refractive index effect, even in this size range.

(iii) The Aerodynamic Particle Sizer undersizes irregular particles, possibly due to increased drag caused by edges and angularities and by tumbling motion along the measurement path.

So far as suitability for the purpose of measuring gas-borne fine particulates is concerned, the following observations are made:

(a) Single-particle light-scattering counters are very sensitive to shape and refractive index effects and must be pre-calibrated using the aerosol of interest sized by an independent technique, such as those proposed by Marple and Rubow (1976) and Fissan et al (1984). The Malvern instrument is rather less sensitive to refractive index and shape effects, except at the lower extent of its range where the assumptions upon which the instrument design is based become invalid.
The sizing method used in the Aerodynamic Particle Sizer is insensitive to refractive index and, in principle, to shape, although the results presented here cast some doubt on its validity for irregular particles.

(b) The single-particle light-scattering counters and the Aerodynamic Particle Sizer, being single-particle counters, measure the number distribution directly and unambiguously (apart from the "spreading" effect) providing that their upper concentration limits are not exceeded. This means that, for accurate filtration measurements, they are preferable to the Malvern, which only measures volume distribution. However, single particle counters require special dilution systems for measuring particles at high concentrations. Single particle counters are effectively subject to no lower concentration limit, whereas the Malvern is subject to signal-to-noise ratio problems at low concentrations.

(c) In the gas-borne mode of operation the Malvern is not a sampling instrument and therefore causes no disturbance to the measured flow and is relatively insensitive to duct temperature and pressure. For these reasons, it is preferable to the single-particle counters as an on-line instrument. The requirement that the aerosol to be measured must pass within the focal length of the collection lens is, however, a restriction.

(d) In this application it is desirable to know the distribution of Stokes or aerodynamic diameter in the aerosol. Thus, in principle, the Aerodynamic Particle Sizer is the most attractive instrument of the three types considered, since it measures directly the diameter of
interest. The light scattering instruments measure only the number or volume distribution of spheres of equivalent scattering, which is frequently difficult to relate to the diameter of interest.

In summary, none of the on-line methods discussed here is considered suitable for determining the grade efficiency of a hot fluidised bed filter. For the experiments described in Chapter 6, therefore, off-line Coulter analysis was used, the particle samples having been collected on micropore filters or in a liquid impinger, the design of which is discussed in the next section.

3.2.7 Liquid Impinger Sampler

In the experiments described in Chapter 6, a heavy oil was injected into the fluidised bed filter in order to improve particle retention. This caused operating problems with the conventional membrane-filter gas-sampling system because of condensation of cracking products on the membrane, leading to a rapid rise in pressure drop across the filters. Typically, the correct sampling rate could not be maintained for longer than a few minutes, which is insufficient time to collect an analysable sample. In order to overcome this problem, a "liquid impinger" was designed, according to the recommendations of Drinker and Hatch (1954), Cadle (1975) and Wright (1984). This device, which is illustrated in Figure 3.19, consists of a nozzle facing an impaction plate. In operation, both are normally submerged in the collection liquid and the gas to be sampled is drawn through the nozzle, producing a high velocity jet. The turbulent gas and liquid flow around the impaction plate keeps the plate clean, while the particles in the gas are
Figure 3.19 Liquid Impinger

(Only essential dimensions shown; jar cap omitted)
projected into, and captured by, the liquid. The liquid impinger is thus effectively the same as an inertial impactor in so far as its collection behaviour is concerned. However, it overcomes the well-known impactor problems of bounce and re-entrainment by retaining the collected particles in liquid. Schadt and Cadle (1957) determined the collection efficiencies of a liquid impinger and a conventional inertial impactor designed according to the same rules, for monodisperse aerosols of sodium chloride, glycerol and stearic acid. Collection of the solid sodium chloride aerosols in the impactor was in poor agreement with theory, showing poor collection at both small and large aerosol sizes, with a maximum collection at some intermediate size, which is characteristic of particle "bounce". Collection of the same aerosols in the liquid impinger was found to depend on particle size only, and was not affected by the physical form of the aerosol.

If the collection liquid is a suitable solvent, such as chloroform, the collected carbon particles can be separated from the chloroform-soluble condensation products in the impinger itself. It is even possible to conceive of an arrangement in which the impinger would be used, in conjunction with a Coulter counter, as a continuous particulate monitoring device. In such an arrangement the collection liquid could be the Coulter electrolyte, which would be sampled continuously into the counter, filtered and then returned to the impinger. However, for the work reported here no dedicated counter was available, so the Coulter electrolyte was used as the collection liquid, which was then analysed off-line in the usual way.
Since the impinger is, in effect, an inertial impactor, it is to be expected that its design will conform to similar rules. Figure 3.20 shows the essential design variables. It is conventional to characterise the performance of impactors by means of an impactor Stokes number (e.g. Marple and Willeke, 1976):

\[
St = \frac{\rho_a d_a^2 U C}{\mu d_j}
\]  

(3.10)

where \(\rho_a\) and \(d_a\) are the density and Stokes diameter of the aerosol, \(U\) is the superficial gas velocity in the nozzle, \(C\) is the Cunningham slip correction factor for the aerosol, \(\mu\) is the gas viscosity, and \(d_j\) is the diameter of the nozzle. \(St\) may be thought of as the ratio of the particle stopping distance at velocity \(U\) to the nozzle radius \((d_j/2)\). The Stokes number corresponding to 50% particle collection in the impactor is denoted \(St_{50}\), and corresponds to the "cut-off" particle diameter, \(d_{a,50}\). For an "ideal" impactor all particles larger than \(d_{50}\) will be collected, while all those smaller will not. For a well-designed impactor with a circular nozzle, Hinds (1982) gives \(St_{50} = 0.22\). Substituting for \(U\) in equation (3.10) in terms of the sample flowrate, \(Q\), and re-arranging, we have

\[
d_j = \left[\frac{4\rho_a d_{a,50} C Q}{9\mu \ St_{50}}\right]^{1/3}
\]

(3.11)

from which a design value of \(d_j\) can be calculated, corresponding to a cut-off particle size of \(d_{a,50}\). In practice, this requires iteration since \(C\) depends on \(d_{a,50}\). Table 3.3 sets out the values of \(d_{a,50}/C\) corresponding to the values of \(d_j\) of the three interchangeable nozzles made up for this work. It is worth noting that the aerosol-laden jet emerging from the nozzle expands only slightly until it reaches within about one jet diameter of the impaction plate (Hinds, 1982). Hence, the impactor (or impinger) performance is relatively insensitive to
changes in the nozzle-to-plate distance, \(a\), provided that \(a/d_j > 1\) (Marple and Liu, 1974). The nozzle length, \(b\), must be sufficient for fully developed flow. Collection performance is again insensitive to changes in \(b\) provided that this minimum condition is met. Marple and Liu also recommend that the nozzle Reynolds number, \(\rho g U_j d_j/\mu\), should be greater than 500. As Table 3.3 shows, this condition is met for all three nozzle sizes.

Two impactors were made up to the design shown in Figure 3.19 and the three interchangeable nozzles were tested in turn under hot sampling conditions. The penetration of aerosol through the impinger was found to be negligible, even with the largest nozzle fitted (although a large penetration was recorded with no nozzle fitted and the end of the 6.3 mm internal diameter pipe directed onto the impaction plate). This impinger design, with the largest nozzle fitted, was therefore adopted for further work.
Table 3.3 Impinger Design

<table>
<thead>
<tr>
<th>Nozzle diameter $d_j$ (mm)</th>
<th>0.5</th>
<th>0.75</th>
<th>1.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cut-off diameter $d_{a,50/C}$ (μm)</td>
<td>0.24</td>
<td>0.45</td>
<td>0.69</td>
</tr>
<tr>
<td>Reynolds no. in jet $\rho U_d d_j/\mu$</td>
<td>5500</td>
<td>3670</td>
<td>2750</td>
</tr>
</tbody>
</table>

Calculations for air at room temperature and pressure, gasifier fines aerosol and a sampling rate of $2 l$(actual)/min.

Tests were then carried out to establish the accuracy with which the total mass of sample could be estimated from the integrated volume of all the particles collected in the liquid impinger. Samples of aerosol were collected on membrane filters and weighed on a microbalance. The same samples were then redispersed into 10% wt sodium chloride solution and a size distribution obtained for each using the Coulter counter. Integrating the total volume of solids in each sample and multiplying by the particle density yielded results within 5% and, with care, within 1% of the weighed value.

3.3 Summary

For dispersion of gasifier fines in the size range of interest and at comparatively low concentration the fluidised bed feeder designed by Ghadiri (1980) has been found reliable.
Methods for on-line particle characterisation have been investigated but none has been found suitable for this work. Single-particle light-scattering counters are too sensitive to particle shape and refractive index effects and are severely limited in their concentration range, while the theory of diffraction is not applicable over the whole range of interest, so that the Malvern instrument is inappropriate for this work. The principle behind the Aerodynamic Particle Sizer makes this a promising technique, but errors in sizing non-spherical particles, and concentration limits, mean that this instrument is also unsuitable for fluidised bed filter test work.

For the remainder of the work described in this thesis, therefore, particle characterisation has been carried out off-line, using the Coulter counter, collecting the particles in a liquid impinger to avoid problems with condensibles blocking the collection filters. The collection liquid was 10\% wt. NaCl solution, which eliminates the particle conduction effects experienced when using the Coulter counter with weaker electrolytes.
4.1. Types of Fluidisation Behaviour.

When a fluid is passed upwards through a settled bed of particles, there comes a point (for many particle/fluid systems) at which, as the fluid velocity is increased, the drag force on the particles becomes equal to the buoyant weight of the bed. Since, by definition, the drag force on the particles is equal to the "manometric" pressure drop across the bed (i.e. that part of the pressure drop arising solely from the fluid motion) we can say that at this, the point of incipient fluidisation, the manometric pressure drop, $\Delta P$, is equal to the buoyant weight of the bed, or

$$\frac{\Delta P}{L} = (1-\epsilon_{MP}) (\rho_p - \rho_g) g \quad (4.1)$$

where $L$ is the bed height,

$\epsilon_{MP}$ is the voidage at minimum fluidisation, and

$\rho_p$ and $\rho_g$ are the densities of the particles and the fluid, respectively.

The manometric pressure drop, $\Delta P$, is related to the difference between mean absolute pressures measured at the entrance to and the exit from the bed, $\Delta P'$, by

$$\Delta P' - \rho_g g L = \Delta P \quad (4.2)$$

For gas fluidised beds, where $\rho_g \ll \rho_p$, the distinction between $\Delta P$ and
\( \Delta P \)' be can ignored for all practical purposes.

The superficial fluid velocity at minimum fluidisation, can be found by combining equation (4.1) with a suitable expression for the pressure drop through a settled bed of solids, such as the correlation due to Ergun (1952):

\[
\Delta P = 150 \frac{(1-\varepsilon)^2}{\varepsilon^3} \frac{\mu U}{(\phi_s d_p)^2} + 1.75 \frac{1-\varepsilon}{\varepsilon^3} \frac{\rho_d U^2}{\phi_s d_p} \tag{4.3}
\]

where \( \mu \) is the viscosity of the fluid,

\( U \) is the superficial fluid velocity,

\( \phi_s \) is the particle sphericity, i.e. the ratio of the surface area of the equivalent volume sphere to the surface area of the particle, and

\( d_p \) is the volume-equivalent-sphere diameter.

The first term in equation (4.3) represents the viscous energy losses while the second represents the kinetic losses, which are important only at higher values of the Reynolds number, \( Re_p = \rho_d d_p U / \mu \). At lower values of the Reynolds number (\( Re_p < 20 \)) the second term in equation (4.3) can be neglected; the equation then has the same form as that used by Carman (1937) and known as the Carman-Kozeny equation. \( U_{MF} \) is now simply found by equating (4.1) and (4.3) (evaluated at \( U_{MF} \)) and solving the resulting quadratic equation.

It is clear that the actual value of \( \Delta P \) in equation (4.3) is very sensitive to changes in the voidage, \( \varepsilon \), and that if the voidage is inhomogenous, as it may well be, then the expression for \( \Delta P \) will be more complex than this.
A simple force balance on the bed shows that it is not possible for the pressure drop to exceed the buoyant weight of the particles. At higher fluid velocities, therefore, either the bed voidage must increase so as to maintain the pressure drop at or below this level, or not all the fluid can flow interstitially. In the former case, the increase in bed voidage can occur by uniform expansion, while in the latter, an increase in flow results in (i) the formation of jets, spouts, bubbles, or slugs, (ii) the nucleation of stationary cavities, or (iii) channelling or "rat-holing" from the distributor to the surface. Strictly speaking, all but the last type of behaviour can be described as fluidisation, that state in which both the bed and individual particles within it are wholly supported by the pressure drop, while the last cannot, since, in general the pressure drop will be less than that required to support the bed.

Several attempts have been made to devise theoretical and empirical criteria to distinguish between these behavioural types, most of which relate only to gas-solid fluidisation, reflecting the greater industrial importance of this phenomenon over liquid-solid fluidisation. Of these, the most successful and widely used is the empirical classification of Geldart (1973), who divides fluidisation behaviour according to mean particle size and density difference between the solids and the fluidising gas, as shown in Figure 4.1. Geldart recognises four behavioural groups, designated A, B, C and D. Typical fluidisation behaviour of groups A, B and C is illustrated in Figure 4.2.
Group B particles fluidise easily, with bubbles forming at or only slightly above the minimum fluidising velocity. Bed expansion is small and the bed collapses very rapidly when the gas supply is cut off.

Group C particles are "cohesive" and tend to lift as a plug or to channel badly; conventional fluidisation is difficult or impossible to achieve.

Group A particles are intermediate in particle size and in behaviour between groups B and C, and are distinguished from group B by the fact that appreciable (apparently homogeneous) bed expansion occurs above the minimum fluidisation velocity but before bubbling is observed. Geldart (1973) defines a minimum bubbling velocity, $U_{MB}$, and designates group A particles as those for which $U_{MB}/U_{MP} > 1$. When the gas supply is suddenly cut off the bed collapses slowly, at a rate which is similar to the superficial gas velocity in the dense phase. There is
Figure 4.2 Typical Fluidisation Behaviour of Particles in Geldart's (1973) Groups A, B and C (note that scales are different for each group).
now much experimental evidence, which is discussed further in this chapter, that group A particles are also intermediate in cohesiveness between groups B and C, their interparticle cohesive forces being of the same order as the particle weight.

Group D particles are those which are "large" and/or abnormally dense. The distinction between groups B and D is not clear-cut. Empirically, group D particles are capable of maintaining a stable spout in a bed more than 0.3 m deep; theoretically, the bubbles in group D rise more slowly than the interstitial fluidising gas, which is the opposite behaviour to that predicted and observed in group B. For this group, interparticle forces are negligible by comparison with particle weight and can be safely neglected. Other properties of the groups are summarised in Table 4.1.

Martin (1983) has suggested an extension of Geldart's classification to incorporate liquid-solid systems. His conceptual classification, shown in Figure 4.3, is in terms of mean particle size, density difference between the solids and the fluid, and fluid viscosity. While Geldart's group B ("aggregative" fluidisation), in which behaviour is dominated by gravity forces, can clearly be extended to describe liquid-solid systems, the essential feature of Martin's classification is that it distinguishes between group A ("delayed-bubbling") and a new group, E, which contains those systems which exhibit homogeneous liquid-solid type expansion ("particulate fluidisation"). In group A fluidisation, bed expansion occurs by nucleation of cavities (see Section 4.2), the surrounding particles maintaining surface contacts which are essential for the stability of the structure. In moving from A to E the increase in hydrodynamic drag with fluid viscosity breaks down this cavity structure to give a uniformly expanded bed, in which particle-particle
Table 4.1 Characteristic Features of Geldart's (1973) Classification of Fluidisation Behaviour (after Geldart, 1981)

<table>
<thead>
<tr>
<th>GROUP</th>
<th>C</th>
<th>A</th>
<th>B</th>
<th>D</th>
</tr>
</thead>
<tbody>
<tr>
<td>MOST OBVIOUS CHARACTERISTIC</td>
<td>Cohesive, difficult to fluidise</td>
<td>Bubble-free range of fluidisation</td>
<td>Starts bubbling at Umf</td>
<td>Coarse solids</td>
</tr>
<tr>
<td>TYPICAL EXAMPLES</td>
<td>Flour, Cement</td>
<td>Cracking catalyst</td>
<td>Building sand, table salt</td>
<td>Crushed limestone, coffee beans</td>
</tr>
<tr>
<td>PROPERTY</td>
<td>Low when bed channels; can be high when fluidised</td>
<td>High</td>
<td>Moderate</td>
<td>Low</td>
</tr>
<tr>
<td>1. Bed expansion</td>
<td>Can be very slow</td>
<td>Slow</td>
<td>Fast</td>
<td>Fast</td>
</tr>
<tr>
<td>2. De-aeration rate</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3. Bubble properties</td>
<td>Channels</td>
<td>Splitting and coalescence predominates</td>
<td>No limit on size</td>
<td>No known upper size Small wake</td>
</tr>
<tr>
<td>4. Solids mixing</td>
<td>Very low</td>
<td>High</td>
<td>Moderate</td>
<td>Low</td>
</tr>
<tr>
<td>5. Gas back-mixing</td>
<td>Very low</td>
<td>High</td>
<td>Moderate</td>
<td>Low</td>
</tr>
<tr>
<td>6. Slug properties</td>
<td>Solid slugs</td>
<td>Axi-symmetric, break down to turbulent fluidisation</td>
<td>Asymmetric</td>
<td>Horizontal voids Solid slugs Wall slugs</td>
</tr>
<tr>
<td>7. Spouting</td>
<td>No, except in very shallow beds</td>
<td>Shallow beds only</td>
<td>Shallow beds only</td>
<td>Yes, even in deep beds</td>
</tr>
</tbody>
</table>
contacts are transient. It follows, therefore, that in the latter case interparticle forces do not play a significant role. The distinction is important in the arguments which follow, in that it reveals the misleading nature of comparisons between gas-solid group A behaviour and liquid-solid particulate fluidisation.

Geldart's classification, although widely adopted and undoubtedly of great practical value, is really only applicable to "hard" solids exhibiting only "natural" interparticle forces (i.e. van der Waals forces and moderate humidity effects), fluidised by air at normal pressures and temperatures in the earth's gravitational field. (Even within this range the relative paucity of results for particles of abnormally high or low density makes it difficult to define the group boundaries accurately.) However, the classification of fluidisation in terms of experimentally observed behaviour is of wider application,
even in cases where interparticle forces are artificially enhanced, as will be shown in Chapter 5.

The rest of this chapter is concerned with the effect on fluidisation behaviour of modification in interparticle adhesive forces in gas-solid systems.

4.2. Fluidisation in the Presence of Interparticle Adhesive Forces.

4.2.1 The Nature of Interparticle Forces

The interparticle forces which can influence the behaviour of particulate systems have been extensively reviewed by Rumpf (1962) and more recently by Schubert (1981). Rumpf's classification of forces is summarised in Table 4.2 below. (It should be noted that more than one mechanism may act simultaneously.)

Table 4.2 Rumpf’s Classification of Forces

<table>
<thead>
<tr>
<th>Bonding Type</th>
<th>Origin of Adhesion Forces</th>
</tr>
</thead>
<tbody>
<tr>
<td>Solid bridges</td>
<td>Sintering, heat hardening</td>
</tr>
<tr>
<td></td>
<td>Chemical reaction</td>
</tr>
<tr>
<td></td>
<td>Incipient melting due to pressure and/or friction</td>
</tr>
<tr>
<td></td>
<td>Deposition through drying</td>
</tr>
<tr>
<td>Immobile liquids</td>
<td>Viscous binders</td>
</tr>
<tr>
<td></td>
<td>Adsorption layers</td>
</tr>
<tr>
<td>Mobile liquids</td>
<td>Liquid bridges</td>
</tr>
<tr>
<td>Intermolecular and long-range forces</td>
<td>Van der Waals forces</td>
</tr>
<tr>
<td></td>
<td>Electrostatic forces</td>
</tr>
<tr>
<td></td>
<td>Magnetic forces</td>
</tr>
<tr>
<td>Mechanical interlocking</td>
<td>Shape-related bonding.</td>
</tr>
</tbody>
</table>
In practice, interparticle forces for many of the mechanisms listed above cannot be calculated a priori (Capes, 1980). However, interparticle forces due to van der Waals forces, electrostatic forces, and mobile liquid bridges can be calculated for simple model geometries. The cases of van der Waals forces and mobile liquid bridges, which are of importance in the arguments which follow, are considered briefly below.

4.2.1.1 Van der Waals Forces

Van der Waals forces arise from a number of intermolecular interactions, all of which are ultimately electrostatic in origin. The way in which these intermolecular forces are integrated to provide the force between two microscopic bodies is considered in Appendix A2. Israelachvili (1985) gives an expression for the attractive force between a rigid sphere and a plane surface from which it is separated by a distance $a$:

$$F_W = \frac{A R}{6a^2}$$  \hspace{1cm} (4.4)

where $A$ is the Hamaker constant (J) and $R$ is the radius of the sphere (m). Calculated and some experimental values of the Hamaker constant for different materials are given in Appendix A2. Equation (4.4) obviously implies that as $a$ goes to zero the interparticle force becomes infinite. $a$ is usually taken to have a minimum value which is related to the average interatomic distance, which is about $4 \times 10^{-10}$m for most solids. The appropriate value for $a$ is considered further in Appendix A2. For the case of contact between two spherical particles of radii $R_1$ and $R_2$, van der Waals forces can be obtained from equation (4.4) by substituting:
Since van der Waals forces are of short range, the force between a rough body and a plane must be calculated using a value of \( R \) corresponding to the radius of curvature of the asperity at the contact point (assuming only one point of contact).

### 4.2.1.2 Mobile Liquid Bridges

The four regimes of bonding which can exist between particulates due to mobile low-viscosity liquids are shown in Figure 4.4. At low liquid loadings, discrete lenses of liquid are formed at the contact points; this is termed the "pendular" state, and is the only one of the four of practical interest here.

Figure 4.5 shows a liquid bridge between two equal spheres of radius \( R \). The force between the spheres has two components: the axial component of the surface tension force at the liquid/gas/solid interface and a hydrostatic force due to the difference in pressure, \( \Delta P \), between the liquid interior and the surroundings. This pressure difference, which is known as the capillary pressure, is related to the surface tension of the liquid, \( \gamma \), by Laplace's equation (Shaw, 1980):

\[
\Delta P = \gamma \left[ \frac{1}{r_1} + \frac{1}{r_2} \right]
\]  

(4.6)

where \( r_1 \) and \( r_2 \) are the principal radii of curvature. The pressure difference, \( \Delta P \), (and the force due to \( \Delta P \)) may be either positive (repulsive) or negative (attractive) according to the signs and
Figure 4.4 Distribution of Liquids in Agglomerates. (after Capes, 1980)

Figure 4.5 Liquid Bridge Between Two Equal Spheres (perfect wetting). 
\(a = \text{half particle separation.}\)
relative magnitudes of \( r_1 \) and \( r_2 \). The Laplace equation requires that the free liquid surface must have constant total curvature, \( \frac{1}{\kappa} \), where
\[
\frac{1}{\kappa} = \frac{1}{r_1} + \frac{1}{r_2},
\]
which makes the exact calculation of \( \kappa \), and hence of the interparticle force, rather complex. In practice, however, the calculation of interparticle force is made easier, with little loss in accuracy, by using a toroidal approximation (Fisher, 1926), i.e. by assuming that the meridian curvature of the bridge is a circle. The total attractive force, \( F_C \), is then easily determined for the case of zero liquid/solid contact angle (perfect wetting) and zero particle separation (see Appendix A3):

\[
F_C = \frac{2\pi\gamma R}{1 + \tan(\beta/2)} \tag{4.7}
\]

where \( \gamma \) is the surface tension of the liquid (N/m) and \( \beta \) is the half-angle of the liquid bridge. Equation (4.7) predicts an increase in the magnitude of \( F_C \) for a decrease in the size of the liquid bridge (represented by \( \beta \)) until a maximum value is reached at zero liquid content. Although careful laboratory experiments, such as those of Cross and Picknett (1963) and Mason and Clark (1965), reproduced this trend down to very low liquid volumes, this behaviour is nevertheless the opposite of what one would intuitively expect, i.e. wetter powders commonly appear stronger than drier ones (until the capillary state is reached, at which point the strength is reduced). Pietsch (1968) attempted to resolve this apparently paradoxical conflict between theory and experimental result by suggesting that all real contacts are rough and that an effective sphere separation, \( a \), should be included in the theory, as shown in Figure 4.6. Calculation of \( F_C \) for non-zero particle separations is considered in Appendix A3 and values of \( F_C \) are presented in Figure 4.7, as functions of the liquid bridge half-angle,
The interparticle force is predicted to show a maximum value at a bridge half-angle which depends on the separation, $a$. In general, of course, $\beta$ and $a$ are not known; Pietsch (1969) suggests that for engineering purposes it would be reasonable to take

$$F_C = 4\gamma R$$  \hspace{1cm} (4.8)

for most of the pendular range, provided that the liquid fully wets the solid. Thus, $F_C$ in equation (4.8) constitutes about two thirds of its maximum possible value. Coughlin et al. (1982) have shown both theoretically and experimentally that pendular liquid bridges give rise to rather different interparticle force behaviour when the contact is conical, the force increasing with the amount of liquid in the bridge (although the maximum force remains similar to that given by equation (4.7)). This result is considered further in Appendix A3 and presents a further explanation for the inadequacy of Fisher's original theory.

It should be noted that liquid bridge forces differ from van der Waals forces in at least three important respects. While the particle-particle attraction due to van der Waals forces decreases rapidly but continuously with increasing separation, the force-separation curve for liquid bridges shows a discontinuity at the point of bridge instability and breakage. The particles must subsequently re-establish contact in order to re-form the bridge. Secondly, van der Waals forces (and electrostatic forces) have short relaxation times, so that there is no practical difference between their static and dynamic values (Clift, 1985), while rapid separation of particles linked by liquid bridges can give rise to viscous forces many times greater than those arising from surface tension and capillary forces alone (see Appendix A3). Thirdly, whereas the magnitude of the van der Waals attraction always depends on
Figure 4.6 Schematic Representation of a Liquid Bridge Between Two Particles With Surface Asperities. (after Pietsch, 1968)

Figure 4.7 The Dependence of Normalised Adhesion Force, $F/2\pi r_y$, Between Two Equal Spheres on the Bridge Half-Angle, $\beta$. 
the radius of the contact point or points, even if this is very
different from the radius of the particle, liquid bridge forces depend
on the scale of the asperities and the bridge. When very small amounts
of liquid are present, the appropriate contact radius may be that of a
small asperity; at larger liquid loadings, however, the asperity
contact becomes irrelevant (except in determining the effective
particle separation, as in Figure 4.6) and the appropriate radius to be
used in equations (4.7) and (4.8) is that of the particle itself.

4.2.1.3 Comparison of Interparticle Forces with Particle Weight.

Figure 4.8 presents the theoretical interparticle forces arising from
equations (4.4), (4.7) and (4.8) for the case of point contact between
smooth spheres in air, with particle weight plotted for comparison.

The values of the parameters are as follows:

\[ A = 6.5 \times 10^{-20} \, \text{J} \quad \text{(quartz, Israelachvili, 1985)} \]
\[ \gamma = 72.8 \times 10^{-2} \, \text{N/m} \quad \text{(water)} \]
\[ \rho = 3 \times 10^3 \, \text{kg/m}^3 \quad \text{(typical value for sand)} \]

Values for van der Waals force are presented for interparticle
separations of $4 \hat{R}$ (Krupp, 1967) and $1.65 \hat{R}$ (Israelachvili, 1985). The
dashed lines indicate the interparticle forces assuming an asperity-to-
plane contact with an asperity radius of 0.1 \( \mu \text{m} \).
Figure 4.8 Comparison Between Capillary Force, van der Waals Force and Particle Weight, for the Conditions listed in Section 4.2.1.3. (Dashed lines indicate air-particle contact.)
4.2.2 "Natural Forces" : Van der Waals Forces and Capillary Forces due to Gas Humidity

It has long been recognised that the difficulty encountered in attempting to fluidise very fine particles is associated with the presence of interparticle forces which are large by comparison with the individual particle weight. If the particles are sufficiently small, van der Waals forces alone are sufficient to prevent fluidisation, which is the condition giving rise to the A/C boundary in Figure 4.1.

Baerns (1966) investigated the fluidisation behaviour of a number of materials with mean particle sizes between 1 and 200 μm. He evaluated their interparticle forces by a method due to Cremer et al (1952) which consists of measuring the angle of inclination at which a certain mass of material will slide as a bulk down a rough substrate. Knowing the size of the particles and the total area of contact, an average adhesive shear force per particle, \( F_p \), can be calculated, which, it is claimed, can be related to the forces which determine the stability of channels in very cohesive material. Figure 4.9 shows the relationship between Baerns' "fluidisability index", the ratio between the calculated and the measured fluidisation velocity, and the force ratio \( G_p/(G_p + F_p) \) where \( G_p \) is the particle weight. Baerns concludes, on the evidence of this diagram and simultaneous measurements of heat transfer coefficients in the bed, that fluidisation is possible for \( G_p/(G_p + F_p) > 0.05 \) (\( F_p/G_p < 19 \)) and is impossible for \( G_p/(G_p + F_p) < 0.003 \) (\( F_p/G_p > 332 \)). In general the boundary between fluidisable and non-fluidisable systems is imprecise, as reflected in Baerns' range of values for the critical particle adhesion/particle weight ratio.
Figure 4.9  Ratio of Calculated to Measured Minimum Fluidisation Velocity as A Function of Force Ratios $G_p/(G_p+F_p)$ and $F_p/G_p$ (after Baerns, 1966).
- Nickel spheres
- Iron spheres
+ Hollow glass spheres
- Irregularly shaped alumina
More recently Geldart et al (1984) and Geldart and Wong (1984) have systematically investigated the differences in fluidisation behaviour between group A and C particles. Differences in the ratio between the expanded and settled bed heights, and in the deaeration rates were explained by the increasing effects of cohesive forces as the particle size is decreased. These differences were found to correlate with the value of the Hausner ratio (Hausner, 1967), the ratio of the highest and lowest bulk densities (i.e. the "tapped" and the "poured" bulk densities, respectively). The Hausner ratio is clearly another indirect measure of interparticle force; looser packings can be stabilised by higher interparticle forces, while the densest packing is unaffected by interparticle forces. Higher values of the Hausner ratio therefore imply higher cohesive forces. Geldart et al. (1984) estimate that the transition from group A to group C behaviour occurs at Hausner ratios between 1.25 and 1.4. There has been no attempt as yet to derive values for interparticle forces from measurements of the Hausner ratio.

Chaouki et al (1986) studied the fluidisation behaviour of very fine, very light aerogels (d_p < 20 µm; \( \rho_p \approx 66 \text{ kg/m}^3 \)) which should be well within Geldart's group C. In fact, above a certain critical superficial velocity they spontaneously agglomerate to form loose clusters with diameters of order 1 mm. The cluster weight is now comparable with the inter-cluster force (which is determined by the original \( \approx 20 \mu m \) particle diameter) and they fluidise smoothly, exhibiting a large expansion without bubbling. Thus, their behaviour has been transformed by spontaneous agglomeration from group C to group A.
Our current understanding of the behaviour of particles within group A of Geldart's classification is largely due to the work of the two research groups under Professor Massimilla at the University of Naples and Professor Rietema at the University of Eindhoven. This work will now be considered in detail because of the light which it sheds on the effect of interparticle forces on fluidisation in general. The aim is to show that the same van der Waals forces which are responsible for preventing fluidisation in group C particles are also responsible for the stabilisation of the expanded bubble-free bed which is the characteristic of group A particles at fluidising gas velocities between $U_{MF}$ and $U_{MB}$.

Several workers (the observations are often first attributed to Davies and Richardson, 1966) have observed that particles now designated as group A exhibit bed expansion before bubbling and it has been possible to correlate the extent of this expansion in terms of the well-known Richardson and Zaki (1954) equation:

$$\frac{U}{U_t} = \epsilon^n$$

(4.9)

where $U_t$ = particle terminal velocity in an infinite medium and $n$ is a function of the particle Reynolds number at the terminal velocity, $U_t$.

This expression was first used to correlate the homogeneous expansion of liquid fluidised beds (where $n \approx 4.65$), and the fact that it also serves to correlate the expansion of gas fluidised beds in the bubble-free region (though with rather different values for $n$; see below) has been used by some to suggest that the latter occurs by the same mechanism. Dimensional analysis (Coulson and Richardson,
1978) shows that the general form of equation (4.9) must apply, in the absence of interparticle forces, but since it is of such a general form, having no other theoretical justification, it is not surprising that it can be used in these two, rather different, cases. Figure 4.10 shows the expansion data of a number of workers re-plotted by Geldart and Wong (1984) in the form of $n/4.65$ against the particle diameter. Geldart and Wong conclude that values of $n$ greater than 4.65 may be regarded as indicating the importance of interparticle forces.

![Figure 4.10 Normalised Richardson-Zaki Index, $n/4.65$, as a Function of Particle Size - for a range of gases and bed materials. (after Geldart and Wong, 1984).](image)

Massimilla et al (1972) and Donsi and Massimilla (1973c) studied the bubble-free expansion of a range of group A powders including silica and alumina catalysts, PVC powder and sodium bicarbonate. By direct observation through the flat wall of a semi-cylindrical column they were able to examine the structure of the expanded bed. Their observations are as follows:
(1) The relationship between fluidising gas velocity, $U$, and voidage, $\varepsilon$, is as predicted by equation (4.9), but with rather larger values of $n$ than those expected for liquid fluidised beds at the same value of $Re_p$. Thus the average drag per particle in the bed is higher than that predicted at the same voidage for homogeneous (liquid-fluidised bed) expansion.

(2) Bed expansion occurs by nucleation and growth of cavities whose sizes range from a few to about ten particle diameters. The fraction of the bed surface adjacent to the wall made up of cavities larger than $d_p^2$ in area was defined to be the “total surface cavity fraction”, $a_{Ca}$. Values for $a_{Ca}$ vary linearly with $U$ from about 0.07 at $U = U_{MF}$ to as much as 0.3 for finer materials at $U = U_{MB} = 3U_{MF}$. This cavity fraction, if extended into three dimensions, is sufficient to explain the observed bed expansion.

(3) The increase in voidage with increase in $U$ is accommodated by a step increase in the cavity number density at $U = U_{MF}$, followed by a linear increase in average cavity size for $U > U_{MF}$; the cavity number density, $N_{Ca}$, i.e. the number of cavities per unit bed surface at the wall, is inversely proportional to $d_p^2$ and independent of $U$ for $U > U_{MF}$.

(4) The cavity size distribution is approximately log-normal in $A_{Ca}/A_{Ca}$, independent of $d_p$, and increasing in skewness with increase in $U/U_{MF}$.
(5) Cavities generally show an elongated shape, the ratio of the major to minor axes ranging from about 2 to 6 and increasing with increase in $U/U_{MF}$. Cavities tend to elongate preferentially along paths inclined at between $45^\circ$ and $90^\circ$ to the vertical.

(6) Transient vertical channels appear in the velocity range $\frac{1}{2}(U_{MF} + U_{MB}) < U < U_{MB}$. These features are small in number compared with the cavities and are very short lived. They appear as small unstable spouts of diameter about $5 \, d_p$ and length about $50 \, d_p$ and are, in general, the only cause of solids motion. Most of the bed is stable and forms a honeycomb-like cavity structure.

These observations are consistent with the following model. The settled bed ($U < U_{MF}$) consists of particles in contact, with a range of interparticle bond strengths. At $U = U_{MF}$, the weakest of these bonds will break, nucleating cavities with a fairly narrow size distribution. However, the local gas velocity is itself affected by the presence of voids. From potential flow theory the maximum through-flow velocity in an isolated spherical void is equal to $3U$. This value increases if the vertical dimension exceeds the horizontal one (Donsi and Massimilla, 1973b). Thus, the drag forces are greatest in the vicinity of the largest voids and these will show an enhanced capacity for expansion, leading to the observed increase in skewness of the cavity size distribution with increase in $U/U_{MF}$, and the constancy of the cavity number density for $U/U_{MF} > 1$. This mechanism might extend

1 In fact it is unnecessary to assume a range of bond strengths, although this will almost certainly be the case. It can be shown (Donsi and Massimilla, 1973a) that the energy required to cause homogeneous expansion of a cubic array of spheres in the presence of van der Waals forces is in excess of that required to expand the array by the formation of unconnected cavities.
up to bubble development, either directly, or through the formation of channels.

The nucleation of cavities is also consistent with the increased value of the Richardson and Zaki index, $n$, over that expected from liquid fluidisation experiments. Donsi and Massimilla (1973b) compare the fluidising velocity/voidage relationship predicted from uniform expansion of liquid fluidised beds with the relationship which would be expected if only isolated spherical voids formed (Figure 4.11), with the flow through the voids calculated according to the analysis of Lockett et al (1967). The measured points lie closer to the predictions of the isolated voids model than to those of the uniform expansion model, but depart increasingly from the former at higher gas velocities, perhaps reflecting the formation of interconnections between cavities. The distinction between expansion by genuinely homogeneous increase in particle–particle separation and expansion by formation of local voids could correspond to the difference between Martin's (1983) groups A and E.

It remains to show that the interparticle bond strengths are sufficient to stabilise the observed structure. Massimilla and Donsi (1976) have investigated the surface characteristics of a number of fine catalyst types using a scanning electron microscope. Even those which are almost perfectly spherical are revealed to have of order $10^6$ sub-particles and surface asperities per mm$^2$ of surface, ranging in diameter from 0.6 to 0.1 μm (the lower limit of accurate measurement and also the smallest asperity size which is capable of resisting mechanical stresses at the contact point; asperities smaller than a limit of this order, depending on material properties, are thought always to deform plastically on contact (Puttick, 1985)). Not all of
these can be considered as effective contacts between particles because the large asperities prevent the smaller ones from making contact. (Thus the error in disregarding any asperities which may be smaller than 0.1 μm is small). The "contact number density", i.e. the number of effective contact points per unit area, was constant for all particle sizes in the range 40 - 150μm. Using the corrected effective asperity diameter distribution (which had a number mean of about 0.2 μm) equations (4.8) and (A2.5) were then used to obtain the ranges of values for the cohesive forces due to van der Waals and capillary forces which are shown in Figure 4.12. For the Ludox catalyst considered (specific gravity 1.53) the computed van der Waals forces are seen to be of the same order as particle weights in the region 80 < \( d_p < 150\mu m \), while the capillary forces are an order of magnitude larger. Calculation of the actual particle-particle tensile force, \( F_T \), in the expanded bed is complex. Donsi and Massimilla (1973b)
Figure 4.12 Comparison Between Particle Weight and Interparticle Forces at Each Particle-Particle Contact

(Massimilla and Donsi, 1976)

$F_C$ and $F_W$ based on particle radius;

$F'_C$ and $F'_W$ based on mean asperity radius.

Measurements on cracking catalyst (Donsi et al., 1975)
calculated the stress distribution arising from the through-flow of gas in a spherical void. They concluded that tensile forces predominate in the solids surrounding the upper half of the void, with compressive forces acting around the bottom half. The maximum tensile force, $F_{T,\text{MAX}}$, acts as the uppermost point of the void, and is given by:

$$\frac{F_{T,\text{MAX}}}{G_p} \sim \frac{D_{Ca}}{D_p}$$  \hspace{1cm} (4.10)

where $D_{Ca}$ is the diameter of the void. They estimate from their measurements for particles well into group A that values of $F_T/G_p$ of up to 5 are likely. Referring again to the catalyst of Figure 4.10, the maximum interparticle forces due to van der Waals effects alone exceed 5 times the particle weight for particle diameters below about 70 μm. If capillary forces are invoked, this limit is raised to about 150 μm. For this density, 70 μm particles would be expected to exhibit classic group A behaviour, while dry 150 μm particles would be just within group B. The ratio $F_{T,\text{MAX}}/G_p$ increase rapidly with elongation of the cavity in a vertical direction, due to the greater flow concentration; this explains the tendency towards development of vertical channels.

Rietema (1967), Rietema and Mutters (1973) and Mutters and Rietema (1977) present further evidence for the existence of an interparticle stress field, i.e. a structure with a certain mechanical strength, in expanded beds of group A material. Figure 4.13 shows the variation in electrical current at constant voltage passing through a bed of fine charcoal as the superficial gas velocity is increased. This they take as evidence of continuous particle to particle contact even for $U > U_{MB}$. They also observe that the electrical conductivity measured in the vertical direction exceeds that in the horizontal direction (at the
same locus in the bed). This is further evidence of an anisotropic bed structure with a tendency towards formation of vertical channels.

Rietema and Mutters (1973) also showed that as a small fluidised bed in its expanded state is tilted away from the vertical, interparticle forces were sufficient to prevent the bed surface from shearing off until a critical angle, \( \alpha \), was reached. The value of \( \alpha \) decreases with increasing voidage, as shown in Figure 4.14. The critical bed voidage at which tilting was no longer possible (i.e. \( \alpha = 0 \)) coincided with the maximum possible voidage in the bubble-free region, suggesting, as expected, that the mechanical strength of the structure drops to zero at \( U = U_{MB} \). In a variant of this method, using a shallow tilting bed at gas velocities just below \( U_{MF} \), Rietema (1984) and Piepers et al (1984) determined the "cohesion constant", or shear strength at zero normal stress. (For a Mohr-Coulomb material, \( \tau = \mu \sigma + c \), where \( c \) is the cohesion constant.) Experiments were carried out using packed beds
Figure 4.14 Bed Height, $H$, and Maximum Tilting Angle, $\alpha$, Versus Superficial Gas Velocity, $U$, for Cracking Catalyst 0-160µm (after Rietema and Mutsers, 1973)
of different materials, typical values ranging from 0 to 10 N/m². The method of analysis of this experiment is considered further in Chapter 5. It is essential to use a shallow bed, so that the stresses only vary in the direction normal to the distributor. Unfortunately, this method is not suitable for measuring the cohesion of an expanded fluidised bed, due to the very small magnitude of the strength and the experimental difficulty of maintaining uniform gas distribution in a very shallow bed (Oltrogge, 1972). If the gas velocity is above $U_{MF}$, therefore, it is necessary to use a deeper bed, for which invariance of stress cannot be assumed for any direction. Analytical interpretation then becomes impractical and the experiment is useful only for establishing whether or not mechanical strength exists, as in the experiments of Rietema and Mutsers (1973) referred to above. Piepers et al (1984) carried out experiments using both the shallow tilting bed at $U < U_{MF}$ and the deep tilting bed for $U > U_{MF}$ in order to examine the effect of gas pressure and type on the cohesion and fluidisation behaviour of cracking catalyst. They showed that for each of the three gases used (hydrogen, nitrogen, and argon) increase in pressure resulted in increase in cohesion of a settled bed (at the same voidage), which was accompanied by an increased maximum expansion for the homogeneously fluidised bed and an increased value for $U_{MB}$. The results were strongly affected by the type of gas used; independent measurements suggested that this was due to the different levels of gas adsorption on the solid surface.

4.2.3 Liquid Layers

Small amounts of free surface liquid on particle surfaces can give rise to interparticle forces which can be much larger than those arising from van der Waals forces alone (see Figure 4.8). The effects of
liquid addition to a gas-solid fluidised bed are often very marked, and of considerable practical importance. Fluidisation of moist material is undertaken in fluidised bed drying and cooling processes, while liquids are deliberately added to fluidised beds in coating and agglomeration processes (Nienow and Rowe, 1985), in the cultivation of micro-organisms (Zimmermann et al, 1984; Egerer et al, 1985) and, as here, as a retention aid in fluidised bed filters. Table 4.3 lists the experimental investigations of the effects of liquid addition to fluidised beds.

D'Amore et al (1979) investigated the effect of very low moisture levels on the fluidisation behaviour of group A powders. Not surprisingly, they found that the effect of moisture on non-porous glass, sand and limestone was much more marked than on porous catalysts because of the take-up of water within the porous structure. At low values of bed moisture a slight reduction in the settled bed voidage was observed for non-porous particles, having little effect on the minimum fluidisation velocity. Geldart and Wong (1984) showed that addition of moisture to group A particles decreases bed expansion at low gas velocities but increases it at higher velocities. This is explained by an increase in interparticle forces resisting expansion at low values of hydrodynamic drag but giving the bed greater mechanical strength, with the possibility of higher voidage, at larger gas velocities.

Numerous authors have noted the increase in minimum fluidisation velocity with addition of liquid to particles in group B. Parker and Stevens (1959) added water to glass spheres between 330 and 890 μm in diameter, fluidised with moist air, and noted not only an increase in minimum fluidisation velocity but also a decrease in the height of the
Table 4.3 Effects of Liquid Addition on Fluidisation Properties

<table>
<thead>
<tr>
<th>Reference</th>
<th>Particles</th>
<th>Group</th>
<th>Liquid</th>
<th>Gas</th>
<th>Observations</th>
</tr>
</thead>
<tbody>
<tr>
<td>D'Amore et al (1979)</td>
<td>Glass, Sand, Limestone, Cracking catalyst</td>
<td>A</td>
<td>Water</td>
<td>Moist Air</td>
<td>Very low levels of moisture addition. Little effect on porous catalyst; slight reduction in $e_s$ for others.</td>
</tr>
<tr>
<td>Parker and Stevens (1959)</td>
<td>Glass</td>
<td>B</td>
<td>Water</td>
<td>Moist Air</td>
<td>Increase in $U_{MF}$.</td>
</tr>
<tr>
<td>Baerns (1966)</td>
<td>Nickel</td>
<td>B</td>
<td>Vacuum oil</td>
<td>Air</td>
<td>Increase in $U_{MF}$, $e_{MF}$ and interparticle force.</td>
</tr>
<tr>
<td>Vaneczek et al (1967)</td>
<td>Sand</td>
<td>B</td>
<td>Water</td>
<td>Air</td>
<td>Increase in $U_{MF}$, (reduced by addition of fines)</td>
</tr>
<tr>
<td>Zimmermann et al (1984); Egerer et al (1985)</td>
<td>Yeast</td>
<td>D</td>
<td>Water</td>
<td>Air</td>
<td>Increase in $U_{MF}$ and $e_{MF}$. (reduced by addition of fines)</td>
</tr>
<tr>
<td>Gluckman et al (1976)</td>
<td>Cracking catalyst</td>
<td>D</td>
<td>Plastic</td>
<td>Air</td>
<td>Increase in $U_{MF}$</td>
</tr>
<tr>
<td>Maroglou and Nienow (1985)</td>
<td>Glass</td>
<td>B</td>
<td>Water</td>
<td>Air</td>
<td>Continuous system, with evaporation. $U_{MF}$ increases with liquid feed rate and decreases with temperature.</td>
</tr>
<tr>
<td>Tardos et al (1985)</td>
<td>Cracking catalyst</td>
<td>D</td>
<td>Plastic</td>
<td>Air</td>
<td>Increase in $U_{MF}$</td>
</tr>
</tbody>
</table>
fluidised bed, though it is not clear what height was being measured. They attempted to explain this in terms of a gradual transition from particulate to aggregative fluidisation. Baerns (1966) added a thin film of vacuum oil to the surface of 137 \( \mu \)m nickel spheres and noted an increase in minimum fluidisation velocity, settled bed voidage and interparticle force (measured using the tilting plate method; see Section 4.2.2). His results are presented in Figure 4.15.

![Figure 4.15 Relationship Between Modified Minimum Fluidisation Velocity, \( U_{MF}' \), Settled Bed Voidage, \( \varepsilon_S \), and Interparticle Force/Weight Ratio, \( F_p/G_p \) for 137 \( \mu \)m Nickel Spheres Covered in Vacuum Oil (data from Baerns, 1966).](image)

Vanecek et al. (1967) studied the relationship between minimum fluidisation velocity and moisture content of sand in connection with a fluidised bed cooling process in which the cooling effect was produced by evaporation of water from within the bed. A substantial increase in minimum fluidisation velocity was found for all sands used, even for moisture contents below 5% of bed weight (Figure 4.16). In all the cases listed above, the increase in total weight of the bed due to the liquid addition was small (negligibly so in the experiments of Baerns, 1966), so that the observed increase in minimum fluidisation velocity must have been due to the formation of a looser bed structure, of
reduced porosity. The minimum fluidisation velocity must then be increased to provide the pressure drop necessary to fluidise the bed. The increase in interparticle force enables this looser bed structure to remain stable.

Zimmermann et al (1984) and Egerer et al (1985) studied the fluidisation behaviour of wet cylindrical yeast particles, initially 0.8 mm in diameter and 2.3 mm long. This is a complex system, since addition of water alters not only the interparticle forces and hence bed voidage, but also causes appreciable changes in the particle size and density. With reference to the work of the author (Seville and Clift, 1984) they showed that the principal effect was the increase in bed voidage (Figure 4.17). When a flow conditioner (fine silica particles) was added to the bed, both the voidage at minimum fluidisation and the minimum fluidisation velocity were much reduced. Similar effects were noted by Vanecek et al (1967) when fine bentonite and coal dust were added to moist sand (Figure 4.16). The explanation lies in the change of contact radius, as discussed in Section 4.2.1.2.

If the quantity of free surface liquid is sufficient, the effective contact radius (i.e. that which determines the liquid bridge strength) is the macroscopic particle radius, because micron and sub-micron sized surface asperities are completely enveloped. However, fines tens of microns in diameter may not be completely enveloped, in which case they can determine (and reduce) the liquid bridge strength.

In agglomeration technology, the defluidisation of the bed due to excessive liquid content or maldistribution has been termed "wet quenching" (Nienow and Rowe, 1985). This phenomenon was investigated by Gluckman et al (1976) who added a plastic coating compound to cracking catalysts, fluidised at a certain gas velocity, until
Figure 4.16 Relationship Between Minimum Fluidisation Velocity, $U_{mf}$, and Moisture Content, for Wet Sand (after Vanček et al., 1967)
A. 150-400 μm
B. 200-600 μm
C. 100-1800 μm
D. 200-700 μm, with admixture of bentonite and coal dust.

Figure 4.17a Relationship Between Minimum Fluidisation Velocity, $U_{mf}$, and Moisture Content, for Yeast Particles (0.8 mm dia. x 2.3 mm long) (after Zimmermann et al., 1984)

Figure 4.17b Relationship Between Voidage at Minimum Fluidisation, $\epsilon_{mf}$, and Moisture Content, for Yeast Particles (0.8 mm dia. x 2.3 mm long) (after Zimmermann et al., 1984)
defluidisation occurred. A very sharp distinction between the fluidised and the unfluidised states was observed, as shown in Figure 4.18, although it is worth noting that the liquid used was very viscous and dropped directly into the bed rather than being finely dispersed so the particle coating would not have been uniform.

![Figure 4.18](image)

Tardos et al (1985) carried out similar experiments and compared their results with those obtained from a theoretical prediction of the minimum gas velocity required to break up an agglomerated bed with a given liquid content. The theoretical analysis contains a large number of unjustified assumptions, some of which are discussed further in Section 4.2.4, and the numerical agreement between theory and experiment is poor. However, the experimental data on gas velocity at quenching do show the predicted dependence on volume of liquid added and on particle size.
\[
\frac{U_q - U_{MF}}{U_{MF}} \propto \frac{V_M}{d_p^{3/2}} \tag{4.11}
\]

where \(U_q\) is the superficial gas velocity at the quenching point, \(U_{MF}\) is the minimum fluidisation velocity of the "clean" material and \(V_M\) is the volume of liquid added to the bed. Further experiment is required to confirm this relationship. In a system in which evaporation and drying also take place, as in most agglomerators, the minimum velocity at which fluidisation is possible depends on both the liquid feed rate and the bed temperature, an increase in bed temperature allowing operation to occur at lower gas velocities. An example of this effect is shown in Figure 4.19, from the work of Maroglou and Nienow (1985). When the liquid loading is too high, defluidisation can also occur in fluidised bed filters, especially when used to collect liquid droplets (Meissner and Mickley, 1949; Doganoglu, 1975; Tan 1982).

![Figure 4.19](image-url)
In summary, the addition of liquid to a fluidised bed causes an increase in interparticle forces which gives rise to an increase in the minimum fluidisation velocity. If sufficient liquid is added, a material in group B or A of Geldart's (1973) classification is effectively transformed into a group C material, and defluidisation occurs.

4.2.4 High Temperature Forces: Sintering

Operation of fluidised beds at elevated temperatures is frequently limited by the tendency of the particles to agglomerate, often catastrophically, leading to complete defluidisation. The temperature at which this can occur is in most cases well below the melting or softening temperature of the bed material and is due to the increased rate of sintering at high temperature. The phenomenon of high temperature defluidisation has been particularly troublesome in the development of several industrial processes, including coal gasification and combustion, iron ore reduction and cement production. It is worth mentioning, however, that there are a number of cases (some of which are discussed later) in which the tendency to sintering at high temperatures has been used to advantage in industrial processes.

Langston and Stephens (1960), in a much quoted paper, considered the development of a process for direct reduction of iron oxide using hydrogen. The ore must first be ground to small size in order to liberate the iron values. Langston and Stephens proposed to amalgamate the subsequent reduction and agglomeration steps by introducing the fine (<45μm) ore to a fluidised bed of spherical iron pellets (425–840 μm) operating at or around the sintering temperature of the iron (760°C). It was not possible to operate the bed directly on the
fine ore, because of the greater tendency for fine particles to
defluidise at low gas velocities or be elutriated at large ones. The
proposed process has a number of advantages: it operates at a
sufficiently high gas velocity (typically greater than 3m/s) that
defluidisation at high temperature is avoided and heat transfer is
promoted; the fine ore is not elutriated because it sticks to the
surface of the bed particles; and reduction is rapid since the ore is
deposited only on the surface. From their investigation of this
process, Langston and Stephens suggested that the "tendency to stick"
on particle–particle impact was directly proportional to the adhesive
force and area of contact, and inversely proportional to the particle
momentum. None of these terms was defined precisely. Their arguments
agree qualitatively with the observed tendency towards defluidisation
of smaller particles at lower gas velocities and higher temperatures.

Gluckman et al (1976) investigated the defluidisation at high
temperatures of group B copper, polymer, and glass particles. As the
temperature was gradually increased they observed a sudden drop in the
pressure difference across the bed (Figure 4.20), accompanied by the
onset of a more rapid rise in bed temperature as local "hot spots"
developed. For each of the particle size fractions investigated, an
operating diagram such as Figure 4.21 was plotted, defining regimes of
fluidisation and de-fluidisation in the gas velocity/temperature plane.
(The boundaries were observed to be sharp and reproducible). In all
cases it was observed that the temperature at which the measured
defluidisation velocity departed from the predicted behaviour in the
absence of interparticle forces coincided with the "initial sintering
temperature", Ts, as measured in a dilatometer. Siegell (1984)
confirmed that for T>Ts the modified minimum fluidisation velocity
increases linearly with T and observed that if the bed does become
Figure 4.20 Defluidisation Due to Sintering - Variation of Total Bed Pressure Drop and Bed Temperature During Quenching (after Gluckman et al., 1976).

Figure 4.21 High Temperature Defluidisation - Operating Region Diagram and Corresponding Dilatometer Trace (after Gluckman et al., 1976).
defluidised, normal fluidisation may be recovered by increasing the gas velocity, provided that the bed has not been allowed to remain defluidised for too long. This is because sintering occurs in stages, the initial, usually diffusional, stage leading to weaker bonds which are easily broken. Both Gluckman et al and Siegell report a linear decrease in the temperature required to defluidise the bed with increase in the bed height/diameter ratio. However, both operated with beds of rather small diameter (5cm in the case of Gluckman et al) so that slugging would have been likely, and their results cannot be extended with confidence to fluidised beds of much larger diameter.

In coal gasification and combustion systems there are considerable advantages in operating at as high a temperature as possible. However, conventional fluidised bed gasifiers are limited to around 1100°C by the tendency of the ash to form clinker and to defluidise. The agglomerating tendency of the ash is, however, exploited in the Ignifluid gasifier, described by Godel and Cosar (1967) and Yerushalmi et al (1975), which operates at 1200-1400°C and a gas velocity of 9-15 m/s. In this process, ash forms small agglomerates throughout the bed which can be removed in a controlled manner using a moving chain distributor. Caking coals, such as those widely used in the USA, present special fluidisation problems in both gasification and combustion systems.

Goldberger (1967) and Ehrlich et al (1972) present operating regime diagrams similar to Figure 4.21 for industrial coal combustion systems. However, these are non-linear, the increase in gas velocity required to maintain fluidisation increasing more rapidly at higher temperatures. In laboratory experiments on coal ash Basu (1982) found a linear
relationship, with the point of departure from the predicted minimum fluidisation velocity again coinciding with the initial sintering temperature.

Basu (1982) and Liss et al (1983) both attempted to predict the minimum fluidisation velocity of particles in the presence of interparticle forces, by adding a term due to interparticle forces directly to the overall force balance at minimum fluidisation. This clearly violates Newton's laws of motion, since at minimum fluidisation the pressure drop must equal the bed weight alone, by definition. Their theories will therefore not be considered further.

Yamazaki et al (1983) observed directly the increase in minimum fluidisation velocity and voidage at minimum fluidisation for four silica sand fractions (three in group B and one in group A). They found that Ergun's equation provided a good prediction of the minimum fluidisation velocity, if the correct voidage was used. They also measured the increase in interparticle force as a function of temperature, for 10 μm sand, using a split-cell method. The "adhesiveness" of the particles, $A_d$, was then defined as:

$$ A_d = \frac{F}{mg} \tag{4.12} $$

where $F$ = interparticle force (assumed proportional to $d_p$), and $mg$ = single particle weight.

Thus,

$$ A_d = \frac{k'f(T)d_p}{\eta d_p^3} = \frac{A_d}{\eta d_p^3} \tag{4.13} $$
where $f(T)$ is the measured interparticle force divided by its value at $T = 0°C$ (for 10 μm particles) and $k, k'$ are (dimensional) constants. Yamazaki et al. then successfully correlated the voidage at minimum fluidisation with the dimensional group $f(T)/d_p^2$, as shown in Figure 4.22. The analysis is unconvincing, since it involves extrapolating values for interparticle forces measured on very small particles to predict the behaviour of much larger ones. The interparticle force is assumed to scale with $d_p$, which, as observed earlier, is a dangerous assumption for real particles with surface asperities.

![Figure 4.22 Voidage at Minimum Fluidisation, $\varepsilon_{MF}$, as a Function of $f(T)/d_p^2$ (see equation (4.13); after Yamazaki et al., 1983).](image)

Sintering in high temperature fluidised beds has been studied extensively by Tardos and co-workers, at City College, New York. Siegell (1984), Compo et al. (1984) and Tardos et al. (1994) showed that the minimum sintering temperature, $T_s$, of a particulate material could be determined without fluidisation experiments by studying the contraction-dilation characteristics of a small sample in a
dilatometer. Compo et al (1987) studied the sintering and defluidisation behaviour of a range of materials and observed that although the bed pressure drop usually decreased on defluidisation, this was not always the case. They concluded that monitoring the power spectrum of the bed pressure drop gave a better indication of incipient defluidisation. In general they found that amorphous materials (glass, coal, polyethylene) agglomerated rapidly if $T_s$ was exceeded and that the gas velocity required to keep them fluidised at temperatures above $T_s$ increased exponentially with increase in bed temperature. These materials formed strong agglomerates. In contrast, crystalline materials (sodium chloride, calcium chloride, sodium citrate) agglomerated weakly and formed weak agglomerates. The gas velocity required to maintain fluidisation of these materials above $T_s$ was roughly proportional to their temperature.

Tardos et al (1985) attempted to construct models to enable the prediction of fluidisation limits for both ambient temperature beds containing free surface liquids, and high temperature beds above the sintering point. They argued that a complete description of the defluidisation phenomenon would take into account the dynamics of agglomerate formation and break-up so as to allow for fluidisation to occur even though agglomeration is taking place (which is obviously the case in fluidised bed agglomerators, as described in the previous section). However, their model takes a rather more simplistic approach, in an attempt to calculate the superficial gas velocity required to break the largest agglomerate which could be present in the bed, i.e. one which occupies the entire bed cross-section. To do this, the experimentally determined force measurements on a cylinder within a fluidised bed are extrapolated to the point where the agglomerate reaches the bed diameter (which is clearly unrealistic) and the
agglomerate strength is determined from Rumpf's (1962) theory (see Section 5.2.2.1). For sintering particles, the limiting superficial gas velocity, \( U_g \), is then given by:

\[
\frac{U_g - U_{MF}}{U_{MF}} = 3.3 \left[ 1 - \frac{1}{e^2} \right] \left[ \frac{d_B^{1.25} \sigma_s^{0.5}}{d_p^{1.5}} \right] \left[ \frac{\rho_p \sigma_s}{\rho_p \sigma_s^{0.5} \mu_s} \right]^{0.5}
\] (4.14)

where \( d_B \) is the bed diameter (m),

\( d_B \) is a typical bubble diameter at the level of the agglomerate (taken as \( d_B/4 \)),

\( \sigma_s \) is the yield strength of the sinter neck at each contact point in the agglomerate (N/m²), and

\( \mu_s \) is the granule surface viscosity (kg/ms).

The surface viscosity, \( \mu_s \), can be measured by means of a dilatometer, and the yield strength, \( \sigma_s \), can be calculated from Krupp's (1967) equation for the "van der Waals pressure":

\[
\sigma_s = \frac{\hbar w}{9n^2a^3}
\] (4.15)

where \( \hbar w \) is the Lifshitz constant (see Appendix A2) and \( a \) is the surface separation. (Equation (4.15) actually applies to the attraction between two parallel surfaces; it would be more correct to use the sphere-sphere form.) The surface separation is taken as an adjustable parameter, which may not be permissible. Assuming that the agglomerate voidage, \( \varepsilon \), remains constant as the temperature increases, equation (4.14) gives a reasonable fit to the experimental defluidisation temperature data for two particle sizes, plotted in the
form \((U_b - U_{mf})/U_{mf}\) versus \((T - T_g)/T_g\), with the separation, \(a\), set to \(6\Delta\). However, it may be remarked that only two terms in equation (4.14), the gas density, \(\rho_g\), and the surface viscosity, \(\mu_s\), are functions of temperature, and that the surface viscosity must be determined experimentally, so that equation (4.14) does not represent a completely theoretical prediction of \(U_b\). Furthermore, equation (4.14) remains untested against different bed diameters, and its derivation contains a number of assumptions and simplifications of doubtful validity, so that this theory must be considered as only a first attempt at the prediction of defluidisation velocity.

4.2.5 Electrical Forces

4.2.5.1 Electrostatic

Electrostatic forces are known to increase the minimum fluidisation velocity (see, for example, Lewis et al, 1949) but there appears to have been no systematic study of this effect.

4.2.5.2 Electromagnetic Forces

In Sections 4.2.3 and 4.2.4 the addition of liquid to a bed of group B material or an increase in temperature was shown to have a potentially catastrophic effect on the fluidisation behaviour, effectively transforming it to group C. Agbim et al (1971) showed, in a most significant experiment, that permanent magnetisation of particles could produce a more subtle effect, transforming group B type behaviour to group A type. Samples of steel shot of mean diameters 125 and 280 \(\mu m\)
were accidentally magnetised as a result of recovering the material using a magnet. A distinct separation was observed between the minimum fluidisation velocity and the minimum bubbling velocity, a separation which was not observed for a third, unmagnetised fraction, of mean diameter 194 μm. Bed height measurements showed a range of bubble-free expansion as expected for a group A material. Unfortunately, it was not possible to measure the induced interparticle force directly.

The control of the fluidisation of magnetisable materials and mixtures of magnetisable and non-magnetisable materials is now a large subject, which it would not be appropriate to review here because it relies on external imposition of body forces on the particles. Application of a magnetic field to a fluidised bed of ferromagnetic particles can result in a multiplicity of operating regimes, depending on gas velocity, field strength and field direction. Figure 4.23a is an operating region diagram for 177-250 μm steel spheres magnetised colinearly with the gas flow (Rosensweig et al, 1981). At higher values of the applied field a non-bubbling fluidised state exists above the minimum fluidising velocity, with similar properties to those of the expanded non-bubbling group A state. The transition between this state and conventional bubbling behaviour in the absence of the field is clearly defined and reproducible. The difference between this form of interparticle force and the others considered in this chapter appears to be that while the field can stabilise a structure of high voidage, allowing bubble free expansion to occur, there appears to be no effect on the minimum fluidisation velocity (Figure 4.23b).
Figure 4.23a Operating Region Diagram for 177-250 μm Steel Spheres in a Magnetic Field (field colinear with gas flow direction; after Rosensweig et al, 1981).

Figure 4.23b Pressure Drop Behaviour of Magnetically Stabilised and Unstabilised Fluidised Beds for 177 - 250μm Steel Spheres (field colinear with gas flow direction; after Rosensweig et al, 1981)
(Note that $U_T$ is equivalent to $U_{MB}$ for a group A material.)
4.3. Criteria for the Transitions in Fluidisation Behaviour

Since we are concerned here with the effect of interparticle forces on fluidisation behaviour, the transition between Geldart's groups B and D, which depends on fluid and particle inertial effects only, is not of interest. On the other hand, the transition between groups C and B is clearly influenced by the presence of interparticle forces, as was shown qualitatively by the work summarised in Section 4.2. This transition has received little theoretical attention in the past, but the transition between groups A and B has, however, been extensively investigated, both in terms of interparticle forces and without resource to them. Some of these investigations, and the theoretical expressions for the transition which result from them are briefly reviewed below.

Attempts to describe the bubbling/non-bubbling transition in purely hydrodynamic terms (see, for example, Foscolo and Gibilaro, 1984) will not be considered here, because of the ample experimental evidence that interparticle forces are important in this behaviour. Hydrodynamic models are capable of giving good prediction of the range of stability of liquid-solid fluidised beds, but, as already discussed (Martin, 1983), the comparisons between gas-solid group A behaviour and liquid-solid particulate fluidisation are somewhat misleading. Purely hydrodynamic models are likely to be successful only for particles in Martin's group E and are not applicable to group A. Grace (1986) has compared the predictions of Foscolo and Gibilaro (1984) with experimental data for the A-B transition over a wide range of solid and fluid density without finding satisfactory agreement (Figure 4.24).
Figure 4.24 Classification of Fluidisation Behaviour in Terms of Dimensionless Density Difference Between Particles and Fluid and Dimensionless Particle Size (including data for gases other than air and high temperatures and pressures; after Grace, 1986).

\[ \frac{\Delta \rho}{\rho} = \frac{\rho_{g} \Delta \rho}{\mu^{2}} \]

\[ \Delta \rho = \rho_{p} - \rho_{g} \]
4.3.1 Geldart (1973)

As noted earlier, Geldart (1973) defined group A powders as those for which $U_{MB}/U_{MF} > 1$. Correlations for $U_{MF}$ are well established; Geldart correlated the available data on $U_{MB}$ and obtained the simple relationship

$$U_{MB} = K_{MB} d_p$$  \hspace{2cm} (4.16)

where $K_{MB}$ is a constant (~ 100 s$^{-1}$) and $d_p$ is the surface-volume mean particle size (m).

The criterion for the A/B transition is then given by

$$\frac{U_{MF}}{U_{MB}} = \frac{U_{MF}}{K_{MB} d_p} = 1$$  \hspace{2cm} (4.17)

Geldart originally used Davies and Richardson's (1966) expression for $U_{MF}$. Geldart and Abrahamson (1978) subsequently used their own correlation for $U_{MF}$, which, when substituted into equation (4.17) gives the transition condition:

$$\frac{2300 \rho_g 0.126 \mu 0.523 \exp(0.716F)}{d_p 0.8 0.934 (\rho_s - \rho_g) 0.934} = 1$$  \hspace{2cm} (4.18)

where $F = \text{the weight fraction of the solids with } d_p < 45 \mu m$. This expression has generally been found to be reliable, even at high pressure (King and Harrison, 1982). For air, at ambient temperature, it reduces to:

$$d_p^{0.857} (\rho_p - \rho_g) = 0.93$$  \hspace{2cm} (4.19)

This equation is plotted in Figure 4.25.
Molerus (1982) suggested that the differences between groups A, B and C arise from the value of the ratio of the interparticle force to the drag of the fluidising gas, and developed semi-empirical conditions which confirmed Geldart's group boundaries. He defined group B type behaviour as that for which interparticle forces are negligible by comparison with fluid drag. By definition, in a fluidised bed the drag on the particle must counterbalance its immersed weight. Therefore, as a simple expression for the B/A boundary, Molerus postulates:

\[
\frac{\pi d_p^3}{6} \left( \rho_p - \rho_g \right) \frac{g}{F_h} = \text{constant}, K \tag{4.20}
\]

where \(F_h\) is the adhesion force transmitted in a single contact between two adjacent particles. Molerus then considers the case of 100 \(\mu m\) glass ballotini, of density \(2.5 \times 10^3 \text{ kg/m}^3\), which lie on the border between the groups. Using equation (A2.5), with a characteristic asperity radius of 0.1 \(\mu m\), he obtained a value for \(F_h\) due to van der Waals forces alone of \(8.8 \times 10^{-8} \text{N}\), yielding \(K = 0.16\). (Molerus' value; an exact calculation gives \(K = 0.147\)). The condition resulting from equation (4.20) is plotted in Figure 4.25 as a hatched region, the limits representing the transitions to be expected for "hard" and "soft" materials. For "soft" materials group A extends further to the right on the diagram.

Although equation (4.20) adequately fits the data (which covers a limited range of particle densities) this does not constitute confirmation of Molerus' calculated values for \(F_h\) arising from van der Waals forces alone, since \(F_h\) and \(K\) are not independent; the transition
has been forced to fit the case of 100 μm glass ballotini. In fact there is some doubt about the value to be used for $F_H$. Appendix A2 reviews the methods for calculation of interparticle attractions due to van der Waals forces, and the values of $F_H$ and $K$ in equation (4.20) are considered further in Chapter 5.

4.3.3 Rietema and Co-Workers

The theoretical fluid mechanical stability of the state of uniform fluidisation has been considered by a number of workers, the earliest analysis being that of Jackson (1963). In the analysis that follows it is assumed that variables such as velocity and stress are to be averaged over regions large compared with the particle spacing but small compared with the scale of the motions of interest, and that the gas phase is incompressible.

The two continuity equations and the two momentum equations for the bed are given in the general form (Jackson, 1985) as:

**Fluid Continuity**
\[
\frac{\partial \rho}{\partial t} + \text{div} \left( \rho \mathbf{u} \right) = 0 \tag{4.21}
\]

**Particle Continuity**
\[
\frac{\partial}{\partial t} \left( 1 - \epsilon \right) + \text{div} \left[ \left( 1 - \epsilon \right) \mathbf{v} \right] = 0 \tag{4.22}
\]

**Fluid Momentum**
\[
\rho \mathbf{g} \times \left[ \frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \times \text{grad} \mathbf{u} \right] = \epsilon \mathbf{v} \times \mathbf{e}_g - \epsilon \mathbf{F} + \epsilon \rho g \mathbf{g} \tag{4.23}
\]

**Particle Momentum**
\[
\rho_s (1 - \epsilon) \left[ \frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \times \text{grad} \mathbf{v} \right] = \left[ (1 - \epsilon) \mathbf{v} \times \mathbf{e}_g + \epsilon \mathbf{F} \right] + (1 - \epsilon) \rho_g \mathbf{g} + \mathbf{v} \times \mathbf{e}_g \tag{4.24}
\]
where \( \bar{u}, \bar{v} \) = fluid and particle local average velocities (thus \( u = U/E \), where \( U \) is the superficial gas velocity);

\( \bar{F} \) = local average drag force exerted on the particles by the fluid per unit bed volume, in the absence of gravity;

\( \bar{\sigma}_g, \bar{\sigma}_s \) = stress tensors associated with the fluid and particle phases respectively, in which, following Jackson (1971), the normal stress components are defined to be negative in compression (thus \( \nabla \cdot \bar{\sigma}_g = -\partial p/\partial x \));

\( \bar{g} \) = specific gravity force vector = \(-g\hat{i}\) where \( \hat{i} \) is the unit vector, positive upwards.

Thus, the terms on the right hand side of the momentum balances represent respectively the buoyancy force exerted by the fluid on the particles, the hydrodynamic drag forces exerted by the fluid on the particles, the gravitational force, and, in the case of the particles only, any interparticle force contribution. The formulations for the expressions for \( \bar{F}, \bar{\sigma}_g \) and \( \bar{\sigma}_s \) in the momentum equations differ widely between different workers. The approach generally taken is to replace \( \bar{F} \) with a term depending on the void fraction and the relative velocity of the phases:

\[
\bar{F} = \beta(\varepsilon, |\bar{u} - \bar{v}|) (\bar{u} - \bar{v}) \quad (4.25)
\]

(At low Reynolds numbers \( \beta \) is a function of \( \varepsilon \) only.)

The analysis of the different approaches to the detailed formulation of the momentum balances is beyond the scope of this thesis. In general, the procedure is now to apply linearised stability theory, introducing infinitesimally small perturbations and subtracting the steady state solution. When this is done it has been generally found that all
fluidised beds are predicted to be unstable to voidage perturbations, liquid and gas-fluidised beds differing only in the rate at which the disturbances grow. Disturbances in a gas-fluidised bed grow approximately one hundred times as fast as in a liquid-fluidised bed. This explains why gas fluidised beds generally exhibit bubbling behaviour while liquid fluidised beds do not, but it is at variance with the observed stability of group A material at gas velocities between $U_{mf}$ and $U_{mb}$.

In most early work on the stability of uniform fluidisation, the solids stress tensor $\tau_s$ was taken as zero i.e. interparticle cohesive forces were implicitly neglected. However, it can be shown that if this term is sufficiently large, the stability is predicted at low gas velocities for all wavelengths of disturbance. The analysis is that of Rietema and co-workers (Rietema and Mutsers, 1973, Mutsers and Rietema, 1977).

Re-writing the continuity equations considering only the vertical components, which are taken as positive upwards, we have:

$$\frac{\partial s}{\partial t} + \frac{\partial (us)}{\partial h} = 0$$  \hspace{1cm} (4.21')

$$\frac{\partial s}{\partial t} - \frac{\partial [v(1 - e)]}{\partial h} = 0$$  \hspace{1cm} (4.22')

where $u$, $v$ are the fluid and particle local average velocities in the $h$ (vertical) direction.

Considering a gas-fluidised system and neglecting all terms in $\rho_g$ in equation (4.23) and taking a low value of Reynolds number, the fluid momentum equation becomes:
where \( F = \beta(e) (u-v) \) \( (4.25') \)

and \( p \) is the manometric pressure (see equation (4.2))

Note that Rietema and Hutser use the opposite sign convention for \( F \); the convention here is as used by Jackson.

The particle momentum equation now becomes:

\[
\rho_s (1 - e) \left[ \frac{\partial v}{\partial t} + v \frac{\partial v}{\partial x} \right] = \left[ -(1 - e) \frac{\partial p}{\partial h} + eF \right] - (1 - e) \rho_s g + (v \cdot \tau_s) h \]  

\( (4.24') \)

Combining (4.23'), (4.24') and (4.25') we have:

\[
\rho_s (1 - e) \left[ \frac{\partial v}{\partial t} + v \frac{\partial v}{\partial x} \right] = \beta(u-v) - (1 - e) \rho_s g + (v \cdot \tau_s) h \]  

\( (4.26) \)

Rietema now considers the term \((v \cdot \tau_s) h\) to be identical to \(-(v \cdot \tau_s) h\) where \(\tau_s\) is the stress tensor in the solid phase due to particle-particle interactions in the expanded cavity structure, now taken to be positive in compression. Now,

\[
(v \cdot \tau_s) h = \frac{\partial}{\partial h} T_{hh} + \frac{\partial}{\partial x} T_{xh} + \frac{\partial}{\partial y} T_{yh} \]  

\( (4.27) \)

where \( x \) and \( y \) are horizontal coordinates. The stresses are depicted in Figure 4.26. In a column of wide cross-section, only the vertical direction is of importance and

\[
\frac{\partial \tau_{yh}}{\partial y} = \frac{\partial \tau_{xh}}{\partial x} = 0 \]  

\( (4.28) \)
Rietema now postulates an "elasticity", $E$, of the expanded particulate structure, analogous to the elastic resistance of a gas to compression. (It is worth noting that interparticle collisions alone cannot generate a sufficiently large value for $E$ to stabilise the structure; the elasticity must arise from semi-permanent particle-particle contact.)

Hence:

$$\frac{\partial}{\partial n} T_{hh} = E \frac{\partial \varepsilon}{\partial n} = (\mathbf{v} \cdot \nabla) \varepsilon_h \quad (4.29)$$

From (4.26):

$$\rho_s (1-\epsilon) \left[ \frac{\partial v}{\partial t} + v \frac{\partial v}{\partial n} \right] = \beta(u-v) - (1-\epsilon) \rho_s g + E \frac{\partial \varepsilon}{\partial n} \quad (4.30)$$
In the equilibrium state
\[ \varepsilon = \varepsilon_0; \; \beta = \beta_0; \; \nu = 0 \]
\[ \varepsilon_{MF} < \varepsilon_0 < \varepsilon_{MB} \]
\[ \beta_e \; \nu_e - (1-\varepsilon_0) \rho g = 0 \]  
\hspace{1cm} (4.31)

Now impose linear perturbations:
\[ v = v'; \; u = u_e + u'; \; e = e_e + e'; \; \beta = \beta_e + \beta' \]

From (4.30); neglecting second order terms:
\[ \rho_g (1-\varepsilon_0) \frac{\partial v'}{\partial t} = (\beta_e + \beta') (u_e + u' - v') - (1-\varepsilon_e-\varepsilon') \rho g \]
\[ + E \frac{\partial \varepsilon'}{\partial h} \]  
\hspace{1cm} (4.32)

Substitute for \( \beta_e u_e \) from (4.31):
\[ \rho_g (1-\varepsilon_0) \frac{\partial v'}{\partial t} = \beta' u_e + \beta_e (u' - v') + e' \rho g + E \frac{\partial \varepsilon'}{\partial h} \]  
\hspace{1cm} (4.33)

Differentiating (4.33) with respect to \( h \):
\[ \rho_g (1-\varepsilon_0) \frac{\partial^2 v'}{\partial t \partial h} = u_e \frac{\partial \beta}{\partial \varepsilon} \frac{\partial \varepsilon'}{\partial h} + \beta_e \left[ \frac{\partial u'}{\partial h} - \frac{\partial v'}{\partial h} \right] + \rho g \frac{\partial \varepsilon'}{\partial h} + E \frac{\partial^2 \varepsilon'}{\partial h^2} \]  
\hspace{1cm} (4.34)

Applying perturbations to the continuity equations, (4.21') and
(4.22'), we have:
\[ \frac{\partial \varepsilon'}{\partial t} + u_e \frac{\partial \varepsilon'}{\partial h} + e_e \frac{\partial u'}{\partial h} = 0 \]  
\hspace{1cm} (4.35)
\[ \frac{\partial \varepsilon'}{\partial t} - (1-\varepsilon_0) \frac{\partial v'}{\partial h} = 0 \]  
\hspace{1cm} (4.36)

which can be re-arranged thus:
\[ \frac{\partial u'}{\partial h} = -\frac{1}{e_e} \left[ \frac{\partial e'}{\partial t} + u_e \frac{\partial e'}{\partial h} \right] \quad (4.35') \]

\[ \frac{\partial v'}{\partial h} = \frac{1}{1-e_e} \frac{\partial e'}{\partial t} \quad (4.36') \]

Also, differentiating (4.36') with respect to t we have:

\[ \frac{\partial^2 v'}{\partial h \partial t} = \frac{1}{1-e_e} \frac{\partial^2 e'}{\partial t^2} \quad (4.37) \]

Equations (4.35'), (4.36') and (4.37) can now be substituted into equation (4.34):

\[ \rho_s \frac{\partial^2 e'}{\partial t^2} = u_e \frac{\partial}{\partial e} \left[ \frac{1}{e_e} \left( \frac{\partial e'}{\partial t} + u_e \frac{\partial e'}{\partial h} \right) \right] - \frac{1}{1-e_e} \frac{\partial e'}{\partial t} \]

\[ + \rho_bg \frac{\partial e'}{\partial h} + E \frac{\partial^2 e'}{\partial h^2} \quad (4.38) \]

Therefore:

\[ \frac{\partial^2 e'}{\partial t^2} = \frac{E}{\rho_s} \frac{\partial^2 e'}{\partial h^2} + \frac{\partial e'}{\partial h} + \rho_g \frac{\partial e'}{\partial e} \]

\[ = g \frac{\partial e'}{\partial h} + u_e \frac{\partial}{\partial e} \frac{\partial e'}{\partial h} \quad (4.39) \]

Now substitute for g from equation (4.31):

\[ \frac{\partial^2 e'}{\partial t^2} = \frac{E}{\rho_s} \frac{\partial^2 e'}{\partial h^2} + \frac{\partial e'}{\partial h} + \rho_g \frac{\partial e'}{\partial e} \]

\[ = \frac{\rho_g u_e}{\rho_s (1-e_e)} \frac{\partial e'}{\partial h} + u_e \frac{\partial}{\partial e} \frac{\partial e'}{\partial h} \quad (4.40) \]
Therefore:

\[
\frac{\partial^2 \varepsilon'}{\partial t^2} - \frac{E}{\rho_s} \cdot \frac{\partial^2 \varepsilon'}{\partial h^2} + \frac{\beta}{\rho_s e_e (1-e_e)} \left[ \frac{\partial \varepsilon'}{\partial t} + u_e (1-2e_e) \frac{\partial \varepsilon'}{\partial h} \right] = 0
\]

or

\[
\frac{\partial^2 \varepsilon'}{\partial t^2} - V_d^2 \frac{\partial^2 \varepsilon'}{\partial h^2} + B \left[ \frac{\partial \varepsilon'}{\partial t} + V_w \frac{\partial \varepsilon'}{\partial h} \right] = 0
\]

where \( V_d^2 = E/\rho_s \)

\[
B = \frac{\beta}{\rho_s e_e (1-e_e)}
\]

\[
V_w = u_e (3-2e_e)
\]

It is now necessary to assume a functional form for \( \beta \). Since they were concerned only with low Reynolds numbers, Mutsers and Rietema (1977) took:

\[
\beta = 150 \frac{(1-e)^2}{e^2} \frac{\mu}{d_p^2}
\]
According to Wallis (1969), $V_d$ is the propagation velocity of a dynamic wave and $V_w$ the propagation velocity of a continuity wave. If $V_w > V_d$ then the perturbation is growing in amplitude and the bed is unstable. Thus the stability criterion is:

$$V_w \leq V_d \quad (4.46)$$

Inserting expressions for $V_w$ and $V_d$ we have:

$$u_e (3-2\varepsilon_e) < (E/\rho_g)^{1/2} \quad (4.47)$$

Substituting for $u_e$ from (4.31) and $\beta_e$ from equation (4.42):

$$\beta_e u_e = (1-\varepsilon_e) \rho_g g = \frac{150}{\varepsilon_e^2} \frac{\mu}{\sigma_p^2} u_e \quad (4.48)$$

Therefore, at the point of transition from stable to unstable behaviour, $E = E_{MB}$ and $\varepsilon_e = \varepsilon_{MB}$, so from (4.47) and (4.48):

$$\frac{\rho_s^3 \sigma_p^4 g^2}{\mu^2 \varepsilon_e^2} = \frac{150 (1-\varepsilon_e)^2}{\varepsilon_e^2 (3-2\varepsilon_e)} \quad (4.49)$$

In physical terms, the stability analysis predicts that the transition between non-bubbling and bubbling behaviour occurs when the velocity of a voidage fluctuation (the dynamic wave velocity, which depends here on the elasticity $E$) exceeds the velocity of an equilibrium disturbance (which is a function of the voidage, $\varepsilon$).
In their experiments, Rietema and co-workers applied equation (4.49) at the point of minimum bubbling in order to calculate the value of $E_{mb}$ as a function of $e_{MB}$. From their results, some of which are presented in Figure 4.27, they concluded that:

(i) $E_{mb}$ decreases exponentially with increase in $e_{MB}$, due to the reduction in the total number of contacts in any cross-section;

(ii) $E_{mb}$ is not greatly dependent on particle diameter but increases slightly with increase in the standard deviation of the size distribution;

(iii) $E_{mb}$ differs considerably for different materials.

Mutsers and Rietema's results are in fact entirely consistent with Geldart's equation (4.16), as will now be shown. For expansion we can apply the Richardson-Zaki (1954) equation

$$\frac{U}{U_t} = e^n$$  \hspace{1cm} (4.50)

where $U_t$ = particle terminal velocity in an infinite medium,
and $n = f(Re_p,t)$.

Here we shall use Richardson and Zaki's value for $n$ appropriate to $Re_p < 0.2$, although, as noted in Section 4.2.2, other workers have suggested that higher values are more applicable in the case of expansion of group A particles. Substituting for $U_t$ at low Reynolds numbers and making use of equation (4.16) we have:

$$e_{MB} = \left[ \frac{U_{MB}}{U_t} \right]^{1/n} = \left[ \frac{1800 \mu}{\rho_p g d_p} \right]^{1/4.65}$$  \hspace{1cm} (4.51)
Figure 4.27 Calculated Elasticity Coefficient, $E_{mb}$, As a Function of Voidage at Minimum Bubbling, For Fresh Cracking Catalyst (after Mutsers and Rietema, 1977).

Fluidisation With
- air
- hydrogen
- propane
- neon

$\sigma$ = standard deviation of size distribution
Figure 4.28 Maximum Expanded Bed Voidage, $\varepsilon_{\text{MAX}}$, Plotted Against the Reciprocal of Particle Diameter, $d_P$, and Against Gas Viscosity, $\mu$ (data from Mutters and Rietema, 1977).
Figure 4.28 presents Mutsers and Rietema's (1977) data for \( e_{MB} \) of cracking catalyst, plotted as a function of particle diameter (omitting wide size distributions) and, for a single mean particle size, as a function of gas viscosity. It can be seen that equation (4.51) provides a good fit to the measured data, which might be improved by adjusting the value of \( n \). Thus equation (4.51) provides a ready means of calculating \( e_{MB} \), and hence, from equation (4.49), the elasticity \( E \).

According to Molerus (1967), if the stability of the expanded non-bubbling bed has a hydrodynamic origin then \( e_{MB} \) should be a function of the Gallileo number, \( \rho g^2 d_p^3 g/\mu \). Rietema and Mutsers (1978) used a centrifuge to investigate the effect of changes in the gravity force on the voidage at minimum bubbling, reasoning that if the elasticity arose from hydrodynamic factors \( e_{MB} \) should be a function of \((g/\mu^2)\), whereas their earlier analysis would predict a dependence on \((g/\mu)^2\). The results showed a much better agreement with the latter hypothesis.

The physical basis of Rietema and Mutsers' ideas is now examined in more detail. Rietema (1973) attempted to relate the "elasticity" to calculated interparticle forces arising from van der Waals interactions, and Rietema and Mutsers (1975) attempted direct measurement using vibrating coils submerged within the bed. Neither analysis shows conclusive proof of elasticity, and the physical basis of the theory remains unproven. Elasticity is considered to arise from the rearrangement of particles without bond breakage. Figure 4.29 shows the idealised "zig-zag" structure of the bed. On the right is an undistorted bed and on the left a distorted one, with the voidage as a function of height represented alongside. The "string" of particles is envisaged to behave rather like a spring, with an elastic force.
(proportional to $\partial e/\partial h$) resisting compression and expansion. The exponential decrease in elasticity with voidage (Figure 4.27) is explained by the reduction in the total number of bonds with increase in voidage and the consequent reduction in rigidity.

Figure 4.29 Stylised Diagram of Distorted and Undistorted Particle "String" (after Rietema and Mutsers, 1973).

If the expanded structure is to show elastic behaviour in the way which is suggested then it must be possible for energy to be stored during expansion (and compression) and subsequently recovered from somewhere in the system. The only way in which this might occur is for energy to be stored at the particle-particle contacts, which will occur if the particles are themselves elastic. Consider van der Waals forces alone, although the argument which follows applies qualitatively to other interparticle forces. The attractive force between two rigid spheres
of radii $R_1$ and $R_2$ is given (Appendix A2) by:

$$F_w = \frac{AR}{6a^2} = 4\pi r y$$  \hspace{1cm} (4.52)

where $A$ is the Hamaker constant (J)

$$R = \frac{R_1 R_2}{(R_1 + R_2)} \text{ (m)}$$

$a$ is the inter-body spacing (m)

$\gamma$ is the surface energy of the material in vacuo (J/m$^2$)

As $a$ is reduced, $F_w$ increases until contact occurs, at a value of which is not accurately known but which is related to the interatomic spacing. Israelachvili (1985) recommends the use of $1.65\AA$ as the "cut-off" distance for $a$ and shows that when this value is put into equation (4.52) the resulting surface energies agree with those experimentally determined. Thus, the maximum value of $F_w$ can be obtained. If the spheres were perfectly rigid, that would be the end of the process, but for real materials, surface deformation, both plastic and elastic, will now occur. Plastic deformation cannot give rise to the required gross elasticity, but elastic deformation might, by generating a repulsive force to resist the van der Waals attraction. Eventually, equilibrium will be reached, and the two forces will balance. This situation is shown schematically in Figure 4.30. A similar equilibrium arises for certain particle/liquid systems where the repulsive force is provided by electrical double-layer interaction. It is postulated that at every stage in the expansion of the non-bubbling bed there is no net force transmitted at the contact points, each of which is in equilibrium (Verkooijen, 1986). A small disturbance of a contact will result in a restoring force which will act so as to return the particles to their original relative
Figure 4.30 Particle Interaction Energy, \( W \), and Force, \( F \), As Functions of Separation Distance, \( a \) - Idealised Behaviour for Elastic Particles.
separation. If there is more than one contact point per particle-particle interaction, this does not materially alter the argument; a position of net stable equilibrium can still be found. A structure bound in this way will show "bulk elasticity" over a very small range, provided that the deformation is partially or wholly elastic. Theoretically, it ought to be possible to calculate the order of magnitude of this bulk elasticity, at least for elastic spheres. According to Hertz (1896), as reported by Johnson et al (1971), when two elastic spheres of radii $R_1$ and $R_2$ come into contact under an applied load $F_R$, distant points within them come together by a distance $\delta$, given by:

$$\delta^3 = \frac{F_R^2}{K^2 R} \quad (4.53)$$

where

$$K = \frac{4}{3} \left[ \frac{1-\nu_1^2}{Y_1} + \frac{1-\nu_2^2}{Y_2} \right]$$

$\nu_1 = \text{Poisson's ratio for the material of sphere 1}$

$Y_1 = \text{Young's modulus for the material of sphere 1}$

$R = R_1 R_2 / (R_1 + R_2)$

Conversely, an imposed deformation of $\delta$ produces a repulsive elastic force, $F_R$. Assuming that the maximum attractive force due to van der Waals interactions is maintained at its value given by equation (4.52) after contact has occurred, we can now write an expression for the total interaction force:

$$F_H = 4\pi R y - \delta^{3/2} K^{1/2} R^{1/2} \quad (4.54)$$

which is positive in attraction. $F_H$ is plotted as a function of $\delta$ in Figure 4.31.
Figure 4.31 Particle Interaction Force, $F$, from Equation (4.54).
Equilibrium occurs when $F_H = 0$, i.e.

$$\frac{\sqrt{2}}{2} \sigma = \frac{4\pi \gamma R^{1/2}}{K} \quad (4.55)$$

Now Rietema's elasticity is defined according to equation (4.29), from which we obtain:

$$E_h = - \frac{\partial \tau_{hh}}{\partial \sigma} = - \frac{\partial \tau_{hh}}{\partial \sigma} . \frac{\partial \sigma}{\partial \sigma_h} \quad (4.56)$$

where the subscript denotes changes in the $h$ direction only.

According to Rumpf (1962) (see also Section 5.2.2.1), in a randomly packed bed of monosized spheres:

$$\tau_{hh} \approx \frac{9}{8} \epsilon \frac{F_H}{\sigma^2} \quad (4.57)$$

where $F_H$ is the force transmitted per particle–particle contact (not necessarily in the $h$ direction).

For a cubic structure expanded in the $h$ direction only:

$$\sigma_h = \frac{d_p^2 (d_p + \delta) - \frac{\pi d_p^3}{6}}{d_p^2 (d_p + \delta)} = 1 - \frac{\pi d_p}{6 (d_p + \delta)} \quad (4.58)$$

$$\therefore \frac{\partial \sigma_h}{\partial \delta} = - \frac{\pi d_p}{6 (d_p + \delta)^2} \approx \frac{\pi}{6 d_p} \text{ since } \delta \ll d_p \quad (4.59)$$

It is now necessary to evaluate $\partial \tau_{hh}/\partial \sigma$ in equation (4.56). To a first approximation, the expansion of the structure in the $h$ direction affects only the magnitude of $F_H$, since $\epsilon$ in the plane perpendicular to
the h direction is not changed by the expansion. Hence, we can write

\[ \frac{\partial \phi_{h}}{\partial \phi} \approx \frac{9}{8} \frac{1-\epsilon}{\epsilon} \frac{1}{\alpha} \frac{\partial \phi_{h}}{\partial \phi} \quad (4.60) \]

4.56

Hence, from (4.57), (4.59) and (4.60) we have:

\[ E_{h} = -\frac{6\alpha}{\pi} \cdot \frac{9}{8} \cdot \frac{1-\epsilon}{\epsilon} \cdot \frac{1}{\alpha} \frac{\partial \phi_{h}}{\partial \phi} \]

\[ = -\frac{27}{4\pi} \cdot \frac{1-\epsilon}{\epsilon} \cdot \frac{1}{\alpha} \frac{\partial \phi_{h}}{\partial \phi} \quad (4.61) \]

4.54

From (4.49):

\[ \frac{\partial \phi_{h}}{\partial \phi} = -\frac{3}{2} \cdot \frac{1}{2} \cdot \frac{1}{2} \cdot K R \frac{1}{2} \quad (4.62) \]

which when evaluated at the equilibrium point, using equation (4.55), gives:

\[ \left[ \frac{\partial \phi_{h}}{\partial \phi} \right]_{e} = -\frac{3}{2} K R \frac{1}{2} \left( \frac{4\pi R}{K} \right)^{1/2} \]

\[ = -\left( \frac{27}{2} \pi K^{2} R^{2} \gamma \right)^{1/3} \quad (4.63) \]

Substituting in equation (4.61) we have:

\[ E_{h} = \frac{27}{4\pi} \cdot \frac{1-\epsilon}{\epsilon} \cdot \frac{1}{\alpha} \left[ \frac{27\pi}{2} K^{2} R^{2} \gamma \right]^{1/3} \quad (4.64) \]

Now according to equation (4.53), \( R = \alpha / 4 \). Therefore,

\[ E_{h} = \frac{27}{4\pi} \cdot \frac{1-\epsilon}{\epsilon} \left[ \frac{27\pi}{32} \frac{K^{2}}{\alpha} \gamma \right]^{1/3} \quad (4.65) \]
Consider smooth quartz spheres of diameter 50 μm. Israelachvili (1985) gives the Hamaker constant for quartz as 6.5 x 10^{-20} J, which gives a surface energy of 0.031 J/m^2. Taking Young’s modulus for quartz as about 10^9 N/m^2 (Anon, 1971) and Poisson’s ratio as 0.4, K ≈ Y ≈ 10^9 N/m^2. These values give an equilibrium deformation, σ_e, of about 124%. Putting values for K, R, γ and dp into equation (4.64), and taking a typical value for εMB of 0.6 we have E_H ≈ 1.7 x 10^7 N/m^2 which is about eight orders of magnitude larger than Rietema’s experimental values.

If we now consider F_H to be determined by local asperities, equation (4.64) still holds, where R is now equal to the radius of the asperity, say 0.1 μm. Using this value, E_H is reduced to 6.8 x 10^5 N/m^2, which is still much larger than the experimentally determined values. Thus it appears unlikely that Rietema’s concept of elasticity can adequately explain the observed stability of the expanded non-bubbling bed.

In order to investigate the effect of interparticle forces on fluidisation it is necessary to have a direct experimental measure of the interparticle forces themselves. The next chapter deals with the development of a method to do this and the application of this method to a fluidised bed of cohesive group B particles.
5.1 Qualitative Effects of Liquid Addition.

When particles in a fluidised bed filter are coated with a layer of liquid retention aid there comes as point, as the loading of the coating is increased, at which the capillary forces between the coated particles exceed the mechanical forces in the bed (which, in general, will tend to separate the particles) and the bed will cease to fluidise. This process is known in agglomeration technology as "wet quenching", as distinct from "dry quenching" which is the result of the formation of solid bonds between particles as the liquid evaporates. Dry quenching leads to the formation of large solid agglomerates which segregate to the bottom of the bed unless the fluidising velocity is increased to compensate for this effect. Despite a large volume of literature on agglomeration, satisfactory general rules for the avoidance of wet and dry quenching have not yet been derived.

The problem of wet quenching is a potentially serious drawback in the industrial use of fluidised bed filtration with retention aid addition. At lower temperatures it would be desirable to use a light loading of a comparatively involatile liquid as the retention aid, so that all the particles in the bed are continuously surface-wet. This is a rather different mode of operation from that which is common in fluidised and spouted bed agglomeration, where usually only particles in the immediate vicinity of the spray nozzle will be appreciably wet, the remainder of the bed acting as a drying zone and particle reservoir. It is therefore of interest to determine the effect of a gradual increase
in the loading of an involatile liquid on the particle surfaces, introducing the liquid as uniformly as possible throughout the bed, rather than at one spray point only. Experiments were carried out in which a light involatile oil (di-2-ethyl hexyl sebacate) was added to the filter bed under ambient conditions. The objectives were to establish criteria for tolerable levels of liquid on the bed, and to identify simple measurements which can be made to indicate the onset of quenching.

Glass ballotini in narrow size ranges between 400 and 1000 μm were gradually loaded with sebacate by fluidising them in a 0.152 m diameter column with a flat perforated plate distributor. The oil was atomised continuously into the fluidising air using a Collison atomiser (B.S. 2577, 1955) so that it was collected onto the bed particles by the mechanisms described in Chapter 2. Small samples of the bed material were withdrawn at intervals during the loading process, and analysed for sebacate loading by washing with ethanol in an ultrasonically vibrated bath, and determining sebacate concentration in the resulting solution using a Perkin-Elmer Lambda 5 UV Spectrophotometer at a wavelength of 215 nm. The pressure drop across the bed was monitored until fluidisation ceased. Figure 5.1 shows the general behaviour of the pressure drop trace as the liquid loading increased, for one ballotini size fraction. No sudden defluidisation point was observed, but "quenching" took place over a range of increasing liquid loading, with the random bubbling characteristic of the clean ballotini changing gradually, through sluggish particle movement with localised bubble eruption, to channeling and spouting above some or all of the distributor orifices. Spouting may proceed for a prolonged period until the bed surface becomes permanently uneven, stable gas channels
Figure 5.1  Typical Quenching Behaviour With Time
710-850 μm ballotini
$U = 0.8 \text{ m/s}$

$\frac{\Delta P}{\Delta P_0}$

Figure 5.2  Bed Pressure Drop Behaviour During Quenching
425-500 μm ballotini
$U = 0.3 \text{ m/s}$
$H_{MF} = 47 \text{ mm}$
The quenching process is a complex one, but a number of general features of the pressure drop behaviour during quenching can be identified:

(a) Quenching is always accompanied by a reduction in pressure drop across the bed. This has varied in experiments from 10% (710 - 850 μm ballotini) to 70% (425 - 500 μm ballotini; channelled bed).

(b) Bed pressure drop traces usually show a shallow minimum before settling down to a steady value representing the pressure drop for a fully quenched bed.

(c) Bed pressure drop traces often show a small maximum just before any evidence of quenching can be seen. Detection of this small maximum (usually accompanied by a decrease in the magnitude of pressure fluctuations) represents one possibility of observing (and therefore avoiding) the onset of quenching at ambient temperature.

Plates 5.1 and 5.2 are microscope photographs of glass ballotini samples removed from the bed during and after coating. Plate 5.1 shows the uniform size of the liquid bridges after heavy coating. This confirms that the liquid coating is relatively uniform, as would be expected from the method of application. Plate 5.2 a-e is a sequence of pictures of contact points chosen at random from samples extracted during the coating process. Liquid bridges are apparent at every contact point, and their size is observed to increase as the coating time increases. This evidence, though qualitative, suggests that it is justifiable to apply to this situation a model in which equal sized
Plate 5.1  Liquid Bridges Between Glass Ballotini. (710-850 μm; magnification x 40)
Plate 5.2  Liquid Bridges Between Glass Ballotini. (1000–1275; magnification x 40; coated with sebacate at 1.0 m/s in a 0.15 m diameter bed.)

5.2a  \( t = 0 \)

5.2b  \( t = 4 \text{ min} \)
liquid bridges exist at every contact point. The forces at the contacts are therefore a function of the total liquid loading only, provided that complete or partial defluidisation does not occur. (For the samples shown in Plates 5-1 and 5-2 vigorous fluidisation was maintained throughout).

Figure 5.2 shows measurements of bed pressure drop per unit bed depth over the top and bottom halves of a 47 mm deep bed. Quenching is seen to occur first in the bottom half of the bed, and the pressure gradient remains smaller in the bottom half of the bed after quenching is complete. The evidence of Figure 5.2 suggests that agglomeration begins to occur just above the distributor, possibly in the "dead" zones between the distributor orifices. The bed then progressively quenches from bottom to top as channels form in the quenched zone and effectively increase the thickness of the distributor, so reducing the fluidised bed height and therefore the overall pressure drop. If agglomeration were occurring simultaneously everywhere in the bed, one would expect the bed to behave as though it were composed of larger particles and hence the amplitude of pressure fluctuations would be expected to increase as larger bubbles passed up through the bed and burst at the surface. This is not the observed behaviour. The observation that quenching occurs first close to the distributor was also made by Tan (1982), who used a similar arrangement to load the bed, and Smith and Nienow (1981), who used a submerged nozzle to introduce binder into a fluidised bed agglomerator.

Attempts to determine the liquid level at which loss of fluidisation occurred were frustrated by the difficulty in identifying a reproducible and distinct quenching point. As discussed in Chapter 4, other workers have experienced sudden catastrophic defluidisation, but
this has usually been as a consequence of a rapid increase in interparticle force. Here, the increase is very gradual, which may explain the different behaviour.

In these experiments it was found more satisfactory to determine the effect of liquid addition by measuring the minimum fluidisation velocity in the conventional way, repeating the measurement as the liquid loading was increased. Bed pressure drop as a function of superficial gas velocity is plotted for 500 – 600 μm ballotini in Figure 5.3, for clean particles, and at two levels of sebacate loading. A considerable increase in minimum fluidisation velocity is observed, with hysteresis and pressure "overshoot" at the lower liquid loading and the formation of a stable channelled bed structure at the higher loading, which was broken up and made to fluidise at some three times the minimum fluidisation velocity of the clean ballotini.

It is clear, from the experiments summarised above, that at high liquid loadings the interparticle forces are sufficiently large as to cause a material which would normally behave like a Geldart group B powder to show classic "cohesive" behaviour, as if it were in group C. Extending this argument, if Molerus's (1982) reasoning, as outlined in Chapter 4, is correct, it should be possible, by loading the ballotini more carefully, to observe the two transitions from B to A and from A to C. Experiments of this kind were carried out, and these are described in detail in Section 5.3.
Figure 5.3  Fluidisation Characteristics
500-600 μm ballotini loaded with sebacate; log-log scales.
5.2 Measurement of Interparticle Forces.

In order to apply analogous reasoning to that of Molerus (1982) to the case of particles coated with liquid layers, it is necessary either to estimate or measure the interparticle force as a function of the liquid loading. In view of the uncertainties in the theoretical estimation of forces due to liquid bridges (the choice of an appropriate contact radius, for example) it was decided to attempt measurement, preferably without removing the particles from the bed.

Table 5.1 lists the methods which have been used for measurement of interparticle forces, excluding methods applicable to single particles only, and distinguishing between "direct" and "indirect" methods. For the indirect methods, the precise connection between the measurement and the value of the interparticle force remains unresolved. These methods were therefore rejected. The inclined plane, centrifugal detachment, and split cell methods require elaborate equipment to measure the separation force and are not appropriate to in-bed measurement. The remaining methods are the tilting fluidised bed, which was briefly described in Chapter 4, and the inverted bed. Both of these are considered in detail below.
Table 5.1 Measurement of Interparticle Forces.
(excluding single particle measurements)

<table>
<thead>
<tr>
<th>Indirect Measurements</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ratio of tapped to loose bulk density</td>
<td>Geldart et al (1984)</td>
</tr>
<tr>
<td>(&quot;Hausner ratio&quot;)</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Direct Measurements</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Inclined plane (shear strength)</td>
<td>Cremer et al (1952)</td>
</tr>
<tr>
<td>Centrifugal detachment</td>
<td>Boehme et al (1962)</td>
</tr>
<tr>
<td>Split cell</td>
<td>Hartley and Parfitt (1984)</td>
</tr>
<tr>
<td>Tilting fluidised bed (shear strength)</td>
<td>Mutsers and Rietema (1977), Oltrogge (1972)</td>
</tr>
<tr>
<td>Stability of inverted bed</td>
<td>Buysman and Peersman (1967)</td>
</tr>
</tbody>
</table>

5.2.1 Tilting Bed Method

Consider an infinitesimal element within the tilting bed illustrated in Figure 5.4. The two-dimensional equations for equilibrium in the presence of a gas flow in the negative x direction are:

\[
\frac{\partial \sigma_{xx}}{\partial x} - \frac{\partial \tau_{yx}}{\partial y} - \rho_B g \cos \alpha + \frac{dp}{dx} = 0
\]  
\[ (5.1) \]

\[
\frac{\partial \sigma_{yy}}{\partial y} + \frac{\partial \tau_{xy}}{\partial x} - \rho_B g \sin \alpha = 0
\]  
\[ (5.2) \]
Figure 5.4 Stress Convention in Tilting Bed

Figure 5.5 Mohr's Circle Representation of Stress State in Tilting Bed

\[ OQ = \rho_p (1 - \epsilon) gh \]
where \( \rho_B \) is the bulk density of the bed \((=\rho_p(1-\varepsilon))\). Normal stresses are taken as positive in compression and shear stresses are positive anti-clockwise, so that \( \tau_{xy} = -\tau_{yx} \).

The presence of solid walls complicates the problem because each wall can exert normal and tangential stresses on the powder. However, if the bed is very shallow, this complication can be ignored, and the stresses can be assumed invariant in the \( y \) and \( z \) directions. Equations (5.1) and (5.2) can then be integrated, to give

\[
\sigma_{xx} = \rho_p(1-\varepsilon)g\cos\alpha - \left[ \frac{dp}{dx} \right] x
\]

(5.3)

\[
\tau_{xy} = \rho_p(1-\varepsilon)g\sin\alpha
\]

(5.4)

where \( (dp/dx) = \rho_p(1-\varepsilon)g U/U_{MF} \) for \( U < U_{MF} \) if the particle Reynolds number is small.

Hence, \( \sigma_{xx} = \rho_p(1-\varepsilon)g x [\cos\alpha - U/U_{MF}] \)

(5.5)

For an incipiently fluidised bed, \( U = U_{MF} \), so that

\[
\sigma_{xx} = \rho_p(1-\varepsilon)g x (\cos\alpha - 1)
\]

(5.6)

and any inclination of the bed will lead to a tensile stress.

Figure 5-5 is a Mohr's circle representation of the stresses in the tilting bed for the case where \( U = dp/dx = 0 \), i.e. no gas flow. The line CB represents the yield locus; for a Mohr-Coulomb material

\[ \tau = \sigma \tan \phi + c \],

where \( \phi \) is the angle of internal friction and \( c \) is the "cohesion constant", whose magnitude is given by OC on the diagram.
Stresses in the xy plane are represented by the line OQ, so that the point O represents the stress state at the free surface of the bed (both normal and shear stresses at zero) and at Q

\[ \sigma_{xx} = \rho_p (1-e) g h \cos \alpha_{CRIT} \]  
\[ \tau_{xy} = \rho_p (1-e) g h \sin \alpha_{CRIT} \]  

(5.7)  
(5.8)

where \( \alpha_{CRIT} \) is the critical angle of tilt at which failure first occurs. The failure plane is then parallel to the distributor.

Determination of a set of values for \( \alpha_{CRIT} \) for different values of h enables the failure locus to be constructed. The calculations for the case of gas flow through the distributor are similar except that the normal stress at the distributor at failure is now given by:

\[ \sigma_{xx} = \rho_p (1-e) g h \left( \cos \alpha_{CRIT} - \frac{U}{U_{MF}} \right) \]  

(5.9)

for low particle Reynolds numbers.

Thus \( \sigma \) and \( \tau \) can be varied independently and the failure locus can be plotted down to very low (and, in theory, negative) values of the compressive normal stress.

The disadvantages of the method are that it is difficult to apply at gas velocities above \( U_{MF} \), because of the very low levels of cohesion, and that it yields \( c \), the shear strength at zero normal stress, which is not as easily related to the single particle-particle adhesion force as the tensile strength, \( \sigma_T \). Due to the generally unknown shape of the failure locus for tensile normal stresses, \( c \) cannot readily be related to \( \sigma_T \).
5.2.2 Inverted Bed Method

The inverted bed method is a technique first used by Buysman and Peersman (1967) to estimate "natural" interparticle forces caused by van der Waals and electrostatic interactions, and developed further for this work (Seville and Clift, 1984). Figure 5.6 illustrates the column in which the measurements are made, which consists of a cylinder (with, in this case, a diameter of 76 mm and a height of 370 mm) fitted at the bottom with a perforated plate distributor and at the top with a fine wire-mesh screen. Pressure tappings are provided as shown. The whole column can be inverted and mounted upside down. The material to be investigated is first charged to the column, as in Figure 5.6a, enabling the minimum fluidisation velocity to be determined in the usual way. The apparatus is then inverted and the new bed, supported by the gauze screen, is fluidised briefly by reversing the direction of the air flow through the column. The air flow is now stopped and the column tapped gently to level the bed. The air flow is then re-established in the original direction (i.e. downwards) and set to a high value so as to hold the bed against the retaining screen. The apparatus is righted, so that the bed remains suspended by the upward gas flow, as shown in Figure 5.6. The air flow rate is decreased slowly while the pressure drop between two tappings in the column wall is recorded. Eventually a point is reached at which, according to the cohesion of the material, either the particles "rain off" smoothly in successive layers from the bottom of the bed (Plate 5.3a) or the whole bed collapses down the tube as a plug (Plate 5.3b). The pressure drop between the two tappings at the failure point is noted.
Figure 5.6  Apparatus Used for Determination of (a) $U_{mf}$ and (b) Interparticle Forces

Figure 5.7  Pressure Distribution in Suspended Bed at the Moment of Collapse
(Curve A: no interparticle forces; Curve B: with interparticle attraction)
For particles which "rain off" from the bottom of the bed, a modification of the analysis of Buysman and Peersman (1967) was developed. Figure 5.7 shows the pressure distribution in the bed at the instant when "raining" occurs. In a system where interparticle forces are negligible (curve A) and for which wall effects can be ignored, the pressure drop $\Delta P_0$ across the bottom layer, of thickness $\delta$, is equal to the weight of this layer per unit area;

\[ i.e. \quad nG_p = \Delta P_0 \]  

(5.10)

where $n$ is the number of particles in the layer per unit bed cross section, and $G_p$ is the average immersed weight of each particle. In the presence of interparticle forces (curve B), equation (5.10) becomes

\[ nG_p = T + \Delta P_F \]  

(5.11)

where $T$ is the tensile stress exerted on the bottom layer by the adjacent particles and $\Delta P_F$ is the pressure drop across the layer at the point of "raining off". From equations (5.10) and (5.11),

\[ T = \Delta P_0 - \Delta P_F \]  

(5.12)

Because the particles in the "raining" layer form the upstream face of the bed, the pressure drop across this layer is less than that across a layer of thickness $\delta$ in the bulk of the bed. However, the ratio of the pressure drop across the upstream layer to that across a section within the bed may be assumed constant provided that the bed particle Reynolds number is comparable in each case (Buysman and Peersman, 1967; Rowe and Henwood, 1961). Hence
where $\Delta P_0$ and $\Delta P_F$ are the pressure drop between the two measuring points in the bed at the point of "raining off", for particles without and with interparticle forces respectively. From equations (5.12) and (5.13)

$$\frac{\Delta P_0}{\Delta P_o} = \frac{\Delta P_F}{\Delta P_F} = C \quad (5.13)$$

while, from equations (5.10) and (5.13),

$$T = C (\Delta P_0 - \Delta P_F) \quad (5.14)$$

while, from equations (5.10) and (5.13),

$$C = \frac{nG_p}{\Delta P_0} \quad (5.15)$$

Hence, from equations (5.14) and (5.15),

$$T = nG_p \left[ 1 - \frac{\Delta P_F}{\Delta P_0} \right] \quad (5.16)$$

The average tensile force transmitted per particle in the layer is then

$$F_T = \frac{T}{n} = G_p \left[ 1 - \frac{\Delta P_F}{\Delta P_0} \right] \quad (5.17)$$

which is the result obtained by Buysman and Peersman (1967).
In this work we are interested not in the average tensile force transmitted per particle but in the average adhesion force transmitted at a single contact between particles, $F_h$, which follows from an analysis by Rumpf (1962). The Rumpf model is based on the following assumptions:

(i) the number of bonds in the failure cross-section is large;
(ii) the bonds are statistically distributed across the section and over all directions in space;
(iii) the particles are spherical, of a single diameter, $d_p$, and are statistically distributed in the bed;
(iv) the effective bonding forces are distributed around a mean value which can replace them in calculations and may be assumed to be equal at different points on the cross-section.

Rumpf's analysis was developed in order to predict the tensile strength of dry agglomerates, for which it has been criticised (see, for example, Kendall et al, 1986), mainly on the grounds that even small imperfections in the structure of an agglomerate can lead to large localised stress concentrations, so that a fracture mechanics approach must be taken. The use of Rumpf's approach is justified here on the grounds that

(a) the size of the liquid bridges appears to be uniform throughout, and, more importantly,
(b) the bed failure is observed to be simultaneous across the entire cross-section.

Figure 5.8a shows the theoretical (planar) and actual failure areas in the bed. The assumption of statistical distribution of particles requires that no cross-section be distinguished. It follows that the void fraction in the failure cross-section, $\varepsilon_f$, is equal to the void
Figure 5.8a Rumpf's Analysis - Fracture Area.

Figure 5.8b Rumpf's Analysis - Contacts Per Particle
fraction in the total volume, $\varepsilon$. Now consider a single particle in the
failure plane, as shown in Figure 5.8b. The surface of this particle
which is actually involved in the failure (the "active" surface) has
area $A(z)$, where

$$A(z) = 2\pi r(r-z) \quad (5.18)$$

The average number of particle-particle contacts per particle in the
failure cross-section is given by:

$$\bar{k}_c = \frac{A}{\pi d^2} \cdot \bar{k} \quad (5.19)$$

where $\bar{k}$ is the mean coordination number, and

$\bar{A}$ is the mean area of the sphere surface in the failure
cross-section,

$$\bar{A} = \frac{1}{r} \int_0^r A(z) \, dz = \frac{2\pi}{r} \int_0^r r(r-z) \, dz = \frac{\pi d^2}{4} \quad (5.20)$$

Hence,

$$\bar{k}_c = \frac{\bar{k}}{4} \quad (5.21)$$

and the angle pertaining to $\bar{A}$ is given by

$$\theta_0 = 2\pi/3 \quad (5.22)$$

Since the tensile forces at the contacts are randomly distributed over
the active surface of the spheres and act normally to it, their
contributions must be resolved in the $z$ direction by multiplying the
sum of their absolute values by the quotient

$$\int_0^{\theta_0} 2\pi r^2 \sin\delta \, c_{05} \delta \, d\delta \quad (5.23)$$

$$\int_0^{\theta_0} 2\pi r^2 \sin\delta \, d\delta = \frac{3}{4}$$

Hence, the average tensile force transmitted per particle in the
failure plane, $F_T$, is given by:
According to Smith et al. (1929), the mean coordination number, \( k \) is related to the voidage, \( \varepsilon \), by the approximate expression

\[
\bar{k} \varepsilon \approx \pi
\]

Hence

\[
F_T \approx \frac{3\pi}{15} \frac{F_H}{\varepsilon}
\]

This differs slightly from the expression derived by Molerus from Rumpf's equations:

\[
F_T \approx \frac{\pi}{6} \frac{F_H}{\varepsilon}
\]

Since \( \varepsilon \) is usually about 0.5, \( F_T \) is usually approximately equal to \( F_H \), whichever equation is used. In view of the approximation inherent in any estimate of \( \bar{k} \), Molerus' expression has been used in the calculations which follow. Substituting for \( F_H \) in equation 5.17:

\[
F_H = \frac{6\varepsilon F_T}{\pi} = \frac{6\varepsilon G}{\pi} \left[ 1 - \frac{\Delta P_F}{\Delta P_O} \right]
\]

In Buysman and Peersman's (1967) experiments \( \Delta P_O \) was obtained for the same particles in water rather than air, since it was supposed that no interparticle forces would act in water. This is not in fact the case (see, for example, Israelachvili, 1985). Furthermore, the assumption is inconsistent with equation (5.13), since the Reynolds numbers are not similar in the two experiments, with and without interparticle forces. In this work, however, the interparticle forces due to the presence of
liquid bridges are at least an order of magnitude larger than the residual van der Waals and electrostatic forces, so it is permissible to use a bed of clean, dry particles to determine \( \Delta P_0 \).

As suggested by Buysman and Peersman (1967), and observed in these experiments, when the interparticle forces are large the inverted bed collapses not by "raining" but by failure at or near the top closure, to fall as a plug. Buysman and Peersman (1967) did not investigate this behaviour, concluding that this regime was useless for determining the interparticle forces since the above analysis cannot be applied to it. In fact, the transition from raining to plug-fall modes of failure proves to be of considerable interest. An analysis of this transition is now considered.

Consider curve B in Figure 5-7, for conditions where the bed either rains off or falls as a plug. For any plug of thickness \( z \) \((z \geq 8)\) to be stable:

\[
(\rho_p - \rho_g)(1 - e)gz - \left[ \frac{dP'}{dz} \right] s - \frac{dP}{dz}(z-s) < T
\]  

(5.29)

where \( \rho_p \) and \( \rho_g \) are the particle and fluid densities, and \( dP/dz \) and \( dP'/dz \) are the pressure gradients in the bulk of the bed and in the bottom layer respectively. As in equation (5.13), we may write

\[
\left[ \frac{dP'}{dz} \right] = c \left[ \frac{dP}{dz} \right]
\]  

(5.30)

where \( c \) is a constant less than unity. Therefore equation (5.29) can be restated as

\[
(\rho_p - \rho_g)(1 - e)gz - T < \frac{dP}{dz} [z - s (1 - c)]
\]  

(5.31)

In a randomly packed and randomly coated bed the limiting tensile stress, \( T \), is constant for all \( z \). Moreover, if the adhesion between the bed and the top closure exceeds \( T \), the bed will fail internally.
rather than by detaching from the retaining screen; that this occurred in the present experiments was confirmed in all cases by a residual layer of bed particles left on the gauze screen after collapse, as shown in Plate 5.3c. The failure condition given by equation (5.31) can therefore be represented as in Figure 5.9. The left-hand side is a property of the material; for high cohesion (i.e. high $T$) it appears as line 1. In the experiment, $(dp/dz)$ is initially high (line 2A) and is reduced until raining or plug fall occurs; thus the gradient of the line is reduced until line 2 is reached. Line 2 crosses line 1 at the top of the bed ($z = L$); i.e. equation (5.31) is no longer satisfied, and failure occurs at the top of the bed so that the material falls as a plug. For less cohesive particles (lower $T$) the left hand side of equation (5.31) is displaced upwards on the diagram; the limiting case is line 1A, which corresponds to the condition at which failure by raining or plug flow are equally likely. For lower values of $T$ still, failure always occurs at $z = \delta$; i.e. "raining off" occurs. The limiting condition, line 1A, is clearly represented by

$$8(1-c) = \frac{T}{(\rho_p - \rho_g)(1 - \varepsilon) g}$$

(5.32)

i.e.

$$T = (\rho_p - \rho_g)(1 - \varepsilon) g 8(1 - c)$$

(5.33)

The number of particle/particle contacts per unit area broken in a failure plane can be approximated by

$$n = \frac{B}{d_p^2}$$

(5.34)

where $d_p$ is the mean particle diameter and $B$ is a constant for given $\varepsilon$. 
Figure 5.9  Failure Conditions for Suspended Bed (see equation 5.29)
Also, the thickness of a "raining" layer can be written

\[ s = A d_p \quad (5.35) \]

with \( A \) also constant for given \( \varepsilon \). Thus the average tensile force transmitted per particle at the limiting condition given by equation (5.33) can be expressed as

\[
F_T = (\rho_p - \rho_g)(1 - \varepsilon) g A d_p (1 - c) / (B/d_p^2) \quad (5.36)
\]

\[
= K' (\rho_p - \rho_g) g d_p^3 \quad (5.37)
\]

where \( K' \) is a constant for constant \( \varepsilon \). From equation (5.28) the average adhesion force between two particles is then

\[
F_H = K(\rho_p - \rho_g) g (\pi d_p^3 / 6) \quad (5.38)
\]

where \( K (= 36 \varepsilon K' / \pi^2) \) is constant for constant \( \varepsilon \).

Equation (5.38) is identical in form to Molerus' condition for the transition between groups A and B (equation (4.20)). It therefore suggests a simple correspondence between the transition from B to A behaviour in a fluidised bed and the transition from "raining" to "plug" failure in a suspended bed.

The analysis above presents not only a comparatively simple method for determining interparticle forces directly but also provides an experimental test of Molerus' (1982) explanation of the Geldart (1973) groups. This method was therefore adopted and used in the experimental work described below.
5.3 Experimental Results.

Experiments were carried out using two size fractions of lead glass ballotini, which were coated with sebacate using the method of Section 5.1. The two fractions were coated to the desired level in the 0.152 m diameter column, sampled for sebacate loading, and then a suitable measured proportion was placed in the inverted bed column, in which the minimum fluidisation velocity was determined in the conventional way. For each size fraction, at each stage of loading, the raining/plug-fall test was also performed as set out in Section 5.2.2, and the value for pressure drop at failure was recorded.

The minimum fluidisation velocity determinations are presented in Figures 5.10 and 5.11. In each figure, the ratio of pressure drop across the bed to bed weight per unit area is plotted as a function of the superficial gas velocity in the column for a number of different loadings. The curves are identified according to the coating level. Figures 5.12 and 5.13 show measurements of settled bed voidage, corresponding to the $U_{mf}$ determinations of Figure 5.10 and 5.11. Voidage was obtained by weighing the charge into the bed and measuring the corresponding bed height after a few moments of fluidisation to obtain a homogeneous bed.

Very light liquid loading appears to cause a slight reduction in the minimum fluidisation velocity, as reported for group A powders by d'Amore et al (1979). In agreement with the work reviewed in Chapter 4, the minimum fluidisation velocity at higher liquid loadings is markedly increased. However, what is most striking about these results is that above a certain loading level the bed expands without bubbling when the minimum fluidisation velocity is exceeded. Figure 5.14
Figure 5.10 Ratio of Pressure Drop to Bed Weight Per Unit Area, As a Function of Gas Velocity, for 310-425 μm Ballotini

Figure 5.11 Ratio of Pressure Drop to Bed Weight Per Unit Area, As a Function of Gas Velocity, for 505-700μm Ballotini
Figure 5.12 Voidage of Settled Bed, 310-425 μm Ballotini

Figure 5.13 Voidage of Settled Bed, 505-700 μm Ballotini
Figure 5.14 Richardson–Zaki Plot of Expansion of Heavily Loaded 505–700 μm Ballotini
shows the expanded bed voidage plotted against gas velocity for two heavily loaded samples. The relationship between them is seen to be linear on log-log coordinates, confirming the applicability of the Richardson-Zaki relationship, equation (4.9). This behaviour is characteristic of group A powders, and confirms that the bed behaviour can be caused to move from group B, through group A, and into group C by a gradual increase in interparticle cohesive forces.

Figures 5.15 and 5.16 show the values of $F_T$ calculated from equation (5.17), as functions of the the average mass percentage of sebacate, for the two sizes of ballotini. Figure 5.17 shows the voidage measurements of the suspended beds used in the determinations of $F_T$ presented in Figures 5.15 and 5.16. Voidage was obtained by weighing the charge into the bed and measuring the corresponding bed height. For each determination the bed was briefly fluidised in the inverted position, as mentioned previously. The apparatus was then tapped gently to level the surface, the air flow direction was reversed, and the apparatus righted, leaving the bed suspended against the top closure. The voidage of the suspended bed is, to some extent, a function of the care with which it is prepared. Figure 5.17 shows that for the work presented here the voidage was maintained at an approximately constant value. Therefore, the analysis of the previous section can be applied. In a number of earlier exploratory experiments attempts were made to consolidate the suspended bed by prolonged vibration. Beds which were prepared in this way did not behave satisfactorily when the suspending gas flow was reduced. For lightly loaded beds, the first few centimetres of bed would rain off at constant gas flowrate and raining would then stop until the gas flow was reduced still further. Similarly, for heavily loaded beds, the first few centimetres of bed would fall as a plug, leaving the
Figure 5.15 Average Interparticle Force As a Function of Liquid Loading, for 310-425 μm Ballotini

Figure 5.16 Average Interparticle Force As a Function of Liquid Loading, for 505-700 μm Ballotini
remainder to fall as a second plug when the gas flow was further reduced. In both cases, this a clear indication that the lower part of the bed had a lower voidage. The most reproducible behaviour occurred for beds which were as little consolidated as possible; the relatively high voidages of Figure 5.17 are a consequence of this.

Some attempt was made to discover the effect of suspended bed height on critical "ceiling" conditions. Experiments were carried out for initial bed heights of 10.8, 8.5, and 6.3 cm, all in the 7.6 cm diameter column. In each case, raining and plug-fall were initiated at the same values of suspending gas flowrate and suspended bed pressure drop, the latter being measured between the tappings shown in Figure 5.17.
5.7. It therefore appears, as predicted, that the analysis of suspended bed failure does not depend on the thickness of the suspended bed, provided that the voidage is uniform.

Clearly equation (5.17) only applies for liquid loadings up to the transition from "raining" to plug collapse. The values of \( F_T \) in this range are approximately linear functions of sebacate loading. Departure of \( F_T \) from the initial trend defines the point at which equation (5.17) becomes inapplicable; i.e. the point at which transition from "raining" to plug collapse occurs. It is not fruitful to apply the plug failure analysis above this point because the terms on the left of equation (5.29) are of comparable magnitude so that \( T \) cannot be determined with accuracy. For both particle sizes, the transition corresponds roughly to

\[
F_T = 0.55 \, G_p = 0.55 \left( \rho_p - \rho_g \right) \left( \pi \frac{d^3}{6} \right) g
\]  

(5.39)

i.e. using equation (5.28) with the average value \( \epsilon = 0.41 \):

\[
\left( \rho_p - \rho_g \right) \left[ \pi \frac{g \, d^3}{6} \right] / F_H = 2.3
\]  

(5.40)

Comparison with Figures 5.10 and 5.11 shows that the liquid loading at the transition from "raining" to plug collapse is essentially the same as at the transition from group B to group A fluidisation; thus equations (5.39) and (5.40) appear to describe both transitions. As confirmation, a clean Group A powder (fluid cracking catalyst of mean diameter 75 \( \mu \)m) was tested by the procedure in Section 3.1; it showed plug collapse rather than "raining".
5.4 Discussion

5.4.1 Comparison of Measured Interparticle Forces with Theoretical Predictions.

For both size fractions of ballotini considered in the previous section, the transition between raining and plug-fall occurs at a value of the average adhesive force transmitted at a single contact, \( F_H \), of approximately \( \frac{G_p}{2.3} \), where \( G_p \) is the particle weight. Taking the appropriate particle diameter as the geometric mean for the interval, this gives values for \( F_H \) of \( 3.15 \times 10^{-7} \) N and \( 1.39 \times 10^{-6} \) N at the transition, for the smaller and larger fractions respectively. From equation (4.7), the maximum liquid bridge force between two equal spheres of radius \( R \) is \( 2\pi R\gamma \), where \( \gamma \) is the surface tension of the liquid (0.032 N/m for sebacate). Hence the maximum liquid bridge forces for these same size fractions, assuming the bridge strength to be determined by the gross sphere radius, are \( 3.65 \times 10^{-5} \) N and \( 5.98 \times 10^{-5} \) N respectively. Thus the measured forces are 0.9% and 2.3% of the maxima. It is clear, therefore, that the bridge forces are not determined by the gross sphere radius at these levels of liquid loading, but by the surface asperities. The maximum liquid bridge force for the sphere-plane system is given by \( 4\pi R\gamma \), where \( R \) is the radius of the sphere. Using this equation, asperity radii of 0.8 and 3.5 \( \mu \)m, respectively, are predicted. Scanning electron microscope pictures of "clean" ballotini surfaces confirm that asperities of this order of size are present. The results are thus consistent with the idea that as liquid loading is increased the liquid bridge force is determined by larger and larger asperity sizes, making a priori prediction virtually impossible.
The results presented in Section 5.3 give qualitative support for the use of Molerus' (1982) expression for the B-A transition:

\[
\frac{(\rho_p - \rho_g) n d_p^3 g}{F_H} = K
\]

(5.41)

where \( K \) is a constant, estimated here as 2.3 and by Molerus as about 0.16. Molerus' estimate is based on a purely theoretical expression for \( F_H \) due to van der Waals forces and plastic deformation (Appendix A2), so it is of interest to compare this estimate for \( F_H \) with others.

Buysman and Peersman (1967) and Donsi et al (1975) both attempted direct measurement of the interparticle attractions between particles close to the B-A transition. Their results are presented in Figure 5.18 and compared with Molerus' value. Massimilla and Donsi (1976) also estimated the value of \( F_H \) for cracking catalyst, using Krupp's (1967) equation (equation A2.5), but taking into account the measured distribution of surface asperities as discussed in Section 4.2.2. Their estimates are also presented in Figure 5.18. Comparison with these measured and estimated values for \( F_H \) due to van der Waals forces indicates that Molerus' value is at least an order of magnitude larger than it should be. Taking Buysman and Peersman's range of measured values, for example, and substituting into equation (5.41) gives a range of values for \( K \) from 2.0 to 2.9, encompassing the present estimate of 2.3 but an order of magnitude above Molerus' value.
Figure 5.18 Comparison of Estimates for Interparticle Van der Waals Attraction

- $F_W$ based on particle radius;
- $F'_W$ based on mean asperity radius
  (Massimilla and Donsi, 1976)
- Measurements on cracking catalyst
  (Donsi et al, 1975)
- Measurements on aluminium silicate
  (Buysman and Peersman, 1967)

- Particle weight
- Line representing Molerus' (1982) assumption for glass and cracking catalyst (van der Waals forces only).
Figure 5.19 Effect of Interparticle Force on Minimum Fluidisation Velocity - Glass Ballotini

\( U_{MF} = \text{minimum fluidisation velocity for "clean" material; } U_{MF}' = \text{minimum fluidisation velocity for coated material.} \)

- 310-425 \( \mu \text{m} \)
- 505-700 \( \mu \text{m} \)

5.4.3 The Effect of Interparticle Cohesion on Settled Bed Voidage.

Figure 5.19 relates the relative increase in minimum fluidisation velocity to the ratio of interparticle force to particle weight.

Figures 5.12 and 5.13 show that liquid loading has a perceptible effect on settled bed voidage, \( \varepsilon_S \). Because of the strong influence of \( \varepsilon_S \) on pressure gradient, this effect gives rise to the influence on pressure gradient and \( U_{MF} \) shown in Figures 5.10, 5.11 and 5.19. Low liquid loadings appear to cause a slight decrease in both \( \varepsilon_S \) and \( U_{MF} \), as observed for Group A particles by d'Amore et al (1979).

When loading is increased further, \( \varepsilon_S \) increases, the pressure gradient for a given gas velocity is therefore reduced, and hence \( U_{MF} \) is increased. This increase in settled bed voidage is consistent with the conclusions of Donsi and Massimilla (1973) and Rietema (1967) that
cohesion between solid particles can stabilise structures of higher porosity. Taken with the present observation that the B/A transition coincides with the transition between failure modes for a suspended bed, a qualitative explanation can be proposed for the difference in fluidisation characteristics between groups A and B. In a group A material, small voids are stable against "raining in" of the particles forming the roof; formation of local voids may therefore cause the bed expansion observed at gas velocities just above $U_{mf}$. For group B particles, such voids will not be stable against "raining in", so that voids must be stabilised by dynamic processes, i.e. by rising as the familiar bubbles.

For group C materials, the interparticle forces are sufficiently large as to maintain the cohesive bed structure even at high gas flowrates. As the gas flowrate is increased voids may grow into vertical channels, which reduces the pressure drop across the bed for a given flowrate so that the gas is not able to penetrate and break up the bed structure unless external agitation is introduced.

5.5 Conclusions

Increasing interparticle cohesive forces in a bed of group B material by loading the particles with an involatile liquid causes the fluidisation behaviour to move from group B through group A to group C. These observations are qualitatively similar to other reported effects of interparticle forces, for example the observations of Agbim et al (1971) on the effects of magnetising particles, and are consistent with Molerus' (1982) explanation of fluidisation behaviour in terms of interparticle cohesive forces. The transition from group B to A is
observed to coincide with the transition between "raining" and "plug-fall" modes of failure of a bed of solids suspended by an upward gas flow. The results of such experiments confirm the form of Molerus' expression for the B/A transition, but with a different and probably more realistic value for the dimensionless constant involved. Increasing interparticle cohesion generally increases bed voidage, and hence increases the minimum fluidisation velocity. This results from stabilisation of local void structures, rather than stabilisation of the uniformly expanded state.
Fluidised Bed Filtration - Experimental

6.1 Selection of the Retention Aid

Of the aids to retention used by previous workers (Chapter 2), most appear unsuitable for use in a fluidised bed filter designed for the clean-up of gasifier off-gases. For example, boric oxide, as used by Ghadiri (1980), reacts readily with water vapour to form a volatile species and is rapidly lost from the collector. It is therefore necessary to consider alternatives. The requirements of a retention aid for the proposed duty are as follows:

(i) it must possess adhesive properties at 800-1000°C;
(ii) it must wet both the collector and the aerosol particles;
(iii) it must be capable of dispersion into the bed;
(iv) it must not produce secondary particles on dispersion;
(v) it must not produce secondary particles by attrition from the collector;
(vi) it must not create harmful or corrosive gaseous species, such as alkali metal salts;
(vii) it must be cheap and readily obtainable.

Among the retention aids which have been suggested are:

(a) aluminium chloride (USDoE, 1979);
(b) synthetic high-temperature lubricants, such as the polyphenyl ethers (Byford, 1982);
(c) residual heavy fuel oil, with or without a particulate additive such as ash or high-ash coal.

Aluminium chloride was rejected because of its tendency to hydrolyse under process conditions (Reed, 1982) and the possibility that it might produce corrosive species. Synthetic lubricants were not considered sufficiently cheap and easily available for an industrial process. The work which follows therefore relates to the use of heavy fuel oil alone. Additives were not used, in order to reduce the possibility of generating secondary particulates at the dispersion stage, although whether this is a problem remains to be established. The oil which was selected is a 3500s (Redwood) residual fuel oil.

In the temperature range of interest for gasification, oils "crack" yielding light hydrocarbons and tarry residues. This is the basis of thermal cracking processes such as the Lurgi "sand cracker" (Schmalfeld, 1963) and the Japanese "KK" process (Shohji et al, 1983). The mass of tarry residues is typically 20-30% of the oil mass for a heavy fuel oil (Linden and Peck, 1955; Schulz et al, 1955). The aim of this work is to use the tarry residue as the retention aid, since tar wets both carbon char and sand. In an industrial application, the light cracking products would give a slight increase in the calorific value of the off-gases. The cracking time for heavy oils in nitrogen at 800-900°C is approximately one second (Pettyjohn and Linden, 1952), although this may be extended by using steam rather than nitrogen as the nozzle atomising fluid (if a two fluid-nozzle is used). It is therefore necessary for the oil to be dispersed very rapidly onto the collector surfaces, both to avoid premature cracking in the dispersion nozzle and to allow sufficient time for the aerosol particles to contact the collector surface before cracking is complete.
6.2. Retention Aid Dispersion

Although no prior work has been reported in which oil was dispersed into a fluidised bed with the aim of providing a residue to improve filtration, a number of workers have investigated the related areas of liquid dispersion in fluidised bed agglomeration, and in fluidised bed gasification and combustion of heavy oils.

Liquid addition from a two-fluid nozzle located in the freeboard and spraying downwards onto the bed surface is commonly used in fluidised bed agglomeration (Nienow and Rowe, 1985), but this can lead to excessive loss of spray-dried droplets and problems of "caking" on the nozzle and on the walls of the bed. Submerging the nozzle within the bed has been found to prevent caking (e.g. Smith and Nienow, 1981), and modification of the nozzle to promote external rather than internal mixing has also been found helpful (Maroglou and Nienow, 1985). In fluidised bed coating processes, where agglomeration of the bed particles must be avoided, nozzles located at the distributor and spraying upwards into the bed are favoured (e.g. Vintner, 1982; Huddleston, 1982).

Legler (1967) used a commercial cooled two-fluid nozzle (Spraying Systems Inc.) to atomise a radioactive waste solution into a fluidised bed calciner operating at 300 - 500°C. The waste salts (principally aluminium nitrate) decomposed in the bed, the volatile components being driven off while the remaining metallic oxides were deposited as layers on the bed particles. The present work has two objectives in common with that of Legler:
(i) adequate coating of the bed particles must be achieved without agglomeration or defluidisation;

(ii) production of particles or droplets small enough to be entrained by the gases leaving the bed must be minimised.

Legler found that satisfactory results were obtained with the nozzle inserted through the bed wall, just below the bed surface, and with the fluids entering the bed horizontally. Spraying into the freeboard caused caking on the nozzle and elutriation of spray-dried feed. Introducing the nozzle through the distributor to spray the liquid vertically upwards was not tested.

Reid et al (1953), investigated cracking of oils of different C/H ratios in a fluidised bed. They report that oils of high C/H value with high carbon residue were difficult to feed and tended to promote agglomeration, but that satisfactory operation could be achieved by spraying the heavy oil using a two-fluid steam atomising nozzle inserted horizontally through the wall of the bed. Pillai and Elliott (1976), attempted to burn heavy fuel oil at 850°C in fluidised beds of 455 \( \mu m \) silica sand with static depths in the range 30 to 150 mm. The injector was an air-cooled stainless steel tube, adjustable in position so that injection could take place just above or below the bed surface or close to the distributor. These authors also experienced agglomeration problems, but succeeded in eliminating injector blockage completely by submerging the nozzle within the bed. It should be noted that both Reid et al and Pillai and Elliott used oil addition rates much greater than those contemplated here.
Three nozzle designs were initially considered, all of which were two-fluid types. Preliminary calculations showed that the liquid flow-rate required for a 0.15 m diameter bed would be of order millilitres per minute, which prevents the use of single fluid nozzles since these are not generally capable of operation at this low liquid throughput. It was also considered that the atomising gas, being cooler than the fluidising gas, would help to delay the cracking of the oil in the nozzle itself and in the region surrounding it. Three nozzle types were considered:

(i) the commercial Spraying Systems Inc. "1/4 J" range (available with a range of interchangeable liquid and gas "caps");

(ii) the "Y-jet" atomiser (Mullinger and Chigier, 1974);

(iii) the B.P. climbing film atomiser (Sutton, 1982)

The Spraying Systems 1/4 J range of nozzles (Figure 6.1) was used by Legler (1967) and is employed in fluidised bed agglomeration and coating processes (Smith and Nienow, 1981). It was rejected for this work because of the large liquid hold-up in the nozzle body, which would be likely to lead to premature cracking in the nozzle itself.

The B.P. climbing film atomiser (Figure 6.2) was developed specifically for combustion of oils in fluidised beds. Experiments with a perspex model showed that the liquid residence time in the nozzle was again too long for it to be used in this application.
Figure 6.1 Spraying Systems Nozzle

Figure 6.2 B.P. Climbing-Film Nozzle
A perspex model of the 'Y-jet' atomiser was designed and built according to the design recommendations of Mullinger and Chigier (1974) (Figure 6.3). This was tested in a 0.15 m diameter fluidised bed of sand at ambient temperature using a liquid consisting of a 20% v/v solution of di-2-ethyl hexyl sebacate in ethanol. The bed depth was varied between zero and 0.11 m. Two injection configurations were investigated:

(i) horizontally, through the bed wall, as recommended by Legler (1967) and Reid et al (1953), the nozzle axis being fixed 39 mm above the distributor so that the nozzle was normally submerged beneath the bed surface;

(ii) vertically upwards through the centre of the distributor.

In these experiments, the total number of droplets greater than 0.5 μm in diameter in the off-gas from the bed was measured at a single point in the freeboard using a Bausch and Lomb 40-1A single-particle light-scattering counter (see Chapter 3). The collector was silica sand in the size range 710-850 μm, fluidised with air at 1.0 m/s, and the liquid flowrate to the nozzle was 0.75 ml/min. Figure 6.4 indicates the effect of bed depth and atomising air flow on over-sprayed particle count for both nozzle entry configurations. These results cannot be considered as representative of the behaviour to be expected for oil atomisation at high temperature, since the properties of the liquid, particularly its viscosity, are very different in the two cases. However, they do indicate the expected superiority of the horizontal entry arrangement in reducing carry-over of droplets into the freeboard, and the importance of keeping the
Figure 6.3  Y-Jet Nozzle
Figure 6.4 Penetration of Retention Aid Droplets Through Bed—Variation With Bed Depth.
Collector: sand 710 - 850 μm
U = 1.0 m/s
Retention Aid: 20% v/v sebacate in ethanol; 0.75 ml/min
Measuring point: 0.33 m above distributor
atomising gas flow to a minimum. (However, reduction of the atomising flow to below 20 l/min destroyed the atomisation and caused immediate agglomeration of the bed material in the vicinity of the nozzle. There is therefore a lower limit to the atomising gas flow). The horizontal arrangement also has the advantages that it is easier both to construct and to keep cool, since the bulk of the nozzle and the feed pipes lie outside the bed column. The Y−jet nozzle, adapted for horizontal entry into the bed, was therefore made up in stainless steel for use at high temperature.

6.3 Apparatus

The high temperature filtration rig used in this work is described in detail by Ghadiri (1980). In outline (Figure 6.5) it consists of:

(i) facilities for supplying nitrogen (in place of the reducing atmosphere in the gasifier) at controlled flowrates;

(ii) a device for feeding and dispersing gasifier fines into the gas stream (described in Chapter 3);

(iii) an electrically−heated preheater, capable of raising the temperature of the gas to 1000°C;

(iv) a 0.152 m diameter fluidised bed, 0.475 m in height, with a perforated plate distributor, constructed of Inconel 600 and located within an electrically heated furnace capable of maintaining the temperature inside the bed at up to 1000°C;
Figure 6.5 Schematic Diagram of Fluidised Bed Filter Rig
(after Ghadiri, 1980)
(v) sampling probes below the distributor, and in the freeboard of the bed, 0.4 m above the distributor plate.

6.3.1 Distributor Plate

The distributor plate used in the initial experiments was a flat circular Inconel plate perforated with 109 x 1.5 mm diameter orifices on a 1.4 cm triangular pitch, giving a free area of 1.06%. The orifice profile is shown in Figure 6.6 and is of the general form recommended by Clift et al (1981), consisting of a 30° countersunk hole with a short 1.5 mm diameter cylindrical portion on the downstream side. The countersink prevents aerosol deposition on the upstream side of the distributor, and also reduces the pressure drop across it. In all the experiments reported here, the ratio between the distributor pressure drop and the bed pressure drop was well in excess of that which is necessary to ensure uniform fluidisation (Geldart, 1981).

Figure 6.6 Distributor Orifice Profile (after Ghadiri, 1980)
Following the initial experiments with the Y-jet nozzle some of the orifices were blocked using high temperature cement to give an arrangement of 31 x 1.5 mm orifices on a 2.8 cm triangular pitch, identical to that used by Ghadiri (1980). This arrangement, which gives a free area of 0.30%, was used for all subsequent experiments.

6.3.2 Sampling and Particle Size Analysis

Sampling of the gases upstream and downstream of the filter was performed by isokinetically extracting a small sample of the gas (typically 2 l/min) through 16 mm diameter sharp-edged probes located on the bed centre line 5 cm below and 40 cm above the distributor. The sample gas was then passed either through a nominal 0.2 µm pore size cellulose acetate micropore filter or a liquid impinger. The design and construction of the liquid impinger are considered in detail in Chapter 3. The short length of sample pipe outside the bed housing was sufficient to cool the gas sample to a temperature at which it caused no damage to the filter. If filter sampling is used, the overall filtration efficiency can be determined directly, allowing for the attrition product weight, from the weight gain of the filters. The size analysis methods are considered in detail in Chapter 3. In brief, the particles collected on the filter can be viewed directly using an optical or scanning electron microscope or washed from the filter in an ultrasonic bath, suspended in aqueous sodium chloride electrolyte, and analysed for equivalent-volume sphere diameter distribution using the Coulter counter. If liquid impingers are used, the particle dispersion step is unnecessary, as the collection liquid can be the electrolyte itself. However, it should be noted that in this case the overall
gravimetric efficiency cannot be obtained directly, but must be calculated from the total volumes of particulates measured by the Coulter counter.

6.3.3 Collector Particles

The collector particles used for all runs were high quality low-iron silica sand (Ball and Co.) with a specification given in Table 6.1. The sand was first sieved to between 850 and 1000μm. A weighed amount was then inserted into the bed via the exit pipe. At the end of each run the bed material, still fluidised, was extracted by inserting an Inconel pipe into the bed from above and connecting this, via a cyclone, to the inlet of an industrial vacuum cleaner.

<table>
<thead>
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<th>Table 6.1 Collector Sand Properties</th>
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<table>
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<tr>
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</tr>
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</table>
6.3.4 Retention Aid Supply and Dispersion

Figure 6.7 shows the side entry nozzle arrangement whose development was described in Section 6.2. In order to prevent premature cracking of the retention aid in the nozzle or the liquid feed line, a cooling air pipe was installed concentric with the oil feed line, the end of the pipe being perforated so that the jet of air issuing from it provided some cooling of the nozzle area. All the lines carrying fuel oil were electrically heated to around 70°C in order to reduce the oil viscosity to an acceptable level for handling. The oil pump was a Type HM "Metripump" (Metering Pumps Ltd.) of the plunger type, with micrometer stroke control, the delivery rate being adjustable from zero to a nominal 0.73 litres/hour. In case of nozzle blockage a 1.7 bar g relief valve was fitted to the oil delivery line and a miniature Bourdon tube pressure gauge was fitted to indicate oil delivery pressure. The atomising nitrogen supply pressure was also continuously monitored in order to reveal nozzle blockages.

6.4 Experimental Method

The experimental method adopted in all filtration tests was as follows:

(i) The collector sand was sieved, weighed, and added to the bed.

(ii) The bed was fluidised in air at the temperature and velocity to be used in the filtration experiment. The atomising and oil cooling flows were set at their required values. The bed was kept fluidised for at least an hour.
Figure 6.7  Y-Jet Atomiser Arrangement - Schematic
(iii) Keeping the bed fluidised, the exit gases were sampled for attrition product for a further hour, by withdrawing gas through a micropore filter for subsequent Coulter analysis.

(iv) The oil heating mantle and tracing tape were switched on and adjusted to give an oil temperature of 70°C.

(v) The fluidising gas was switched to nitrogen and the fluidised bed gasifier fines disperser brought on line (keeping the total gas flow constant). The disperser was run for half an hour.

(vi) If retention aid was to be used, the delivery pump was switched on for five minutes and the oil delivery pressure and temperature monitored for abnormalities.

(vii) The entrance and exit flows were sampled, either onto micropore filters, or into liquid impingers for one hour.

(viii) The oil delivery line was emptied by removing the oil reservoir and allowing pumping to continue for a period.

(ix) The collector was rapidly removed via a tube inserted down the exit pipe of the rig.

(x) The fluidising gas was switched back to air and the flow reduced.

(xi) After careful examination for agglomerates and uniformity of coating a small sample of the bed material was retained and the remainder returned to the rig so as to burn off any collected
carbon. The sand was then removed a second time, sieved and re-used. (Attrition was not found to be a serious problem. A 2 kg batch of sand kept at 800°C and fluidised periodically for 168 hours was sieved at the end of the period and found to contain 8.9 % wt of undersize material).

6.5 Results - Y-Jet Atomiser

The results of a preliminary filtration experiment at one set of conditions are presented in Table 6.2. Calculated grade efficiencies are plotted in Figure 6.8 where the values are compared with those to be expected if retention were complete, computed using the theoretical model for fluidised bed filtration described in Appendix A1. The "retention efficiency", calculated by dividing the measured collection efficiency by that predicted for complete retention, is also shown in Figure 6.8. The inferred retention efficiency falls off sharply for particles larger than about 1μm. This is similar to effects observed by Coury (1983) in fixed bed filters, where it was concluded that incomplete retention of larger particles occurred because these particles are collected by inertial impaction and yet may impact with kinetic energy greater than the adhesion energy associated with van der Waals' forces so that they simply rebound and are removed from the system (see also Section 2.1.2). Thus Figure 6.8 suggests that retention is still limiting due either to use of an ineffective retention aid or to inadequate coverage.

During subsequent experimental runs with the Y-jet atomiser serious nozzle blockages occurred, due to premature cracking of the oil in the nozzle. Increasing the cooling air flow failed to prevent the problem. It was therefore decided to design a new nozzle and feed system, to
overcome this problem and also the operating difficulty that in order to remove the Y-jet atomiser for cleaning it was necessary to completely shut down and cool the rig, resulting in a delay of at least two days between experiments.

Table 6.2 Results of Filtration Experiments -Y-Jet Atomiser

(using high free-area distributor)

| Expt NO | Temperature (°C) | Bed Mass (kg) | $H_{MF}$ (m) | $U$ (m/s) | $U/\dot{U}_{MF}$ (-) | $U_{OR}$ (m/s) | $\Delta P$ (kN/m²) | Retention Aid Flow (ml/min) | Atomising Gas Flow (l/min) | $C_{IN}$ (kg/actual m³) x 10⁵ | $C_{OUT}$ (kg/actual m³) x 10⁵ | $C_{ATT}$ (kg/actual m³) x 10⁵ | Overall efficiency (%) | Sampling Method |
|---------|----------------|--------------|-------------|---------|-----------------|-------------|-------------|---------------------|----------------------|------------------------|--------------------------|--------------------------|------------------------|--------------|---------------|
| 1       | 800            | 2.0          | 0.07        | 0.525   | 1.75            | 49.7        | 1.41        | 1.2                 | 28.3                 | 1.99                   | 1.41                     | 0.17                    | 26.2                    | Filter       |

*1 Estimated using $\varepsilon_{MF} = 0.41$

*2 $\dot{U}_{MF} = 0.300$ m/s; estimated according to Wen & Yu (1966)

*3 Measured across bed plus distributor

*4 Measured at 70°C

*5 At 15°C & 1 bar a
6.6 Concentric Tube Atomiser Development

For ease of cleaning, it was decided to construct the new atomiser as a single concentric tube, the retention aid feed pipe being in the centre (Figure 6.9). A simple ambient temperature model was constructed and found to work well for water in a fluidised bed of glass ballotini. The coarseness of the atomisation is effectively controlled by the gas flowrate in the annulus. Not surprisingly, the droplet size distribution produced by the concentric tube atomiser is both coarser on average and wider than that produced by the Y-jet, but this is not regarded as a disadvantage in this particular application since larger droplets are captured more efficiently by the collector particles, leading to a reduction in the mass of droplets by-passing the collector bed. Departure from exact concentricity of the gas and
Figure 6.9 Concentric Tube Atomiser
liquid pipes was found to have little effect on the atomisation performance.

A high temperature version of the atomiser was made up in stainless steel. The outer pipe was sized to provide adequate cooling of the oil as it passes through the furnace which surrounds the filter bed, while the diameter was reduced at the hot end to provide an adequate gas velocity for good atomisation in the bed. Between experiments it proved possible, without reducing the bed temperature, to disconnect and remove the central oil feed pipe for cleaning or to rod it out in place. In order to measure the temperature of the oil as it left the nozzle a 0.5mm diameter thermocouple was inserted up the oil feed line to the tip.

Preliminary trials of the new atomiser showed an unacceptably high pressure drop in the oil delivery line. A traverse along the oil line using the fine thermocouple showed this to be due to the oil temperature dropping below 40°C for a portion of its length inside the furnace insulation. This was remedied by electrically preheating the cooling/atomising gas to 70°C.

6.7 Results - Concentric Tube Atomiser

Whilst impossible to monitor the concentric tube atomiser performance directly during a coating run, observation of the bed material after a run indicated a uniform thin coating of cracked hydrocarbon, with no uncoated particles and no agglomerates. No deposits were found on the bed walls or on the distributor. For higher oil feed rates a dark smoke with a strong hydrocarbon odour was observed issuing from the rig exit pipe. At intermediate flowrates the smoke density was observed to
pulsate at the same frequency as the oil feed pump. No smoke was visible at lower oil feed rates. These observations, together with the temperatures recorded at the tip of the oil feed line as the oil flowrate was varied, are recorded in Table 6.3. During these runs the combined pressure drop across the distributor and the bed, and the bed temperature, were continuously monitored in order to detect whether loss of fluidisation occurred. No change in either variable was noted, even at the highest oil addition rate, confirming that loss of fluidisation did not present a problem over the range to be used in the filtration experiments.

Table 6.3 Oil Feed Carry-Over - Concentric Tube Atomiser. (Expt 14)

<table>
<thead>
<tr>
<th>Oil Flowrate (ml/min @ 70°C)</th>
<th>Atomisation Temp. (°C)</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.15</td>
<td>254</td>
<td>Much smoke</td>
</tr>
<tr>
<td>3.45</td>
<td>305</td>
<td>Smoke</td>
</tr>
<tr>
<td>2.80</td>
<td>314</td>
<td></td>
</tr>
<tr>
<td>2.25</td>
<td>335</td>
<td>Smoke barely visible</td>
</tr>
<tr>
<td>1.70</td>
<td>349</td>
<td></td>
</tr>
<tr>
<td>1.15</td>
<td>370</td>
<td>No smoke visible</td>
</tr>
<tr>
<td>0.65</td>
<td>-</td>
<td>Excessive pressure on oil feed line.</td>
</tr>
<tr>
<td>0</td>
<td>410</td>
<td></td>
</tr>
</tbody>
</table>
Results of filtration experiments using the concentric tube atomiser are given in Table 6.4. Experiments were carried out at 700°C, in the hope that the less severe cracking conditions would be less likely to cause nozzle blockages and more likely to assist retention by allowing the cracking fuel oil to remain in a tarry state for longer. These, and all subsequent runs were carried out using the low free-area distributor.

The overall efficiency obtained for no oil addition (expt. 12) is close to that measured under similar conditions by Ghadiri (1980). Plate 6.1 is a scanning electron micrograph of the surface of a sand particle used to collect gasifier fines in the absence of retention aid. Comparison with Plate 3.1, the gasifier fines aerosol collected at the upstream sample point and viewed at the same magnification, shows the almost complete absence of particles larger than about 1.5µm in diameter, confirming that bounce occurs for aerosol particles above this size. However, no useful information on filter performance can be obtained from the filtration experiments 13 and 15 because of the large amount of carry-over of particulates derived from the oil itself.

There is some indication that at least part of the oil-derived material carried over from the filter is liquid condensed from the vapour on cooling, because the pressure drop across the membrane filters used to sample the gases leaving the bed was observed to build up much more rapidly than when the gases contained only solid particles. Scanning electron micrographs of the collection filter and the collector particles corresponding to experiment 13 are presented as Plates 6.2 and 6.3 respectively. The agglomerated state of the aerosol collected on the filter in Plate 6.2a contrasts with Plate 3.1 where the collected particles are distinct. It is not possible to determine on
this evidence alone whether agglomeration took place in the fluidised bed itself or at some point downstream of the bed, although the uneven distribution of material on the micropore filter suggests that agglomeration occurred before the aerosol was collected there. Condensed cracking product can be seen covering the surface in Plate 6.2; this is clearly the reason for the abnormally high pressure drop across the sampling filters. In Plates 6.3a-d it can be seen that the oil contacts the particles before cracking is complete and that the deposit wets the sand surface and flows over it. It appears that cracking then proceeds on the surface itself, liberation of volatiles leading to the formation of a characteristic "pock-marked" appearance. From the range of impact sites observed it appears that the size distribution of the atomised oil drops ranges from about 10 to 100μm. The surface cracking observed in Plate 6.3d may be the result of particle - particle impacts in the bed or it may be due to the rapid quenching which occurs when the sand is extracted from the filter vessel at the end of the run.

The rapid rise in pressure drop across the sampling filters during filtration runs when oil is sprayed into the bed renders this method of sampling unsuitable for prolonged testing. It was therefore decided to adopt a new sampling system, incorporating a liquid impinger, as described in Chapter 3. Although the collection efficiency of the impinger was found to be very high, a glass fibre collection filter was normally placed downstream of each impinger, both to check for any carry-over of particles and to protect the sampling rotameters.
Table 6.4  Results of Filtration Experiments - Concentric Tube Atomiser

(Using low free-area distributor)

<table>
<thead>
<tr>
<th>Expt No.</th>
<th>12</th>
<th>15</th>
<th>13</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature (°C)</td>
<td>700</td>
<td>700</td>
<td>700</td>
</tr>
<tr>
<td>Bed Mass (kg)</td>
<td>2.0</td>
<td>2.0</td>
<td>2.0</td>
</tr>
<tr>
<td>$H_{MF}$ (m)*1</td>
<td>0.07</td>
<td>0.07</td>
<td>0.07</td>
</tr>
<tr>
<td>$U$(m/s)</td>
<td>0.525</td>
<td>0.525</td>
<td>0.525</td>
</tr>
<tr>
<td>$U/U_{MF}$(-)*2</td>
<td>1.66</td>
<td>1.66</td>
<td>1.66</td>
</tr>
<tr>
<td>$U_{or}$(m/s)</td>
<td>175</td>
<td>175</td>
<td>175</td>
</tr>
<tr>
<td>$\Delta P$(kN/m$^2$)*3</td>
<td>6.26</td>
<td>6.02</td>
<td>6.26</td>
</tr>
<tr>
<td>Retention Aid Flow (ml/min)*4</td>
<td>None</td>
<td>1.15</td>
<td>2.25</td>
</tr>
<tr>
<td>Cracking Temperature (°C)</td>
<td>-</td>
<td>344</td>
<td>287</td>
</tr>
<tr>
<td>Atomising Gas Flow (l/min)*5</td>
<td>42.4</td>
<td>42.4</td>
<td>42.4</td>
</tr>
<tr>
<td>$C_{IN}$ (kg/actual m$^3$) x 10$^5$</td>
<td>2.45</td>
<td>3.58</td>
<td>2.25</td>
</tr>
<tr>
<td>$C_{OUT}$ (kg/actual m$^3$) x 10$^5$</td>
<td>2.11</td>
<td>8.80</td>
<td>36.04</td>
</tr>
<tr>
<td>$C_{ATT}$ (kg/actual m$^3$) x 10$^5$</td>
<td>0.55</td>
<td>0.25</td>
<td>0.42</td>
</tr>
<tr>
<td>Overall Efficiency (%)</td>
<td>20.5</td>
<td>-194</td>
<td>-1879</td>
</tr>
<tr>
<td>Sampling Method</td>
<td>Filter</td>
<td>Filter</td>
<td>Filter</td>
</tr>
</tbody>
</table>

*1 Estimated using $e_{MF} = 0.41$

*2 $U_{MF} = 0.316$ m/s; estimated according to Wen & Yu (1966)

*3 Measured across bed plus distributor

*4 Measured at 70°C

*5 At 15°C & 1 bar a.
Plate 6.1 Gasifier Fines Collected on Sand Grains in the Absence of Retention Aid. (Scanning Electron Microscope.)
Plates 6.2a/b  Penetrating Aerosol Collected on 0.2 μm Micropore Filter During Filtration Run With Retention Aid.  
(Scanning Electron Microscope; expt. 13.)
Plates 6.3a/b  Collector Particles Removed From Bed After Filtration Run with Retention Aid.
(Scanning Electron Microscope; expt. 13.)
Plates 6.3c/d  Collector Particles Removed From Bed After Filtration Run With Retention Aid
(Scanning Electron Microscope; expt. 13.)
Using the arrangement described above, with chloroform as the collection liquid, a series of tests was carried out in which the retention aid was sprayed into an empty column.

During the sampling period the following observations were made:

(i) The chloroform collected a quantity of insoluble black particulate material with an upper size of approximately 100µm.

(ii) The chloroform became a distinct yellow/brown, presumably as a result of absorption of light cracking products or (less likely) un-cracked oil.

(iii) The pressure-drop across the glass-fibre filter remained constant but the filter surface became brown, by surface staining. Subsequent SEM examination suggested a thin coating of sub-micron droplets; these would have been able to pass through the impinger without collection.

(iv) A small number of (1-5mm) particulates was observed in the filter off-gases. These are thought to originate from periodic build-up/break-off of deposits on the nozzle; this supposition is supported by the fact that periodic increases in the nozzle atomising gas back-pressure were also observed, each followed by a rapid decrease to the previous value.
After filtering through a 0.2 \( \mu \)m cellulose nitrate filter, the chloroform was analysed at Coal Research Establishment, Stoke Orchard, using gas chromatography. The results of this analysis are presented in Table 6.5.

### Table 6.5 Analysis of Chemical Constituents in Filter Off-Gases

<table>
<thead>
<tr>
<th>Component</th>
<th>Concentration (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tetradecane</td>
<td>1</td>
</tr>
<tr>
<td>Hexadecane</td>
<td>6</td>
</tr>
<tr>
<td>Octadecane</td>
<td>7</td>
</tr>
<tr>
<td>Gicosane</td>
<td>4</td>
</tr>
<tr>
<td>Docosane</td>
<td>2</td>
</tr>
<tr>
<td>Tetracosane</td>
<td>1</td>
</tr>
<tr>
<td>Other alkanes each &lt;1 ppm totalled</td>
<td>2</td>
</tr>
<tr>
<td>Peak eluting just before C16 alkane</td>
<td>2</td>
</tr>
<tr>
<td>&quot; &quot; &quot; &quot; C18 &quot; } possible series</td>
<td>2</td>
</tr>
<tr>
<td>&quot; &quot; &quot; &quot; C20 &quot; } of alkanes or</td>
<td>2</td>
</tr>
<tr>
<td>&quot; &quot; &quot; &quot; C22 &quot; } cycloalkanes</td>
<td>1</td>
</tr>
<tr>
<td>Unidentified peaks</td>
<td>5</td>
</tr>
<tr>
<td>TOTAL ELUTED FROM COLUMN</td>
<td>35 ppm</td>
</tr>
</tbody>
</table>
Table 6.6

Experimental Conditions (Experiments 32-39)

Collector  Silica sand (Table 6.1)

Size  850 - 1000 µm

$U_{mf}$  0.30 m/s (at 800°C)

Mass  3.0 kg

Height (at $U_{mf}$)$^{a1}$  10.5 cm

Dispersion  Gasifier Fines (Figure 3.3)

Retention Aid  3500s Heavy Fuel Oil

Sampling

Below distributor: onto 0.2 µm cellulose nitrate micropore filter

Above distributor: into liquid impinger containing 10% wt NaCl solution.

Particle Characterisation

Coulter Counter Model ZM, using 60µm orifice tube.

Particles > 20µm, removed by fast filtration using a fine nylon mesh.

$^{a1}$ Estimated using $e_{mf} = 0.41$
The experimental conditions for the filtration tests carried out using the liquid impinger sampling train are given in Table 6.6. When particle characterisation is carried out using the Coulter counter, it is most convenient to use the Coulter electrolyte (in this case pre-filtered 10% wt. sodium chloride solution) as the collection liquid with a small amount of dispersant "Nonidet P40" added to prevent re-agglomeration of the suspended particles. A few particles larger than 20μm in diameter were collected in the impinger; these caused particle counting problems when using the Coulter counter due to repeated blockage of the orifice. For all the work reported here, therefore, suspensions were pre-filtered by passing them rapidly through a fine "Simonyl" nylon mesh (Henry Simon Ltd.), a technique which is recommended by the manufacturers of the Coulter counter (Wharton, 1985) and found not to affect the measured size distribution below the mesh size.

Results of the experiments carried out using the liquid impinger sampling system are presented in Table 6.7, and Figures 6.10 - 6.17. In this series of experiments the bed temperature and the gas and oil flowrates were all varied. Results are presented in the form of dust loadings in g/actual m³ and dust mass flowrates in g/hr. In each case these results were obtained by summing the total volume of particles collected in the liquid impinger over a recorded time interval, excluding those particles over 20 μm in size. For each set of experimental conditions three dust loadings are presented, corresponding to the challenging aerosol and the penetrating aerosol for the filtration run, and the penetrating aerosol produced by the addition of oil alone. Overall and grade efficiencies are then calculated from
Table 6.7: Results of Filtration Experiments - Concentric Tube Atomiser
(using liquid impinger sampling method)

<table>
<thead>
<tr>
<th>Experiment Number</th>
<th>34</th>
<th>35</th>
<th>32/33</th>
<th>36/37</th>
<th>38</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature (°C)</td>
<td>800</td>
<td>800</td>
<td>800</td>
<td>800</td>
<td>700</td>
</tr>
<tr>
<td>Oil Flowrate (g/hr)</td>
<td>106</td>
<td>106</td>
<td>106</td>
<td>48</td>
<td>48</td>
</tr>
<tr>
<td>Actual Gas Velocity (m/s)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>In</td>
<td>0.400</td>
<td>0.482</td>
<td>0.579</td>
<td>0.482</td>
<td>0.403</td>
</tr>
<tr>
<td>Out</td>
<td>0.545</td>
<td>0.626</td>
<td>0.722</td>
<td>0.626</td>
<td>0.533</td>
</tr>
<tr>
<td>Distributor Orifice Actual Gas Velocity (m/s)</td>
<td>133.2</td>
<td>160.5</td>
<td>192.5</td>
<td>160.5</td>
<td>134.2</td>
</tr>
<tr>
<td>Pressure Drop Across Bed + Distributor (kPa/m²)</td>
<td>4.35</td>
<td>5.42</td>
<td>6.66</td>
<td>5.42</td>
<td>4.73</td>
</tr>
<tr>
<td>Dust Loadings (g/actual m³) &lt; 20µm</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>In</td>
<td>0.126</td>
<td>0.092</td>
<td>0.087</td>
<td>0.099</td>
<td>0.139</td>
</tr>
<tr>
<td>Out (Oil + GF)</td>
<td>0.048</td>
<td>0.050</td>
<td>0.089</td>
<td>0.012</td>
<td>0.021</td>
</tr>
<tr>
<td>Out (Oil only)</td>
<td>0.026</td>
<td>0.013</td>
<td>0.040</td>
<td>0.007</td>
<td>0.008</td>
</tr>
<tr>
<td>Carry-over*¹</td>
<td>-</td>
<td>-</td>
<td>0.008</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Dust Mass Flowrates (g/hr) &lt; 2 µm</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>In</td>
<td>3.32</td>
<td>2.92</td>
<td>3.28</td>
<td>3.14</td>
<td>3.67</td>
</tr>
<tr>
<td>Out (Oil + GF)</td>
<td>1.70</td>
<td>2.07</td>
<td>4.23</td>
<td>0.48</td>
<td>0.73</td>
</tr>
<tr>
<td>Out (Oil only)</td>
<td>0.94</td>
<td>0.74</td>
<td>1.91</td>
<td>0.28</td>
<td>0.27</td>
</tr>
<tr>
<td>Carry-over*¹</td>
<td>-</td>
<td>-</td>
<td>0.40</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Overall Efficiency (%)</td>
<td>48.6</td>
<td>29.1</td>
<td>-28.8</td>
<td>84.8</td>
<td>80.1</td>
</tr>
<tr>
<td>Overall Efficiency (%) Corrected*²</td>
<td>77.1</td>
<td>54.5</td>
<td>29.5</td>
<td>93.8</td>
<td>87.3</td>
</tr>
</tbody>
</table>

*¹ Sample taken for 30 minutes immediately after oil and aerosol supplies cut off.
*² Correcting for dust mass due to oil alone.
these data, correcting for the additional gas added to disperse the oil feed. Two overall efficiency figures are presented for each set of conditions; both exclude the contribution from particles above 20 µm in size. The first is not corrected for the aerosol produced by the addition of oil alone, i.e.

$$\eta_1 = 1 - \frac{Q_1 c_1}{Q_0 c_0}$$

(6.1)

where \(c_0\) & \(c_1\) are the challenging and penetrating dust concentrations (kg/actual m³) and \(Q_0\) & \(Q_1\) are the corresponding gas flowrates (actual m³). Thus it is possible for \(\eta_1\) to become negative when \(Q_1 c_1 > Q_0 c_0\), i.e. if the mass of aerosol produced by the oil exceeds the mass of gasifier fines retained by the filter. The second overall efficiency figure is corrected for the aerosol produced by the oil alone, i.e.

$$\eta_2 = 1 - \frac{Q_1 (c_1 - c_2)}{Q_0 c_0}$$

(6.2)

where \(c_2\) is the dust concentration due to the feeding of oil alone (kg/actual m³). Grade efficiencies are presented in Figures 6.10, 6.14 and 6.16; these are calculated on the corrected basis, i.e.

$$\eta_{i2} = 1 - \frac{Q_1 (c_{1i} - c_{12})}{Q_0 c_{1o}}$$

(6.3)

where the subscript \(i\) refers to the aerosol diameter interval \(d_i\) to \(d_{i+1}\).
Figure 6.10 Effect of Gas Flowrate on Corrected Grade Efficiencies
6.7.1 Effect of Gas Flowrate

Experiments 32/33 to 35 illustrate the effect of varying the challenging gas flowrate to the filter, maintaining the temperature and oil addition rates constant. Figure 6.10 presents the corrected grade efficiencies, $\eta_{i2}$, for those three runs, which are compared with the theoretically predicted grade efficiencies. The scatter in the measurements for particles below 4 $\mu$m in size obscures any difference between the three cases, although the results are close to the theoretical collection efficiencies, indicating good retention under these conditions. For larger aerosol particles, however, the grade efficiency drops to zero and then becomes negative. It is in this region, for particles above about 5 $\mu$m in size, that the filter performance depends much more critically on the gas velocity, as can be seen from the overall collection efficiency figures in Table 6.7, which are calculated on an overall weight basis and are therefore highly dependent on the collection efficiency of larger particles. This is as expected, since, according to the theory summarised in Section 2.1.2, rebound occurs when the kinetic energy of the aerosol particle exceeds a certain value which depends on the material properties of both aerosol and collector and the surface characteristics of the collector. The kinetic energy is proportional to the aerosol diameter to the third power and to the gas velocity squared, so that rebound is much more likely for larger particles and higher gas velocities. Figure 6.10 shows grade efficiencies which are both slightly greater than 100% for the smallest particles and less than zero for larger ones. This behaviour is consistent with a shift in the aerosol size distribution to larger sizes, i.e. fine particles including those formed from the
EXPT. NO. 34
GAS VELOCITY 0.40 m/s
OIL FLOW RATE 106 g/hr
TEMPERATURE 800 °C

- CHALLENGING GASIFIER FINES
+ PENETRATING AEROSOL (GASIFIER FINES + OIL)
- PENETRATING AEROSOL (OIL ALONE)

Figure 6.11 Differential Size Distributions (Expt. 34)
EXPT. NO. 35
GAS VELOCITY 0.48 m/s
OIL FLOWRATE 106 g/hr
TEMPERATURE 800 °C

- CHALLENGING GASIFIER FINES
- PENETRATING AEROSOL (GASIFIER FINES + OIL)
- PENETRATING AEROSOL (OIL ALONE)

Figure 6.12 Differential Size Distribution (Expt. 35)
Figure 6.13 Differential Size Distributions (Expt. 32/33)

EXPT. NO. 32/33
GAS VELOCITY 0-58 m/s
OIL FLOW RATE 106 g/hr
TEMPERATURE 800 °C

■ CHALLENGING GASIFIER FINES
+ PENETRATING AEROSOL (GASIFIER FINES + OIL)
○ PENETRATING AEROSOL (OIL ALONE)
retention aid, are agglomerated, so that they disappear from the lower size bands but are added to the larger ranges. To check whether this is consistent with the data it is necessary to consider the differential size distributions for the individual challenging and penetrating aerosols; these are presented for superficial challenging gas velocities of 0.40, 0.48 and 0.58 m/s in Figures 6.11, 6.12 and 6.13 respectively. The distributions are plotted as mass flowrates in grammes per hour per micron; a property of this representation is that the area under each differential curve is proportional to the total mass flowrate of dust. The shape of the challenging gasifier fines aerosol distribution is, as expected, substantially unchanged over the three experiments, while the mass flowrates of the aerosols resulting from the addition of oil alone, and oil plus gasifier fines increase with increase in superficial gas velocity. It is noticeable, however, that the modal diameter of particles penetrating the filter (gasifier fines plus oil) occurs at 8-9 \( \mu \)m in Figures 6.12 and 6.13, whereas the mode of the challenging gasifier fines distribution occurs at 3-4 \( \mu \)m. In Figure 6.13 the area under the penetrating aerosol distribution is some 30% larger than the area under the challenging aerosol distribution; it is clear that the whole gasifier fines distribution has been shifted to larger sizes before being lost from the filter.

Thus it appears that two modes of operation are possible: at lower gas flowrates the challenging aerosol is retained on the collector; at higher gas flowrates the challenging aerosol is agglomerated and re-emitted from the bed. It appears that the agglomerates are not formed by attrition of the coating from the bed particles because when
the dust feed is cut off but the gas flow is maintained, the concentration of particulates in the off-gas rapidly falls to low values. In experiment number 32/33 in Table 6.7, for example, the off-gases were sampled for 30 minutes immediately after the oil and gasifier fines feeds had been cut off; the measured dust loading was less than 10% of its former value, even at this, the highest superficial gas flowrate used. The agglomerates must therefore be formed and re-entrained rapidly during the cracking process itself. This interpretation is qualitatively consistent with the scanning electron microscope pictures presented as Plates 6.3 and 6.4.

6.7.2 Effect of Oil Flowrate

Corrected efficiency curves for otherwise identical experimental runs carried out using different flowrates of retention aid oil are compared in Figure 6.14. It can be seen that a lower oil flowrate is preferable if it is desired to operate the bed as an agglomerating filter, because the grade efficiency remains fairly high for particles up to 10 μm in size. At the higher oil flowrates the grade efficiency drops to zero at a diameter of 7 μm and grade efficiencies are consistently lower across the range. Differential size distributions for the two cases compared in Figure 6.14 are plotted in Figures 6.12 and 6.15.

6.7.3 Effect of Temperature

The experiments previously considered were carried out only at 800°C. In order to investigate the effect of temperature, a separate run was carried out at 700°C; the resulting grade efficiency curve is presented
Figure 6.14 Effect of Oil Flowrate on Corrected Grade Efficiencies

TEMP. 800°C

EXPT. 36/37
OIL F.RATE 48 g/hr

EXPT. 35
OIL F.RATE 106 g/hr

$E_i$
Figure 6.15 Differential Size Distribution (Expt. 36/37)

- **EXPT. NO.:** 36/37
- **GAS VELOCITY:** 0.48 m/s
- **OIL FLOWRATE:** 4.8 g/hr
- **TEMPERATURE:** 800 °C

- **CHALLENGING GASIFIER FINES**
- **PENETRATING AEROSOL (GASIFIER FINES + OIL)**
- **PENETRATING AEROSOL (OIL ALONE)**

**EQUIVALENT VOLUME SPHERE DIA. d_v (μm)**

**MASS FLOWRATE (× 10^-2 g/hr/μm)**
in Figure 6.16, where it is compared with a similar run carried out at 800°C. The choice of gas flowrate to be used in these two experiments is not entirely straight-forward. As discussed in Chapter 2, under the conditions of interest here, collection is dominated by behaviour in the jet region close to the distributor. Therefore, for these two experiments Thambimuthu's (1980) group NoR, which correlates collection in the jet region (equations (2.18) - (2.20)), was maintained constant. Thus the collection efficiency in the jet region should also have remained constant.

The removal efficiency of the filter at the lower temperature is clearly very different from that at the higher temperature, and this is reflected in the differential size distributions presented in Figure 6.17. Aerosol removal for particle sizes below 5 μm is much poorer than at the higher temperature but removal is essentially complete above 5 μm and no re-emission appears to occur. This is much closer to, but still slightly better than the grade efficiencies achieved by Ghadiri (1980) when using boric oxide as a retention aid. The results imply a longer sticky phase during cracking at the lower temperature, which should improve retention for all sizes of aerosol. However, this will also increase the cohesive forces between the collector particles, resulting in a reduction in entrainment of collector particles into the grid jets at lower temperatures, an effect which cannot, as yet, be quantified. It may be that it is this second mechanism which is responsible for the poorer grade efficiency for particles below 5 μm. Some qualitative support for the idea that cohesive forces between collector particles were more significant at the lower temperature is
Figure 6.16 Temperature Effect on Corrected Grade Efficiencies
EXPT. NO. 38
GAS VELOCITY 0.40 m/s
OIL FLOWRATE 48 g/hr
TEMPERATURE 700 °C

- CHALLENGING GASIFIER FINES
- PENETRATING AEROSOL (GASIFIER FINES + OIL)
- PENETRATING AEROSOL (OIL ALONE)

Figure 6.17 Differential Size Distributions (Expt. 38)
provided by the fact that the atomising gas pressure drop across the atomiser showed more frequent variations during this run, possibly indicating agglomeration around the end of the nozzle.

6.8 Results — Fluidised Bed Filter/Cyclone Combination

A possible second mode of operation of the fluidised bed filter is as a pre-agglomerator to some form of inertial collector, conveniently a cyclone. To investigate this possibility further, a series of runs was carried out with an unheated 0.12m diameter cyclone in series with the filter rig, the cyclone being constructed according to Stairmand's (1951) high-efficiency design (Figure 1.4), but down-rated so that the cut-point occurred at a particle diameter of about 8 μm. Referring to Figure 6.13, for example, this means that collection of unagglomerated gasifier fines in the cyclone would have been negligible. During these runs, only the gasifier fines aerosol entering the filter, the aerosol leaving the cyclone, and the cyclone bottoms were sampled. Results are presented in Table 6.8.

Sampling of the gas leaving the exit of the cyclone was carried out isokinetically using a probe on the centre-line of the exit pipe, downstream from an anti-spin baffle and a mixing plate, according to the recommendations of Stairmand (1951). At the sampling point the exit gas temperature had fallen to around 140°C, which caused condensation products to come out in the sample tube itself, resulting in frequent blockages, which had to be cleared during the run. The mass flowrates of dust recorded in Table 6.8 are therefore not as accurate as those obtained in the previous runs, and efficiencies calculated from them should be regarded as comparative only.
A significant aspect of the results of the filter/cyclone combination tests is that the cyclone bottoms mass flows differ significantly between the filtration runs and runs in which oil alone was added to the filter. Since the cyclone was down-rated so as to have a very poor collection efficiency for unagglomerated gasifier fines this can only be due to a contribution from agglomerated fines. (Plate 6.4 shows agglomerated fines collected from the cyclone during a filtration run.) Following this reasoning further, if the additional mass collected in the cyclone is entirely due to collection of gasifier fines then the percentage of challenging aerosol re-emitted in agglomerated form can be calculated. These figures are also given in Table 6.8. They are in qualitative agreement with the suggestion that higher oil flowrates lead to re-emission of a larger proportion of the challenging aerosol in agglomerated form. The result for 900°C also suggests that the pre-agglomerating mode of operation may be more suitable for use at this higher temperature, since roughly half of the challenging aerosol is re-emitted under these conditions.
Plate 6.4  Gasifier Fines Agglomerates Collected in Downstream Cyclone.  
(Scanning Electron Microscope; expt. 44.)
### Table 6.8: Results of Filtration Experiments - Filter/Cyclone Combination

(using liquid impinger sampling method)

<table>
<thead>
<tr>
<th>Experiment Number</th>
<th>41</th>
<th>42</th>
<th>43</th>
<th>44</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bed Temperature (°C)</td>
<td>800</td>
<td>800</td>
<td>700</td>
<td>900</td>
</tr>
<tr>
<td>Oil Flowrate (g/hr)</td>
<td>106</td>
<td>48</td>
<td>48</td>
<td>48</td>
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<tr>
<td>Actual Gas Velocity (m/s)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>- Into Bed</td>
<td>0.482</td>
<td>0.482</td>
<td>0.445</td>
<td>0.521</td>
</tr>
<tr>
<td>- Out of Bed</td>
<td>0.626</td>
<td>0.626</td>
<td>0.576</td>
<td>0.678</td>
</tr>
<tr>
<td>Distributor Orifice Actual Gas Velocity (m/s)</td>
<td>160.5</td>
<td>160.5</td>
<td>149.2</td>
<td>173.5</td>
</tr>
<tr>
<td>Pressure Drop Across Bed + Distributor (kN/m²)</td>
<td>5.52</td>
<td>5.49</td>
<td>5.36</td>
<td>6.09</td>
</tr>
<tr>
<td>Dust Loadings (g/m³ of gas at bed temp.) &lt; 20μm</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>- Into Bed</td>
<td>0.081</td>
<td>0.073</td>
<td>0.033</td>
<td>0.033</td>
</tr>
<tr>
<td>- Out (Oil + GF) - cyclone exit</td>
<td>0.017</td>
<td>0.010</td>
<td>0.010</td>
<td>0.015</td>
</tr>
<tr>
<td>- Out (Oil only) - cyclone exit</td>
<td>0.015</td>
<td>0.006</td>
<td>0.011</td>
<td>0.004</td>
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<tr>
<td>Dust Mass Flowrates (g/hr) &lt; 20μm</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>- Into Bed</td>
<td>2.55</td>
<td>2.30</td>
<td>3.0*¹</td>
<td>3.0*¹</td>
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<tr>
<td>- Out (Oil + GF) - cyclone exit</td>
<td>0.68</td>
<td>0.40</td>
<td>0.37</td>
<td>0.69</td>
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<td>- Out (Oil only) - cyclone exit</td>
<td>0.62</td>
<td>0.24</td>
<td>0.40</td>
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<tr>
<td>Cyclone Bottoms Mass Flowrates (g/hr)</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>- Out (Oil + GF)</td>
<td>1.05</td>
<td>0.42</td>
<td>0.36</td>
<td>2.07</td>
</tr>
<tr>
<td>- Out (Oil only)</td>
<td>0.22</td>
<td>0.11</td>
<td>0.06</td>
<td>0.60</td>
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<td>Gasifier Fines Collected in Cyclone (%) *²</td>
<td>32</td>
<td>13</td>
<td>10</td>
<td>49</td>
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<tr>
<td>Gasifier Fines Lost From System (%) *³</td>
<td>2.4</td>
<td>6.9</td>
<td>-1.1</td>
<td>17</td>
</tr>
<tr>
<td>Overall Efficiency (%)</td>
<td>73.4</td>
<td>82.6</td>
<td>87.8</td>
<td>77.5</td>
</tr>
<tr>
<td>Overall Efficiency (%) Corrected *⁴</td>
<td>97.6</td>
<td>93.1</td>
<td>101.1</td>
<td>82.8</td>
</tr>
</tbody>
</table>

*¹ Estimated from previous work  
*² Calculated from cyclone bottoms mass flow rates  
*³ Calculated from dust mass flowrates in gas leaving cyclone  
*⁴ Correcting for dust mass due to oil alone
6.9 Conclusions

(1) The concentric-tube atomiser is suitable for addition of heavy fuel oil as a retention aid in high temperature fluidised bed filtration. Horizontal entry below the level of the bed minimises loss of oil-derived particulates directly into the freeboard.

(2) Theoretically-predicted dust removal efficiencies (greater than 99% at 2μm) can be achieved in practice with the addition of a heavy oil retention aid.

(3) The dust removal efficiency is seriously marred by the aerosol originating from the oil addition itself, which, for the experiments reported here, is larger in total mass than the penetrating gasifier fines. However, much of this aerosol is larger than 7μm in size and is therefore easily removable in a downstream cyclone.

(4) Two modes of filter operation are possible: at lower gas and retention aid flowrates the challenging aerosol is collected and retained on the collector; at higher gas and retention aid flowrates the challenging aerosol is agglomerated and re-emitted from the bed in a distribution with a larger mean size.
Further experimental work on gasification particulate removal should employ higher challenging dust loadings (possibly representative of the gas leaving the gasifier and before passage through the primary and secondary cyclones) and should investigate the use of alternative oil types over longer time periods. This will require experiments to be performed on the off-gases from a working gasifier, where the other components of the gas may well contribute to the retention of captured aerosols.
Chapter 7

Conclusions and Further Work

7.1 Conclusions

In this work, heavy fuel oil has been investigated as an aid to retention in a fluidised bed sand filter designed to collect gasifier fines aerosol at 700—900°C. In separate pieces of work, the problems of particle characterisation and modification of fluidisation behaviour due to the addition of liquid to the bed have been investigated under ambient conditions. The conclusions of this work are as follows.

7.1.1 Hot Gas Cleaning

1. The gasification combined-cycle currently represents an attractive proposition for power generation from coal.

2. In gasification combined-cycle, there are significant economic advantages to cleaning the off-gases from the combustor and the gasifier at the turbine inlet temperature, rather than cooling the gases, cleaning and then re-heating.

3. Tolerable turbine inlet temperatures are likely to increase to at least 1300°C before the end of the century, and any hot gas cleaning system must be designed with this eventual objective in mind.
4. No one hot gas cleaning technology has yet demonstrated a clear advantage for the proposed duty. The development of a suitable cleaning method is the single most critical area of uncertainty in the development of the combined-cycle.

5. The degree of certainty with which turbine manufacturers can specify tolerable particulate loadings for the gasifier off-gases is limited by lack of experience; the current best estimate is 4 mg/m$^3$ with 99% wt. <6 µm and none >10 µm (Reed, 1983). Environmental requirements may impose even tighter constraints.

6. The capital cost of the chosen gas cleaning system depends critically on the number and size of the filter vessels required and hence on the superficial gas velocity in the filter; this makes the fluidised bed filter attractive because its superficial gas velocity (≈1 m/s) is an order of magnitude higher than that usable in most of the rival technologies. However, the pressure drop is relatively high (≈ 0.5 bar at 15.5 bar g system pressure) which would increase the operating cost over those of rival technologies.

7.1.2 Fluidised Bed Filtration

1. Filtration efficiency in correctly designed fluidised bed filters is potentially high enough to fulfil the gas cleaning requirements for a gasification combined-cycle plant.

2. High filtration efficiencies can be achieved by using a low free-area distributor, at the cost of higher pressure drop.
3. For such a distributor, most collection occurs in the gas-entry or "jet" region immediately above the distributor, so the bed need be only a few centimetres deep.

4. Although collection efficiencies for micron-sized gasifier fines are high, the retention efficiency in beds of sand is low under the conditions of interest here, i.e. rebound occurs and the aerosol is re-entrained from the bed.

5. The probability of rebound increases rapidly with increase in aerosol size and gas velocity. Retention efficiencies can be improved by making either the aerosol or the collector adhesive. For fly ash, it has been demonstrated that retention efficiencies can be improved by increasing the bed temperature above the sintering point of the ash (Ghadiri et al., 1986); for gasifier fines, retention efficiencies can only be improved by the addition of a sticky "retention aid" to the filter bed.

6. The addition of a retention aid to the bed also causes an increase in cohesion between the collector particles, which may lead to catastrophic defluidisation.

7. Of the mechanical and electrical aerosol collection mechanisms (inertial impaction, diffusion, interception, gravity, and electrostatic attraction), inertial impaction is by far the most significant collection mechanism in fluidised bed filtration.

8. The most penetrating particle size increases with temperature, i.e. separation is more difficult at higher temperatures.
9. Ambient temperature collection in beds of glass ballotini is well represented by Thambimuthu's (1980) correlation; Nienow and Killick's (1983) results for bronze collectors are lower than this correlation would predict, however, which may be because of a different mode of gas entry in their experiments; i.e. bubbling, rather than "jetting".

10. Jet collection is much reduced in beds of angular collector such as sand, because the rate of entrainment of angular particles into the jet is less than for spherical particles.

11. The height of the jet region under the conditions of interest here is well represented by Merry's (1975) equation.

12. The concentric-tube atomiser is suitable for addition of heavy fuel oil as a retention aid in high temperature fluidised bed filtration. Horizontal entry below the bed surface minimises loss of oil-derived particulates directly into the freeboard.

13. Theoretically-predicted dust removal efficiencies (greater than 99% at 2 \( \mu m \)) can be achieved in practice with the addition of a heavy oil retention aid.

14. The dust removal efficiency is seriously marred by the aerosol originating from the oil addition itself, which, for the experiments reported here, is larger in total mass than the penetrating gasifier fines. However, much of this aerosol is larger than 7 \( \mu m \) in size and is therefore easily removable in a downstream cyclone.
15. Two modes of filter operation are possible: at lower gas and retention aid flowrates the challenging aerosol is collected and retained on the collector; at higher gas and retention aid flowrates the challenging aerosol is agglomerated and re-emitted from the bed in a distribution with a larger mean size.

7.1.3 Aerosol Dispersion and Characterisation

1. The fluidised bed aerosol disperser developed by Ghadiri (1980) is suitable for dispersion of gasifier fines at low concentration (<1 g/m³).

2. The Coulter counter is suitable for measurement of the equivalent-volume diameter of gasifier fines particles, but care must be taken to use a low orifice current and an electrolyte with a high conductivity.

3. The Stokes diameter is the appropriate diameter to use in characterising the filtration efficiency of a fluidised bed filter. However, the equivalent-volume diameter lies within 4% of the Stokes diameter for oblate particles of aspect ratio down to about 0.1 and for prolate particles with aspect ratios up to about 60 (Ghadiri et al, 1986) so that, in practice, the difference can be neglected for most practical purposes.

4. Single-particle light-scattering counters severely undersize gasifier fines, due to their absorbing properties, and are very limited in their concentration range. They are therefore unsuitable for use for particle characterisation in gasification systems.
5. The Malvern Fraunhofer-diffraction sizer gives results for gasifier fines which differ according to whether the particles are dispersed in liquid or in a gas; this clearly indicates a refractive index effect. The lower end of the aerosol size distribution used here is too small for the Fraunhofer theory to be applied.

6. The principle behind the Aerodynamic Particle Size makes this a promising technique, but errors in sizing certain non-spherical particles, and concentration limits mean that this instrument is also unsuitable for fluidised bed filter monitoring.

7. The liquid impinger, designed according to the rules laid down for inertial impactors, is a suitable device for collection of aerosols in circumstances where condensation products cause rapid blocking of micropore filters.

7.1.4 Effects of Interparticle Forces on Fluidisation Behaviour

1. Geldart's group C particles are difficult to fluidise because the interparticle forces between them are large compared with their weight; for group B particles, interparticle forces are relatively insignificant. Group A particles are intermediate in properties (including cohesiveness) between groups C and B.

2. The transitions between groups B and A and groups A and C can be described by simple criteria based on assigning a constant value to the ratio between interparticle force and particle weight for each transition (Molerus, 1982).
3. Interparticle forces due to liquid bridges are larger than those due to van der Waals interactions and both are larger than the particle weight for particles below about 1 mm in size (assuming the contact radius to be the radius of the particles). Estimates of both forces depend critically on the values taken for contact curvature, surface separation, and, in the case of liquid bridges, separation rate.

4. A distinction should be made between group A particles, in which bed expansion before bubbling occurs by nucleation and expansion of cavities, and Martin's (1983) group E, in which expansion is homogeneous. In the former group, which applies to gas-solid systems, interparticle forces are essential for the stability of the expanded cavity structure; in the latter, which applies to liquid-solid systems, particle-particle contacts are transient and interparticle forces do not play a significant role. This difference is reflected in the different values of n, the Richardson-Zaki index, for liquid-solid beds and group A gas-solid fluidisation.

5. The observed stability of expanded group A powders can be predicted from classical stability analysis (Jackson, 1985) by introducing an "elasticity", E, of the expanded particulate structure arising from semi-permanent particle-particle contacts (Rietema and Mutsers, 1973). However, values of E estimated from material properties are very much larger than those necessary to stabilise the expanded structure.

6. Addition of surface liquid to a gas-fluidised bed generally causes an increase in minimum fluidisation velocity, due to the formation of a looser bed structure, of increased voidage. The increase in interparticle force due to the presence of the liquid enables this looser bed structure to remain stable. (Addition of micron-sized fines to the bed can reduce the liquid bridge forces and so reduce the minimum fluidisation velocity again.)
7. Similar increase in minimum fluidisation velocity has been observed for fluidised beds operating above the sintering temperature of the particles, and for particles affected by other attractive forces.

8. There is, as yet, no satisfactory theory for the prediction of minimum fluidisation (or defluidisation) velocities for gas-solid fluidised beds containing liquid or operating above the sintering temperature.

9. When a light involatile oil is added gradually to a fluidised bed of glass ballotini until "wet quenching" occurs

(i) quenching is gradual, with no sudden defluidisation point detected;
(ii) quenching is always accompanied by a reduction in pressure drop across the bed, varying in these experiments from 10% to 70% of the original (dry bed) pressure drop;
(iii) bed pressure drop traces usually show a shallow minimum before settling down to a steady value representing the pressure drop for a fully quenched bed;
(iv) bed pressure drop traces often show a small maximum just before evidence of quenching can be seen, and the magnitude of pressure fluctuations is reduced;
(v) quenching occurs first closest to the distributor, and the pressure gradient remains smaller lower in the bed after quenching is complete.

10. The suspended bed experiment (Buysman and Peersman, 1967) is a relatively simple way of measuring artificially-enhanced interparticle forces directly, provided that the interparticle force is of the same order as the particle weight and a sample of non-cohesive but otherwise identical material is available.
Increasing interparticle cohesive forces in a bed of group B material by loading the particles with an involatile liquid causes the fluidisation behaviour to move from group B through group A to group C. These observations are qualitatively similar to other reported effects of interparticle forces, for example the observations of Agbim et al (1971) on the effects of magnetised particles, and are consistent with Molerus' (1982) explanation of fluidisation behaviour in terms of interparticle cohesive forces. The transition from group B to A is observed to coincide with the transition between "raining" and "plug-fall" modes of failure of a bed of solids suspended by an upward gas flow. The results of such experiments confirm the form of Molerus' expression for the B/A transition, but with a different and probably more realistic value for the dimensionless constant involved.

7.2 Further Work

1. There is a clear need for more reliable estimates of the tolerable particulate loadings for the turbine, especially in gasification systems, for which there is very little information available.

2. Fluidised bed gas cleaning needs to be costed out on the same basis as rival technologies to establish whether its high velocity/high pressure-drop characteristics give it a clear advantage or disadvantage over other systems. More attention needs to be devoted to how such a filter would be integrated into the combined-cycle.
3. Further experimental work on gasification particulate removal should employ higher challenging dust loadings (possibly representative of the gas leaving the gasifier and before passage through the primary and secondary cyclones) and should investigate the use of alternative oil types over longer time periods. This will require experiments to be performed on the off-gases from a working gasifier, where the other components of the gas may well contribute to the retention of captured aerosols.

4. The use of fluidised beds for gas cleaning in non-power applications needs to be investigated. In many such applications the problems of rebound from the collector may not be so severe, or the selection of a suitable retention aid may be easier.

5. There is a need for mechanistic models of aerosol collection in the jet region of fluidised beds. Such models, if successful, will aid the general understanding of gas and particle motion in this region.

6. There is still relatively little understanding of how surface properties affect rebound from the collector, and how particular coatings might modify the rebound condition.

7. There is a need for an on-line method of particle characterisation at high temperatures and relatively high concentrations.

8. Further work on the boundaries between Geldart's groups, employing fluids of unusual density and surface properties, will help to resolve the arguments about the transition conditions. Such work should preferably include some measurement of the interparticle forces acting in the system. The contribution of materials scientists to the understanding of interparticle forces would be welcome.
9. The development of a reliable quantitative prediction of wet quenching velocity is necessary for a number of fluidisation applications. Until (and probably after) such a theory is developed, there will be a need for a device which can give a reliable early indication of impending defluidisation.

10. Donsi and Massimilla (1973b) attempted to calculate the maximum tensile stresses in the expanded group A bed, and thereby to estimate the maximum voidage of the expanded bed as a function of interparticle adhesion force. Such a model might usefully be developed to provide a mechanistic basis for Molerus' (1982) expression for the B/A transition.
Theoretical collection efficiencies quoted throughout this thesis were calculated using a two-region model based on that proposed by Clift et al (1981). The bed is divided into two distinct parts: the bubbling bed, and the entrance or "jet" region immediately above the distributor. Collection in the bubbling bed is estimated by means of the model of Clift et al (1981), based on the fluidised bed reaction model of Orcutt et al (1962). Satisfactory models for collection in the jet region do not yet exist; collection is therefore estimated according to the correlation developed by Ghadiri (1980).

A.1.1 Collection in the Bubbling Bed.

According to equation (2.3) the aerosol collection rate per unit bed volume, $R_v$, is given by

$$ R_v = K_v C $$

where

$$ K_v = \frac{3E_t U(1-\varepsilon)}{2d_C} $$

and $E_t$ is the total single particle collection efficiency. Now consider the aerosol-laden gas passing through a fixed filter bed of cross-sectional area A and height H (Figure A1.1). The rate of collection within the filter element at distance h from the inlet is

$$ R_v Adh = \frac{3E_t U(1-\varepsilon)Adh}{2d_C} $$

An aerosol balance over this element gives

$$ AUdc = -R_v Adh $$

Hence,

$$\frac{dC}{dh} = -k_u \frac{A\epsilon (1-\epsilon)}{2dC} = -kC$$  \hspace{1cm} (A1.4)

where $k$ is a rate constant for filtration; an aerosol particle has a probability $kdh$ of being captured on passing through a filter of depth $dh$.

The difference between the fixed-bed filter of Figure A1.1 and a bubbling bed filter such as that illustrated in Figure A1.2 is that some of the gas flow passing any section in a fluidised bed does so in the form of bubbles. Let $\epsilon_B$ be the fraction of bed cross-sectional area occupied by bubbles, so that the area occupied by the particulate phase is $A(1-\epsilon_B)$. According to the "modified two-phase theory" (Lockett et al, 1967) the gas velocity through this area is equal to its value at minimum fluidisation. Hence, the total gas flow through the particulate phase is $(1-\epsilon_B)A\dot{U}_{MF}$, the remainder passing through the bubble phase. Thus, the fraction of fluidising gas crossing any section within the bubble phase is

$$\beta = \frac{U - \dot{U}_{MF}(1-\epsilon_B)}{U} = 1 - \frac{\dot{U}_{MF}(1-\epsilon_B)}{U}$$  \hspace{1cm} (A1.5)

Now, collection only occurs in the particulate phase, the processes causing aerosol to be presented to the collector being convection by the interstitial fluidising gas and transfer from the bubble phase. Both processes are illustrated in Figure A1.3. Transfer from the bubble phase occurs both by diffusion and by convective "throughflow" (Davidson and Harrison, 1963) although Clift et al (1981) have shown that for air-fluidised beds with bubble diameters of order 0.01 m, transfer of aerosols by convection is about two orders of magnitude greater than by diffusion. More complex models of gas exchange between phases have been proposed (e.g. Kunii and Levenspiel, 1969), to account for the fact that gas flowing through a bubble
Figure A1.1 Schematic Diagram of a Fixed Bed Filter.

Figure A1.2 Schematic Diagram of a Fluidised Bed Filter. (no entrance region shown)

Figure A1.3 Transport of Aerosol to Collector Particle.
may recirculate within a limited "cloud" region surrounding the bubble. However, the existence of the "cloud" as a well-defined region in freely bubbling beds has been questioned (Grace, 1971; Clift, 1983) and when applied to fluidised bed reactors, the additional complexity of such models has not led to improved results (Chavarie and Grace, 1975).

Clift et al (1981) showed that the local collection rate per unit volume of particulate phase can be written

$$R_v = \frac{3E(1-e^{\gamma})}{2d_c} \left[ UMFC_p + \frac{1-e^{-Y_{CB}UC_B}}{(1-e_B)^{2}} \right]$$

(A1.6)

which can be compared with equation (A1.1). The two terms in the square bracket represent collection due to convection in the interstitial gas, and transfer from the bubble phase, respectively; $C_p$ and $C_B$ are the aerosol concentrations in the particulate phase and the bubble phase. The dimensionless group $Y$ describes interphase transfer occurring over a bed depth equivalent to one layer of collector particles, and is related to the conventional "cross-flow factor", $X$, used to describe interphase transfer in the whole bed, by

$$Y = Xd_c/(H-h_j)$$

(A1.7)

where

$$X = \frac{K e (H-h_j)/\beta U}{A}$$

(A1.8)

$K$ is the overall interphase transfer coefficient per unit bubble phase volume, $H$ is the overall bed depth, and $h_j$ is the height of the jet region. $X$ can be interpreted as the number of bubble volumes of gas flowing through the bubble during its rise through the bed. If $X$ is considered as the "number of transfer units" then $Y$ is the number of transfer units per layer of collector.
By analogy with fluidised bed reaction (Orcutt et al, 1962) we now consider two limiting cases: plug-flow and complete mixing of the gas in the particulate phase. By mass balance over an infinitesimal element of the filter in the former case, and over the entire filter in the latter, Clift et al (1981) obtained the following expressions for the penetration through the bubbling bed, \( f_b \):

(i) Plug-Flow

\[
f_b = X \frac{r_2 e^{-r_1} - r_1 e^{-r_2} - \frac{(1-\beta)r_1 r_2 (e^{-r_1} - e^{-r_2})}{X(r_2 - r_1)}}{X(r_2 - r_1)}
\]  

where \( f_b \) is defined as the aerosol concentration at exit from the filter, \( C_h \), divided by that entering the bubbling bed region, \( C_j \); \( r_1, r_2 \) are the roots of

\[
(1-\beta)r^2 + \left[ X + (1-\beta)k \right] r + kX(1-\beta e^{-Y}) = 0
\]  

and

\[
k = \frac{3\mu_n (1-\epsilon_{MF})(H-h_1)}{2d_c} = \kappa_{MF}(H-h_j)
\]

(ii) Fully-Mixed Flow

\[
f_b = 1 - \frac{k(1-\beta e^{-X})(1-\beta e^{-Y})}{1 + k(1-\beta e^{-Y}) - \frac{kX(1-e^{-X})(1-e^{-Y})}{X}}
\]

Clift et al (1981) showed that under the conditions of interest here interphase transfer is rapid, so that the interphase transfer coefficient, \( K \), is large, and both \( X \) and \( Y \) become large for a shallow bed. Since transfer is rapid, the bed behaves as though it consists of a single phase, and the appropriate limiting forms of equations (Al.9) and (Al.12) are

\[
f_b = e^{-k}
\]

for plug-flow in the particulate phase, and

\[
f_b = \frac{1}{1+k}
\]
for full-mixed particulate phase. Convective interphase transfer is independent of particle size, so that, if interphase transfer were limiting, penetration through the bubbling bed would also be substantially independent of aerosol size. However, there is ample evidence that penetration in the bubbling bed is strongly dependent on aerosol size (Doganoglu et al., 1978; Thambimuthu, 1980; Tan, 1982), so that filtration efficiency cannot be limited by interphase transfer.

In fact, filtration efficiency for the conditions of interest here is limited by the collection rate constant, \( k \), which, through the single particle collection efficiency, \( E_t \), is strongly dependent on aerosol size.

### Al.2 Gas Velocity in the Particulate Phase

Clift et al. (1981) concluded that the single particle collection efficiency, \( E_t \), in the particulate phase is equal to its value in a fixed bed at gas velocity \( U_{MF} \). However, using the more extensive data of Tan (1982), Clift et al. (1983) showed that the mean speed of interstitial gas relative to the collector particles is higher than \( U_{MF} \), and estimated its value as

\[
\bar{U} = U_{MF} \left[ 1 + 1.5 \epsilon_B^{2/3} \right]
\]

This expression is obtained as follows. Consider a spherical bubble of diameter \( D \). This is, on average, associated with a spherical shell of particulate phase of diameter \( D/\epsilon_B^{1/3} \). The surface area of the upper hemisphere of this spherical shell is \( \pi D^2/2 \epsilon_B^{2/3} \), and, according to Davidson and Harrison (1963), the volumetric gas flow through the bubble is \( 3\pi D^2 U_{MF}/4 \). If all this gas leaves the spherical shell, the average speed through the upper hemisphere is then \( 3U_{MF} \epsilon_B^{2/3}/2 \), which, when added to the
net upward fluidising velocity gives the result quoted in equation (Al.15). This estimate has been used in the expressions for single particle collection efficiency listed in Chapter 2.

Al.3 Computational Scheme.

The FORTRAN program used to compute theoretical values of the overall penetration is presented at the end of this section. The order of calculation is as follows:

1. Input aerosol particle size, gas (air or nitrogen), gas temperature, pressure and viscosity, aerosol density, humidity (for air only), collector (glass ballotini or sand), collector particle diameter and density, bed voidage at minimum fluidisation, superficial gas velocity under operating conditions and at minimum fluidisation, column diameter and bed height at minimum fluidisation, and distributor orifice diameter and number of orifices.

2. Calculate gas density, mean molecular velocity and mean free path.

3. Calculate aerosol diffusivity and terminal velocity.

4. Calculate collector Reynolds number.

5. Calculate bed fraction occupied by bubbles, \( e_B \) - according to Darton (1979).

6. Calculate gas velocity in the particulate phase - according to equation (Al.15).

7. Calculate single particle collection efficiencies due to diffusion, interception, gravity and inertial collection - according to the expressions in Table 2.1.

8. Calculate cross-flow factor, \( X \) - according to Darton (1979) and Davidson et al (1977).

9. Calculate the Stokes number based on the orifice diameter and orifice gas velocity - equation (2.12).
(10) Calculate jet height - according to Merry's (1975) equation (2.10).

(11) Calculate the fraction of gas passing any section in the bubble phase, $\beta$ -equation (A1.15).

(12) Calculate the dimensionless filtration rate constant, $k$ - equation (A1.11).

(13) Calculate penetration through the bubbling bed for plug-flow and fully-mixed particulate phase for calculated values of $X$ and $Y$ - equations (A1.19) and (A1.20).

(14) Calculate penetration through the bubbling bed for plug-flow and fully-mixed particulate phase for limiting case of infinite $X$ and $Y$ - equations (A1.13) and (A1.14).

(15) Calculate terminal velocity of collector particle.

(16) Calculate penetration through the jet region - according to Thambimuthu (1980) correlation for spherical collector (equations (2.14), (2.15) and (2.16)) or Ghadiri (1980) correlation for sand (equations (2.17) and (2.18)).

(17) Calculate overall penetrations for plug-flow and fully-mixed particulate phase.
**** FLUIDISED BED FILTRATION PROGRAM ****

(INCORPORATING ENHANCED GAS FLOW IN PARTICULATE PHASE)

DATAFILE SHOULD CONTAIN FOLLOWING DATA IN FREE FORMAT.
LINE 0: NO. OF AEROSOL SIZES TO BE CONSIDERED (UP TO 20)
LINE 1: AEROSOL SIZE (UM)
LINE 2: GAS (AIR OR N2) (4 CHARACTERS)
LINE 3: TEMPERATURE (K), PRESSURE (BAR), VISCOSITY (KG/M/SEC), AEROSOL DENSITY (KG/M**3)
LINE 4: WET AIR/WET NITROGEN N(0=AIR, 1=N2), NC(0=BALLOTINI, 1=SAND)
LINE 5: COLLECTOR PARTICLE DIAMETER (M), COLLECTOR DENSITY (KG/M**3), VOIDAGE (-)
LINE 6: MINIMUM FLUIDISATION VELOCITY (M/SEC)
LINE 7: SUPERFICIAL GAS VELOCITY (M/SEC)
LINE 8: COLUMN DIAMETER (M), NUMBER OF ORIFICES (-)
LINE 9: BED HEIGHT AT MINIMUM FLUIDISATION (M)
LINE 10: SINGLE ORIFICE DIAMETER (M)

DIMENSION DAE(20)
PI=4*ATAN(1.)
READ(5,*)IITER
READ(5,*)D(AE(I),I=1,ITER)
READ(5,501)GAS
501 FORMAT(A4)
READ(5,*)-T,P,VISC,RHOA
READ(5,*)XO,N,NC
READ(5,*)DP,RHOP,EMF
READ(5,*)UMF
READ(5,*)U
READ(5,*)DT,AN
READ(5,*)HMF
READ(5,*)DOR

DA=AEROSOL SIZE
GAS=AIR OR NITROGEN
T=TEMPERATURE IN KELVIN
P=PRESSURE IN BAR
VISC=VISCOSITY IN KG/M/SEC
RHOA=AEROSOL DENSITY IN KG/M**3
XO=KG WATER/KG AIR
N=0 IF GAS IS AIR, 1 IF GAS IS NITROGEN
NC=0 IF COLLECTOR IS BALLOTINI, 1 IF COLLECTOR IS SAND
DP=COLLECTOR PARTICLE DIAMETER
RHOP=COLLECTOR DENSITY
EMF=VOIDAGE AT MINIMUM FLUIDISATION
UMF=MINIMUM FLUIDISATION VELOCITY
U=SUPERFICIAL GAS VELOCITY
DT=COLUMN DIAMETER
AN=NO. OF DISTRIBUTOR ORIFICES
C=HMF=BED HEIGHT AT MINIMUM FLUIDISATION VELOCITY
DOR=DIAMETER OF EACH DISTRIBUTOR ORIFICE

WRITE(6,212)
WRITE(6,230)
WRITE(6,212)

CALCULATE GAS DENSITY (RHOG), MEAN MOLECULAR VELOCITY (GMV),
AND MEAN FREE PATH (FP), WHETHER NITROGEN OR AIR
IF(N.EQ.1) GOTO 50
RHOG=1.2929*(273.13/T)*(P/1.01325)*((0.6217*(1+X0))/
&(0.6217+X0))
GOTO 51
50 RHOG=1.2506*(273.13/T)*(P/1.01325)
GMOV=SQRT(8.*3.1416E+03*T/P/29.)
GOTO 51
51 FP=VISC/0.499/RHOG/GMV
WRITE(6,200)BASE,T,P,RHOG,VISC,FP
200 FORMAT(//5X,****GAS PROPERTIES****,//5X,A4,' AT',
&F8.2,'K AND',F8.2,' BAR',//5X, 'DENSITY=',F7.4,' KG/MM**3',
&5X,' VISCOSITY=',E11.4,' KG/M/S',//5X, 'MEAN FREE PATH=',E11.4,
&' M')
DO 38 I=1,ITER
DA=DAE(I)*1.E-06
WRITE(6,240)RHOA
240 FORMAT(//5X,**** AEROSOL PROPERTIES****,//5X, 
&'AEROSOL DENSITY=',F8.1,' KG/MM**3',//5X,'DIAMETER',S8.2,' KNUDSEN',
&5X,' CUNNINGHAM',S8.2,' DIFFUSIVITY',S8.2,' SCHMIDT',S8.2,' TERMINAL',//5X,
&'(MICRON),4X,'NUMBER',S8.2,' FACTOR',S8.2,' NUMBER',S8.2,
&'/V E L.',S8.2')
CALCULATE THE FOLLOWING:
FKN=KNUDSEN NUMBER
CSCF=CUNNINGHAM SLIP CORRECTION FACTOR
DIFF=DIFFUSIVITY(M**2/S)
SC=SCHMIDT NUMBER
FND= DIMENSIONLESS AEROSOL PARTICLE DIAMETER
UT=TERMINAL VELOCITY OF AEROSOL(M/S) (STOKES REGIME)
FKN=FP/DA
CSCF=1.+FKN*(2.514+0.8*EXP(-0.55/FKN))
DIFF=(CSCF*(T)*1.380622E-23)/(3.*P/VISC/DA)
SC=VISC/RHOG/DIFF
FND=13.08*RHOG*(RHOA-RHOG)*DA**3/VISC/VISC
UT=CSCF*VISC/DA/RHOG*FND*(1./24.+FND*(-1.7569E-04+FND*(6.9252E-
&7+07-FND*2.3027E-10)))
WRITE(6,201)DAE(I),FKN,CSCF,DIFF,SC,UT
OUTPUT COLLECTOR PROPERTIES
IF(NC.EQ.0)GO TO 21
WRITE(6,231)
WRITE(6,298)
298 FORMAT(///5X,'COLLECTOR: SAND')
GO TO 22
21 WRITE(6,299)
299 FORMAT(///5X,'COLLECTOR: BACCOTINI')
22 WRITE(6,202)DP,RHOP,EMF,UMF,U
202 FORMAT(///5X,'COLLECTOR DIAMETER=',E10.4,' M',8X,
&' COLLECTOR DENSITY=',E10.4,' KG/MM**3',//5X,
&' DENSE PHASE VOIDAGE=',E10.4,'9X,'MINIMUM FLUIDIZATION VEL.',
&'=',E10.4,' M/S',//5X,'SUPERFICIAL VELOCITY=',E10.4,')
CALCULATE BUBBLING BED HEIGHT (H) AND HENCE BUBBLE-VOID FRACTION (EB) ACCORDING TO DARTON (1979). AU=DISTRIBUTOR AREA ASSOCIATED WITH ONE ORIFICE (M**2)

\[ H = \frac{9.81}{A_0} \times \frac{D{T}^2}{(4.0 \times \text{AN})} \]

\[ R = (1.0 + 2.0 \times (U - \text{UMB} \times 0.8 / (H \times \text{HMF}) + 0.4) \times ((4.0 \times \text{SQR}(A_0) / \text{HMF}) \times 0.6)) \]

\[ \text{IF}(\text{ABS}(\text{DIFF}) \leq 0.00001) \text{GO TO 10} \]

\[ \text{GO TO 10} \]

\[ 20 \text{ CONTINUE} \]

EB = (H - \text{HMF}) / H

TRUE GAS VELOCITY IN THE DENSE PHASE OF THE BUBBLING BED, UBB, CALCULATED ACCORDING TO CLIFT ET AL. (1983)

\[ UBB = 1.0 \times (0.0 + 1.5 \times (E \times (2.0 / 3.0))) \]

CALCULATE COLLECTOR PARTICLE REYNOLDS (REP) AND STOKES (ST) NUMBERS BASED ON UBB

\[ \text{REP} = \frac{U_B \times D_P \times \rho_{OG}}{\nu} \]

\[ \text{ST} = \frac{C_{SCF} \times \rho_{OA} \times D_A \times D_A \times U_{BB}}{9 \times \nu \times B_P} \]

CALCULATE SINGLE PARTICLE COLLECTION EFFICIENCY (ETBB) AND OUTPUT ETBB, REYNOLDS NUMBER AND STOKES NUMBER

\[ E_1 = 4.36 / \text{EMF} \times (\text{DIFF} / UBB / D_P) \times (2.2 / 3.2) \]

\[ E_2 = 0.0375 \times SQR(UT / UBB) \]

\[ E_4 = \frac{0.0375 \times SQR(UT / UBB)}{0.62 \times \text{EMF}} \]

ETBB = E1 + E2 + E3 + E4

E1 = DIFFUSION COLLECTION EFFICIENCY

E2 = INTERCEPTION COLLECTION EFFICIENCY

E3 = GRAVITATIONAL COLLECTION EFFICIENCY

E4 = INERTIAL COLLECTION EFFICIENCY

ETBB = TOTAL SINGLE PARTICLE COLLECTION EFFICIENCY AT VEL. UBB

WRITE(6, 205)


WRITE(6, 206) DAE(1), REP, ST, ETBB

206 FORMAT(/3X, 'F10.4, 8X, E10.4, 6X, E10.4, 8X, E10.4')

CALCULATE CROSS-FLOW FACTOR (X) ACCORDING TO DARTON (1979), USING
DAVIDSON'S BUBBLE GAS EXCHANGE ASSUMPTIONS (DAVIDSON ET AL., 1977)

\[ x = (67.4 * \text{UMF} / ((U - \text{UMF}) ** 0.6 * G ** 0.2 * A0 ** 0.1)) * (1 - (4 * \text{SORT}(A0) / (H + 4 * \text{SORT}(A0))) ** 0.2) + (23.5 * \text{EMF} / (1 + \text{EMF})) * (\text{DIFF} ** 0.5 * G ** 0.1 / ((U - \text{UMF}) ** 0.7 * A0 ** 0.2) + (1 - (4 * \text{SORT}(A0) / (H + 4 * \text{SORT}(A0)))) ** 0.4) \]

\[ \text{CC} \]

CALCULATE ORIFICE STOKES NUMBER FOR JET COLLECTION CORRELATIONS

\[ \text{UOR} = \text{VELOCITY THROUGH ORIFICE (M/S)} \]

\[ \text{STOR} = \text{STOKES NUMBER OF ORIFICE BASED ON PARTICLE DIAMETER & ORIFICE VELOCITY} \]

\[ \text{UOR} = \text{UDES} \times (\text{DT} / \text{DOR}) \times 2 \]

\[ \text{STOR} = \text{SCF} * \text{ROH} \times \text{BDA} \times \text{UOR} / 9 / \text{VISC/DP} \]

JET HEIGHT \((HJ)\) ACCORDING TO MERRY (1975)

\[ \text{HBB} = \text{BUBBLING BED HEIGHT (M)} \]

\[ HJ = 5.2 \times \text{DOR} \times (\text{ROG} \times \text{DOR} / (\text{ROG} \times \text{DP})) \times 0.2 \times (1.3 \times (\text{UOR}^2 / \text{INF} \times \text{DOR}^2)) \times 0.7 \times (\text{UOR}^2 / \text{INF} \times \text{DOR}^2) \]

\[ \text{HBB} = H - HJ \]

CALCULATE BETTA ACCORDING TO MODIFIED TWO PHASE THEORY (LOCKETT ET AL., 1967)

\[ \text{BETTA} = (U - \text{UMF} * (1.0 - \text{EB})) / U \]

\[ \text{AK} = \text{DIMENSIONLESS FILTRATION RATE CONSTANT} \]

\[ \text{AK} = (3 \times \text{ETBB} \times (1.0 - \text{EMF}) \times \text{HBB} / (2 \times \text{DP}) \]

WRITE (6, 207) \( \text{H}, \text{HMF}, \text{HJ}, \text{UOR}, \text{STOR}, \text{AK}, \text{X}, \text{BETTA}, \text{EB} \)

207 FORMAT ('(I2,5X,'H=',E10.4,'(M)',5X,'HMF=',E10.4,'(M)',5X,'HJ=',E10.4,'(M)',5X,'UOR=',E10.4,'(M/S)',
5X,'STOR=',E10.4,'(-)',5X,'AK=',E10.4,'(1/S)',
5X,'BETTA=',E10.4,'(-)',5X,'EB=',E10.4,'(-)')

CALCULATION OF PENETRATION IN BUBBLING BED

\[ \text{R1}, \text{R2} = \text{ROOTS OF QUADRATIC FOR PLUG FLOW CASE} \]

\[ \text{F1}, \text{F2} = \text{PENETRATION FOR PLUG FLOW/ FULLY-MIXED-PARTICULATE-PHASE} \]

\[ \text{FL1}, \text{FL2} = \text{LIMITING VALUES OF F1 AND F2 RESPECTIVELY WHEN X AND Y GO TO INFINITY} \]

\[ Z = \text{BETTA} * \text{EXP}(-X) \]

\[ \text{ALFA} = (P / (1.0 - \text{EMF}) \times (1.0 - \text{EB})) \times 1.0 / 3.0 \]

\[ Y = (X \times \text{ALFA} \times \text{DP}) / \text{HBB} \]

\[ Z = \text{BETTA} \times \text{EXP}(-Y) \]

\[ AA = 1.0 - \text{BETTA} \]

\[ BB = \text{X} - (AA \times \text{AK}) \]

\[ CC = \text{AK} \times \text{X} \times (1.0 - \text{ZY}) \]

\[ CC = \text{SORT}((BB \times BB - 4.0 \times AA \times CC) / (2.0 \times AA)) \]

\[ BB = \text{BB} / (2.0 \times AA) \]

\[ R1 = \text{BB} + \text{CC} \]

\[ R2 = \text{BB} - \text{CC} \]

\[ DD = (1.0 - \text{BETTA}) \times R1 \times R2 \times (\text{EXP}(-R1) - \text{EXP}(-R2)) \]

\[ EE = X \times (R2 \times \text{EXP}(-R1) - 1) \times \text{EXP}(-R2) \]

\[ FF = X \times (R2 - R1) \]

\[ F1 = EE \times DD / FF \]

\[ F2 = 1.0 - ((\text{AK} \times (1.0 - Z) \times (1.0 - \text{ZY})) / (1.0 + \text{AK} \times (1.0 - \text{ZY}) - Z \times \text{AK} \times \text{BETTA} \times (1.0 - \text{EXP}(-Y)) \times (1.0 - \text{EXP}(-X) / X)) \]
FL1=EXP(-AK)
FL2=1./((1.+AK)

CALCULATION OF:

RET=TERMINAL REYNOLDS NUMBER OF COLLECTOR PARTICLE
(CLIFT ET AL., 1978)

UTP=TERMINAL VELOCITY OF COLLECTOR PARTICLE AT ENTRY TO FILTER

AND=DIMENSIONLESS COLLECTOR SIZE

AND=4.*RHOG*(RHOP-RHOG)*G*DP*DP*DP/3./VISC/VISC
W=ALOG10(AND)

IF(AND.GT.73.) GO TO 41
RET=(AND/24.)-(1.7569E-04*AND*AND)+(6.9252E-07*AND**3)-(2.3027E-10 &*AND**4)
GO TO 43

41 IF(AND.GT.580.) GO TO 42
RET=10.**(-1.7075+1.33438*W-0.11591*W**W)
GO TO 43

42 IF(AND.GT.1.55E+07) GO TO 44
RET=10.**(-1.31391+1.34671*W-0.12427*W**W+0.006344*W**3)
GO TO 43

43 UTP=RET*VISC/RHOG/DP

CALCULATE PENETRATION IN JET REGION

ANU=ORIFICE NUMBER

ANU=(3.*RHOG*RHOG*UTP**3)/(4.*(RHOP-RHOG)*G*VISC)
PARN=STOR*ANU**((1./3.)

IF(NC.GT.0.) GO TO 47

CORRELATION FOR JET PENETRATION IN SPHERICAL COLLECTOR
(THAMBIMUTHU, 1980)

IF(PARN.GT.0.4) GO TO 45
FJ=1.-3.096*PARN**3
GO TO 48

45 IF(PARN.GT.0.8) GO TO 46
FJ=1.749-2.62*PARN+0.345*PARN*PARN+0.715*PARN**3
GO TO 48

46 FJ=1.-((PARN/(PARN+0.078))**3
GO TO 48

CORRELATION FOR JET PENETRATION IN SAND
(GHADIRI, 1980)

47 IF(PARN.LT.3.163)GO TO 49
FJ=3.163/PARN**1.855
GO TO 48

49 FJ=1.0
GO TO 48

CALCULATE OVERALL PENETRATIONS FOR PLUG FLOW & FULLY MIXED PARTICULATE PHASE

OVERALL PENETRATION P1=PLUG FLOW; P2=FULLY MIXED
PL1=LIMITING P1; PL2=LIMITING P2

P1=FJ*F1
P2=FJ*F2
PL1=FR*PL1
WRITE(6,211)UTP,PARN
WRITE(6,212)
WRITE(6,213)
WRITE(6,297)
297 FORMAT(/5X,'(FULLY MIXED CASE APPLIES ONLY FOR U/UMF>5)')
WRITE(6,209)F1,F2,FJ,P1,P2
WRITE(6,214)
WRITE(6,215)FL1,FL2,FJ,FJ,PL1,PL2
WRITE(6,212)
209 FORMAT(/5X,'PLUG FLOW',10X,'PERFECT MIXING',/5X,'BUBBLING BED P
&ENETRATION',/5X,'(USING DARTON WITH DAVIDSON MODEL)',10X,E10.4,
&13X,E10.4,/5X,'JET PENETRATION',/5X,'(USING THAMBIMUTHU AND GHAD
&IR',/14X,E10.4,13X,E10.4,/5X,'CORRELATIONS'),/5X,'TOTAL PENETRA
&ION',/26X,E10.4,13X,E10.4,/5X,'/!
211 FORMAT(/5X,'TERMINAL VELOCITY OF COLLECTOR PARTICLE =',
&E10.4,/(M/S)',/5X,'ORIFICE NUMBER =',E10.4,'(-)')
212 FORMAT(/5X,80('**'))
213 FORMAT(/5X,'*** RESULTS FOR CALCULATED X AND Y ***')
214 FORMAT(/5X,*** LIMITING VALUES AS X AND Y TEND TO INFINITY ***
&*)
215 FORMAT(/5X,'PLUG FLOW',10X,'PERFECT MIXING',/5X,'BUBBLING BED P
&ENETRATION',/5X,'LIMITING VALUES',/27X,E10.4,
&13X,E10.4,/5X,'JET PENETRATION',/5X,'(USING THAMBIMUTHU AND GHAD
&IR',/14X,E10.4,13X,E10.4,/5X,'CORRELATIONS'),/5X,'TOTAL PENETRA
&ION',/26X,E10.4,13X,E10.4,/5X,'/!
230 FORMAT(/5X,'***** FLUIDISED BED FILTRATION MODEL *****',/
&11X,'VERSION WITH ENHANCED GAS FLOW')
231 FORMAT(/5X,'***** FILTER BED PROPERTIES *****')
38 CONTINUE
STOP
END

PL2=FJ*FL2
Van der Waals Forces

All intermolecular forces are essentially electrostatic in origin, although they manifest themselves in such different ways that sub-classification has become common. Intermolecular forces may arise from the following interactions (Israelachvili, 1985):

- covalent
- charge-charge (Coulomb)
- charge-dipole
- dipole-dipole (Keesom)
- charge-non-polar
- dipole-non-polar (Debye)
- non-polar-non-polar (dispersion forces)
- hydrogen bonding

Dispersion forces arise from the local polarisations produced in molecules by the random fluctuation of electrons. They are therefore due to attractions between transient induced dipoles, and are the only interactions of those listed above which are always present; the other interactions may or may not occur, according to the nature of the materials. The "van der Waals force" is taken to include dipole-dipole, dipole-non-polar, and dispersion forces.

There are essentially two theories for van der Waals attraction between large (i.e. larger than the molecular scale) bodies: the microscopic (Hamaker, 1937) and the macroscopic (Lifshitz, 1956) theories. It would be inappropriate to review these theories here, but the main results are presented below, for the case of sphere-plane and sphere-sphere interactions only, together with some theoretical and experimental estimates for the resulting forces. The form
of these results is of importance in the arguments of Chapter 4, and there has been some confusion in the literature about the values of the forces which are present.

The non-retarded van der Waals interaction energy between two molecules in vacuum is given (Israelachvili, 1985) by

\[ W = - \frac{C}{\infty} \]  
\[ (A2.1) \]

where \( x \) is the distance between the molecules and \( C \) is a constant. Assuming that the interaction energies between all the molecules in one body and all the molecules in the other body are additive and non-interacting, we may integrate \( W \) over both bodies according to Hamaker's approach to obtain the "two body" potential:

\[ W = -\frac{\pi^2 C \rho_1 \rho_2}{6a} R = -\frac{A R}{6a} \]  
\[ (A2.2) \]

Where \( \rho_1, \rho_2 \) are the number of atoms per unit volume in the two bodies;

\( a \) is the surface separation;

\( A \) is the Hamaker constant = \( \pi^2 C \rho_1 \rho_2 \);

\( R = \frac{R_1 R_2}{R_1 + R_2} \) where \( R_1, R_2 \) are the radii of the two surfaces (thus for a sphere/planet interaction, \( R \) is the radius of the sphere).

The more sophisticated Lifshitz theory avoids the problem of additivity by ignoring the atomic structure of the bodies and treating them as continua. The attraction is calculated in terms of bulk properties such as the dielectric constant and the refractive index. However, the results are of the same form as Hamaker's; effectively, therefore, the Lifshitz theory provides a rigorous method for calculation of the Hamaker constant. Values for the Hamaker constant, calculated using the Lifshitz theory, are given in Table A2.1.
Israelachvili (1985) derives the force between two bodies arising from
the Hamaker interaction potential, \( W \):

\[
F_W = -\frac{dW}{da} = \frac{AR}{6a^2} \tag{A2.3}
\]

This is identical to the expression given by Krupp (1967):

\[
F_W = \frac{\tilde{h}w}{8\pi a^2} \tag{A2.4}
\]

where \( \tilde{h}w \) is the Lifshitz constant (=4/3 \( \pi a \))

The expressions above give the van der Waals interaction forces up to the
point of contact between the bodies, although what is meant by "contact" is
unclear because if \( a \) is allowed to go to zero, the interparticle force
becomes infinite. Krupp (1967) takes \( a \) to be \( 4 \times 10^{-10} \text{m} \), which is close to
the average interatomic distance for many common solids. However,
Israelachvili (1985) recommends the use of a "cut-off distance" substantially
less than the interatomic distance when calculating surface energies (i.e. half
the energy required to separate two surfaces from contact to infinity) from
Hamaker/Lifshitz theory. He shows that a "separation" of \( 1.65 \times 10^{-10} \text{m} \)
gives values within \( \pm 20\% \) of experimental surface energy measurements.

Using the appropriate value for \( a \), equations (A2.3) and (A2.4) give the
adhesion force between two rigid spheres. Real particles, however, are not
rigid, and will deform elastically and /or plastically at the contact point,
even under zero external load. The adhesion of elastic spheres is considered
by Johnson et al (1971) who predict that the pull-off force in this case
should be three-quarters of the value for perfectly rigid spheres, regardless
of the area of contact and the elastic modulus of the spheres. The effect of
elastic deformation is thus relatively small. The increase in pull-off force
due to plastic deformation is more difficult to calculate. Krupp (1967) makes
the simplifying assumption that the pull-off force is equal to the sum of the
van der Waals force at the instant of first contact, plus the force acting on the adhesive area subsequently formed. He thus obtains a value for the adhesion force with plastic deformation, but neglecting elastic recovery:

\[ F_{W'} = \frac{h_w}{8\pi a^2} \left( 1 + \frac{h_w}{8\pi a^3 H} \right) \]

where \( H \) is the hardness of the softer solid (N/m²). This is the equation which was used by Massimilla and Donsi (1976) and Molerus (1982) to predict van der Waals forces between fluidised particles. However, both of these papers use Krupp's (1967) value for \( a \), which makes the second deformation term in equation (A2.5) relatively small, especially for hardness values of \( 10^9 \) N/m² (appropriate for silica) and greater. (For most common materials in vacuum, \( A \approx 5 \times 10^{-20} \) J. Therefore \( A/6\pi a^3 H \approx 0.04 \) i.e. the effect of plastic deformation is small). However, if \( a \) is taken to be \( 1.65 \times 10^{-10} \)m, the value of the deformation term is an order of magnitude greater (\( A/6\pi a^3 H \approx 0.57 \) for the same conditions). In view of the uncertainty about the derivation of the deformation term, and the relatively large hardness values of the materials which are normally considered to show Geldart group A behaviour (cracking catalyst for example), the deformation term in equation (A2.5) has been ignored in the original calculations carried out for Chapter 4 of this thesis.
For two identical media interacting across a vacuum (air), calculated according to Lifshitz theory unless stated:

<table>
<thead>
<tr>
<th>Medium</th>
<th>Hamaker Constant (10^{-20} J)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polystyrene</td>
<td>6.6</td>
<td>Israelachvili (1985)</td>
</tr>
<tr>
<td>PTFE</td>
<td>3.8</td>
<td>&quot;</td>
</tr>
<tr>
<td>Fused quartz</td>
<td>6.5 (5-6) *1</td>
<td>&quot;</td>
</tr>
<tr>
<td>Mica</td>
<td>10 (13.5) *1</td>
<td>&quot;</td>
</tr>
<tr>
<td>Metals</td>
<td>30-50</td>
<td>&quot;</td>
</tr>
<tr>
<td>Diamond</td>
<td>26-30</td>
<td>Krupp (1967)</td>
</tr>
<tr>
<td>Graphite</td>
<td>27.5</td>
<td>&quot;</td>
</tr>
<tr>
<td>Silica</td>
<td>26-28</td>
<td>&quot;</td>
</tr>
<tr>
<td>Quartz</td>
<td>7.6</td>
<td>&quot;</td>
</tr>
<tr>
<td>Polypropylene</td>
<td>6.5</td>
<td>Molerus (1982) *2</td>
</tr>
<tr>
<td>Glass Beads</td>
<td>27</td>
<td>&quot;</td>
</tr>
<tr>
<td>Cracking Catalyst</td>
<td>27</td>
<td>&quot;</td>
</tr>
<tr>
<td>Silica</td>
<td>7.6</td>
<td>Massimilla &amp; Donsi (1976) *2</td>
</tr>
<tr>
<td>Alumina</td>
<td>7.6</td>
<td>&quot;</td>
</tr>
</tbody>
</table>

*1 Experimental values in parentheses.

*2 Molerus (1982) and Massimilla and Donsi (1976) quote Krupp (1967) as the reference, although Krupp does not mention any of these materials specifically. Massimilla and Donsi's values look plausible, but Molerus' values for glass and cracking catalyst look high.
The forces acting between two particles due to a liquid bridge between them may be both capillary and viscous in nature, i.e. static and dynamic respectively. In this appendix only the lowest state of saturation, the pendular state (Figure 4.4) is considered. Both capillary and viscous forces are calculated and their magnitudes are compared under various conditions of contact geometry and separation rate.

In general, the capillary force between two particles is the sum of three terms: the axial component of the surface tension force at the solid/liquid/gas interface, the force due to the reduced hydrostatic pressure in the bridge itself, and the buoyancy force due to the partial immersion of each particle. Princen (1968) and Picknett (1969) demonstrated for spheres that both the buoyancy term and the distortion of the shape of the bridge due to gravity can be neglected if the particle size is small, say less than 1 mm.

Consider two identical spheres of diameter 2R, joined by a liquid bridge of half-angle $\beta$ and separated by a distance 2a (Figure A3.1). In the arguments which follow, zero contact angle (perfect wetting) will be assumed, although the derivations are readily extendable to non-zero contact angles (Coughlin et al, 1982). According to the Laplace equation (Shaw, 1980), the reduced hydrostatic pressure within the bridge, $\Delta P$, is given by

$$\Delta P = \gamma \left[ \frac{1}{r_1} - \frac{1}{r_2} \right]$$  \hspace{1cm} (A3.1)

where $r_1$ and $r_2$ are the principal radii of curvature of the bridge, as shown in Figure A3.1, and $\gamma$ is the liquid surface tension. This equation requires that the liquid surface must have constant mean curvature, which means that the radius marked with a dashed line in Figure A3.1 cannot also equal $r_1$. 
However, Fisher (1926) used a toroidal approximation for the shape of the liquid bridge (i.e. assuming that the curvature at the meridian of the bridge is a circle so that the dashed radius is also equal to \( r_1 \)) and this has been shown theoretically (Orr et al, 1975) and experimentally (Cross and Picknett, 1963; Mason and Clark, 1965) to lead to an accurate estimate of bridge strength provided that the particles are smooth and truly spherical. Consider first the case of zero sphere separation (\( a=0 \)), which is the situation considered by Fisher (1926). The axial surface tension force acting at the dividing plane is given by

\[
F_1 = 2\pi r_2 \gamma
\]

(A3.2)

while the hydrostatic force evaluated at the dividing plane is

\[
F_2 = \pi r_2^2 \Delta P = \pi r_2^2 \gamma \left[ \frac{1}{r_1} - \frac{1}{r_2} \right]
\]

(A3.3)

The total capillary force is then given by

\[
F_C = F_1 + F_2 = \pi \gamma \left[ \frac{r_1 + r_2}{r_1} \right]
\]

(A3.4)

By geometry,

\[
r_1 = R(\sec \beta - 1)
\]

(A3.5)

\[
r_2 = R(1 + \tan \beta - \sec \beta)
\]

(A3.6)

Putting \( t = \tan(\beta/2) \), and substituting for \( r_1 \) and \( r_2 \) in (A3.4) the following simple result is obtained:

\[
F_C = \frac{2\pi \gamma R}{1 + \tan(\beta/2)}
\]

(A3.7)

which is also given as equation (4.7). This result differs slightly from that obtained by Adams and Perchard (1985) who argue that it is correct to evaluate the surface tension and hydrostatic forces at the surface of the sphere, so that equations (A3.2) and (A3.3) become

\[
F_1 = 2\pi \gamma \sin^2 \beta
\]

(A3.8)

and

\[
F_2 = \pi r_2^2 \gamma \sin^2 \beta \left[ \frac{1}{r_1} - \frac{1}{r_2} \right]
\]

(A3.9)
i.e. the reduced hydrostatic pressure acts over the projected wetted area. The main effect of this modification is to alter the proportion of the total force which is attributed to each origin; the sum $F_1 + F_2$ remains almost the same as that given by equation (A3.7), as it should if the spheres are at equilibrium. Adding (A3.8) and (A3.9), and substituting for $r_1$ and $r_2$ as before, we obtain the result

$$F_c = 2\pi R\gamma \left[ \frac{t^2-t+1}{(1+t^2)^2} \right]$$

(A3.10)

where $t = \tan(\beta/2)$. This gives values for $F_c$ within a few percent of those given by equation (A3.7) for values of $\beta$ within the range of interest; the discrepancy is attributable to the error in the toroidal approximation.

The variation of interparticle force with increase in the bridge half-angle $\beta$ is shown in Figure A3.2, for the following values of the variables:

- $2R = 922$ $\mu$m (the mean diameter of sand grains used in the experiments reported in Chapter 6)
- $\gamma = 0.072$ N/m (water)
- $a = 0$

Values of the surface tension and capillary forces and the total force are given according to both approaches. It is clear from Figure A3.2 that the total force is virtually independent of $\beta$ at small values of $\beta$; as predicted by equation (A3.7), $F_c$ tends to a limiting value of $2\pi R\gamma$ ($=2.09 \times 10^{-4}$ N in this case). At larger values of $\beta$, $F_c$ reduces slightly. The range of interest of $\beta$ extends to only about $40^\circ$, since the coalescence limits for liquid bridges between spheres are $30^\circ$ and $45^\circ$ for close packed and cubic arrangements, respectively (Coughlin et al, 1982). It is apparent that for values of $\beta$ below about $10^\circ$ the contribution of the surface tension force is negligible.
Figure A3.1 Liquid Bridge Joining Two Spheres, (only one shown; zero contact angle)

Figure A3.2 Variation of Hydrostatic, Surface Tension and Total Capillary Forces with Liquid Bridge Half-Angle, $\beta$. 

$2\pi R \gamma = 2.09 \times 10^{-4} \text{N}$

$2R = 922 \mu \text{m}$

$\gamma = 0.072 \text{ N/m}$

$\alpha = 0$
As discussed in Chapter 4, the decline in $F_C$ with increase in $\beta$, and hence of liquid volume, is counter-intuitive, and Pietsch (1968) attempted to resolve this by considering a separation between the particles, caused by surface asperities. The effect of finite separation distances can be considered using Adams and Perchard's (1985) approach, re-writing equations (A3.5) and (A3.6) as

$$r_1 = R[(1+a')\sec\beta - 1]$$  \hspace{1cm} (A3.11) \\
and  \\
$$r_2 = R[1+(1+a')\tan\beta-(1+a')\sec\beta]$$ \hspace{1cm} (A3.12)

where $a' = a/R$. Figure A3.3 shows the variation in $F_C$ with $\beta$ for half-separations, $a$, of 0, 1 $\mu$m and 10 $\mu$m, illustrating the dramatic difference that even small separations can make to the interparticle force if $\beta$ is small. The predicted total capillary force now shows a maximum at a certain value of $\beta$, which is greater for large sphere separations.

Most particles of practical interest cannot be considered spherical, especially when viewed on a microscopic scale. It is therefore of interest to consider the forces arising from other contact geometries. Figure A3.4 shows the contact between a cone, of half-angle $\alpha$, and wetted length $l$, making contact with a plane. By a similar approach to that employed for spherical particles, neglecting buoyancy, taking zero contact angle and assuming the surface of the liquid describes an arc of a circle in the plane of the paper as shown, the force acting perpendicular to the plane is given by

$$F_C = 2\pi r y \cos\alpha + \Delta P r y^2$$ \hspace{1cm} (A3.13)

where $\Delta P = \gamma \left[ \frac{l - 1}{r} \right]$ \hspace{1cm} (A3.14)

and $y$ is the radius of the cone at the three-phase line.

Now  \\
y = l \sin\alpha  \hspace{1cm} (A3.15)

and  \\
r = l \left[ \frac{\cos\alpha}{1+\sin\alpha} \right]  \hspace{1cm} (A3.16)
Figure A3.3  Variation of Total Capillary Force With Liquid Bridge Half-Angle For Different Values of Sphere Separation, \(a\).
Figure A3.4  Liquid Bridge at the Contact Between a Cone and a Plane.

Substituting in (A3.13) we obtain

$$F_C = \pi y l \tan \alpha [1 + \cos^2 \alpha + \sin \alpha - \cos \alpha]$$  \hspace{1cm} (A3.17)

which is the result obtained by Coughlin et al (1982). For this geometry, therefore, the capillary force is simply proportional to the wetted length and a geometric factor depending only on the cone half-angle. In contrast to the sphere-sphere case, the interparticle force increases monotonically with increase in the volume of the liquid bridge; for \( l = R \), the sphere radius, values of \( F_C \) will be comparable in magnitude.

The capillary forces so far discussed are not the breakage forces; the stability of liquid bridges and the critical separation limits are discussed by Erle et al (1971). However equations (A3.7) and (A3.17) do enable maximum interparticle forces for quasi-static separation to be calculated, which might be expected to dominate the behaviour of wet granular materials. If separation rates are high, however, viscous forces can become appreciable.
Adams and Perchard (1985) give the following equation for the viscous force, \( F_v \), between two spheres of radius \( R \), and separation \( 2a \), being separated at a rate \( 2v \) (where \( v = da/dt \)):

\[
F_v = 3\mu R^2 v/2a
\]  \hspace{1cm} (A3.18)

where \( \mu \) is the viscosity of the (Newtonian) liquid.

The ratio of \( F_v \) and \( F_c \) has been calculated, as a function of the separation rate, for the following variable values:

- \( \beta = 5^\circ \)
- \( a = 1 \) \( \mu \)m and \( 10 \) \( \mu \)m
- \( 2R = 922 \) \( \mu \)m
- \( \gamma = 0.072 \) N/m (water)
- \( \mu = 10^{-3} \) kg/ms (water)

The results are presented in Figure 3.5, showing that viscous forces can become significant by comparison with capillary forces for separation rates above about 1 cm/s if the particle separation is small. However, for larger separations and larger bridge angles, viscous forces only become comparable with capillary forces for separation rates above about 1 m/s.
Figure A3.5  Ratio of Viscous to Capillary Force, $F_V/F_C$, as a Function of Separation Rate.
REFERENCES


