Attrition in Fluidised Bed Jets

A study of the interactions between single particle properties and jet hydrodynamics

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SUMMARY

Attrition in the jetting region of fluidised beds may pose a serious threat to their efficient operation and use. This dissertation presents the results of a mainly experimental study into the interactions between the attrition propensity of fine particles and the hydrodynamics of fluidised bed jets. A number of state-of-the-art techniques have been used to quantify a number of parameters, such as the jet divergent angle, the solids concentration in the jet, and the particle velocity in the jet. These have been used to verify the predictions of a model of the hydrodynamics of fluidised bed jets, which is in turn used in a model of attrition in fluidised bed jets.

From the experiments, a significant effect of the orifice-to-particle size ratio on the jet hydrodynamics has been identified, which in turn affects the extent and mechanisms of particle breakage. When the orifice-to-particle size ratio decreases below a certain limit, which was found to be about 10, the dilute core of the jet ceases to exist close to the jet nozzle. This results in a rapid shearing flow at the nozzle exit. At orifice-to-particle size ratios above this limit the particles can be entrained and accelerated freely into the jet, until they collide with the dense phase on top of the jet. The latter case can be adequately modelled by a combination of normal single particle impact testing and a model of the hydrodynamics of fluidised bed jets. In this case the model predictions for the dependence of the attrition rate on the operating conditions are in good agreement with experimental findings. In the case of small orifice-to-particle size ratios, below the critical value of about 10, further work is needed for a more realistic model of jet hydrodynamics and particle breakage.

The present work shows that results from commonly used standard attrition tests such as the Forsythe and Hertwig and the ISO 5937 tests cannot be extrapolated to different process geometries and conditions.
Voor mijn ouders
Fais ton bien avec le moindre mahl d’autrui qu’il est possible

J.J. Rousseau - Discours sur l’origine et les fondements de l’inégalité parmi les hommes
In June 1994, I visited Professor Mojtaba Ghadiri and explained to him my passion for fluidised particles. He regretted to have to tell me that the main topic of study of his group was the breakage of the objects of my passion. Little effort was required, however, to convince me of the relevance and fascinating nature of this aspect of powder processing, and it must be said that the number of facets of this aspect has never ceased to rejuvenate my passion. One of the main objectives of this thesis is to reflect the enthusiasm with which the team, consisting of no less than 12 members at one time, has worked in close co-operation on the different aspects of powder processing. This thesis presents my share in this team effort, regarding the breakage of fluidised particles. The success rate of grant applications has been such that a challenging number of state-of-the-art facilities could be acquired and put to use in a number of different applications. This has generated a lively interest from industry and various collaborative research projects have resulted from this. As each project required different expertise, the team members involved changed every time. I recall with great pleasure my first particle sizing adventures with the Coulter Counter and Dimitris, whom I must thank for teaching me how to perform the single particle impact test, and for his comments on the interactions among group members, which have helped to lubricate sometimes rusty relationships. The set-up and maintenance of the high speed camera and the image analysis facilities have taken a large part of my time in the last years, but the successful application in my PhD and in numerous other projects, in which I supported a large number of colleagues has been a very satisfying challenge and experience. I thank Pedro Arteaga, Farideh Bassam, Craig Bentham, Charlotte Couroyer, Wenli Duo, Zemin Ning, Dimitris Papadopoulos, Jesús Subero, and Shi-Hong Zhang for their patience and support during these years.

It is customary to acknowledge the support of your supervisor in a section like this, but here it is anything but a custom, as our relationship has never had a bad moment. Let it be noted first of all that Professor Mojtaba Ghadiri is a man of his word. He is also a master in the art of dosing challenges and providing new challenges at the right times, i.e. very frequently. Apart from thanking him, I feel a certain need to
apologise, as it has been a tough time on both of us living through these challenges. One of the challenges has been the format of this thesis, which is in the form of a short thesis, followed by a number of our publications appended to it, as approved by the University. This has been made possible also by the fact that these publications have in some way or other been subjected to critical review of independent referees. Another challenge has been a number of consultancies, carried out within tight time constraints and with always huge demands on accuracy and consistency, with the BOC project as the most recent example. Here, I should mention also the support of the workshop, who have worked under the same or larger pressures of time and work load.

With great appreciation I mention the support and facilities provided by EPSRC and Rutherford Appleton Laboratories, who manage the EPSRC Equipment Loan Pool, especially Mr Peter Goodyer. Acknowledgements are finally due to Shell Research and Technology Centre Amsterdam and to the Faculty of Engineering of the University of Surrey for the personal grant support that they have provided.

Finally, I want to thank my parents, who went through various ups and downs together and always encouraged me to continue and improve, even though it went against their hearts’ will to know that we were quite far apart.
Attrition in the jetting region of fluidised beds may pose a serious threat to their efficient operation and use. This dissertation presents the results of a mainly experimental study of the interactions between the attrition propensity of particles and the hydrodynamics of fluidised bed jets. A number of state-of-the-art techniques have been used to quantify a number of parameters characterising the hydrodynamics of fluidised bed jets, such as the jet divergent angle and solids concentration, and particle velocities in the jet, in order to gain a fundamental understanding of the influence of jet hydrodynamics on attrition. A significant effect of the orifice-to-particle size ratio on the jet hydrodynamics and consequent effects on the extent and mechanisms of particle breakage have been identified. When the orifice-to-particle size ratio decreases below a certain limit, which was found to be about 10, the dilute core of the jet ceases to exist close to the jet nozzle. This results in a rapid shearing flow at the nozzle exit, whereas at ratios above this limit the particles can be entrained and accelerated freely into the jet, until they collide with the dense phase on top of the jet.

The measured jet parameters have been used as input for the hydrodynamic jet model of Massimilla and co-workers (Massimilla, 1985). Using the results from the hydrodynamic jet model, predictions of the attrition rate could be obtained with the jet attrition model of Ghadiri and co-workers (Ghadiri and Boerefijn, 1996). This model assumes that particle attrition in the jet region can be related to single particle impact breakage, and therefore combines the model of single particle impact attrition of Ghadiri and Zhang (Zhang, 1994) with the jet hydrodynamics model. The jet attrition model has been used to simulate the Forsythe and Hertwig (1949) test method of the friability of fluid cracking catalyst (FCC). This method is used widely in the process industry, and in fact it has been adopted as a dustiness test for sodium perborates in ISO 5937. This test uses a single jet with an orifice size of about 0.4 mm to fluidise a batch of powder for one hour. The predictions from the jet attrition model compare very well with the experimental data obtained in this study for used FCC. For fresh FCC, however, a poor agreement is obtained. In the case of
fresh FCC, the debris consisted of small fines from the surface, whereas the debris of used FCC is clearly caused by necking of microspheroids from secondary agglomerates. The shape of the debris for the latter is similar to that from single particle impact damage, as observed in the study of single particle impacts of these materials. The breakage mechanism of fresh FCC is therefore clearly different from that occurring in the single particle impact test. This suggests that the use of the single particle impact model in conjunction with the jet hydrodynamics model may not be valid for small orifice-to-particle size ratios, as the fresh FCC is susceptible to the rapid shearing flow, and that surface abrasion, rather than necking, is the dominant breakage mechanism in this case. This also suggests that the Forsythe and Hertwig test and the ISO standard test are not useful for the prediction of the performance of materials in different hydrodynamic conditions, or to compare the performance of different materials in the same conditions, if for example the size range is so different that the same hydrodynamic conditions do not prevail in the jet region.

An extensive study of the breakage mechanism of composite materials and agglomerates has accompanied the above jet attrition studies, which has yielded some insight on the breakage mechanisms of these materials. Impact damage studies included the breakage of weak lactose agglomerates, used in inhalation techniques. These weak agglomerates were shown to fail in a mode, similar to the ductile mode for solids, with a large degree of plastic deformation. The dependence of the extent of breakage on the impact velocity was shown to follow the model of Ghadiri and Zhang (Zhang, 1994), which assumes a semi-brittle failure mechanism. Ning et al. (1997) applied the Distinct Element Method to simulate the impact breakage of these agglomerates, and good agreement was found between the simulation results and experimental data. The single particle impact breakage of fresh and used FCC also followed the squared impact velocity dependence, but breakage occurred by means of fracture of microspheroids, originating from secondary agglomeration during spray-drying. The relationship between the velocity dependence of the extent of breakage and the breakage mechanism is therefore not unequivocal, and the velocity dependence is not sufficient to ensure similarity in breakage mechanism. The jet
attrition model should therefore be extended to include parameters which depend more clearly on the breakage mechanism. A characteristic of the breakage product size distribution might be of use here, but a large amount of further work is required to identify the dependence of the size distribution on the breakage mechanism.

Some aspects of the time-dependence of the attrition rate and of the effects of chemical reactions on the attrition rate have also been investigated. Material conversion due to chemical reaction causes differences in particle structure and properties, and hence the attrition propensity. This is a dynamic and rate limiting process, where the competition between attrition and chemical reaction determines the degree of conversion and the extent of attrition. The physical significance of attrition rate constants, used to model the relation between conversion and attrition, and their dependence on material properties and process parameters remains unclear.

The effect of the presence of fines, generated by attrition, on the quality of fluidisation is still poorly understood. In the present study, the presence of fines was found to significantly affect the bubble structure in shallow fluidised beds. The amount of fines varied significantly with the operating parameters in the fluidised bed attrition tests of the Forsythe and Hertwig type. The bed inventory contained sometimes up to 60% fines, which shows that the elutriate may contain only a fraction of the total amount of the attrition products. The interactions between the particle size distribution and the quality of fluidisation need to be clearly quantified in order to justify and validate the use of the elutriate as an indicator of the attrition rate.

In order for the present jet attrition model to obtain validity in the region of small orifice-to-particle size ratios for materials, which are susceptible to surface abrasion and time effects such as work-hardening, the single particle impact test data may be substituted by data from repeated impacts and impacts at an inclined target. Little is known at this stage, however, of the breakage mechanism at inclined targets and of the effects of work-hardening on the attrition propensity of particles, for which no model has yet been developed. The Distinct Element Method may provide a
convenient solution to some of these aspects, as it can model both the surface abrasion and the fracture mechanisms and could form a topic for future work.
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NOMENCLATURE

Roman Symbols

\( b \)  \quad \text{Width of Jet Boundary Layer (m)}
\( d_o \)  \quad \text{Orifice Size (m)}
\( d_p \)  \quad \text{Particle Size (m)}
\( Fr \)  \quad \text{Froude Number (-/-)}
\( Fr^* \)  \quad \text{Two-Phase Froude Number, based on particle size (-/-)}
\( g \)  \quad \text{Gravitational Acceleration Constant (m s}^{-2}\text{)}
\( h \)  \quad \text{Attrition Power Index (dependence on orifice size)}
\( H \)  \quad \text{Hardness (Pa)}
\( k \)  \quad \text{Attrition Power Index (dependence on orifice gas velocity)}
\( K_c \)  \quad \text{Fracture Toughness (N m}^{-3/2}\text{)}
\( l \)  \quad \text{Attrition Power Index (dependence on orifice gas velocity)}
\( L_m \)  \quad \text{Jet Penetration Length (m)}
\( m \)  \quad \text{Attrition Power Index (dependence on single particle impact velocity)}
\( n \)  \quad \text{Attrition Power Index (dependence on orifice gas velocity)}
\( R \)  \quad \text{Attrition Rate (-/-)}
\( R_i \)  \quad \text{Single Particle Impact Attrition Rate (-/-)}
\( s \)  \quad \text{Attrition Power Index (dependence on orifice size)}
\( t \)  \quad \text{Attrition Power Index (dependence on orifice size)}
\( u_o \)  \quad \text{Orifice Gas Velocity (m s}^{-1}\text{)}
\( u_p \)  \quad \text{Particle Velocity (m s}^{-1}\text{)}
\( v \)  \quad \text{Single Particle Impact Velocity (m s}^{-1}\text{)}
\( w_{gl} \)  \quad \text{Lateral Entrainment Gas Mass Flow Rate (kg m}^{-2}\text{ s}^{-1}\text{)}
\( W_s \)  \quad \text{Solids Entrainment Mass Flow Rate (kg s}^{-1}\text{)}
\( W_g \)  \quad \text{Gas Mass Flow Rate (kg s}^{-1}\text{)}
\( x \)  \quad \text{Vertical Distance above Jet Nozzle Exit (m)}
\( y \)  \quad \text{Jet Half-width (m)}
**Greek Symbols**

\( \theta \)  
Jet Divergent Half-Angle (-/ -)  

\( \xi \)  
Single Particle Impact Attrition (-/ -)  

\( \xi^* \)  
Relative Boundary Layer Thickness (-/ -)  

\( \rho_p \)  
Particle Density (kg m\(^{-3}\))  

\( \rho_g \)  
Fluid Density (kg m\(^{-3}\))
1. INTRODUCTION

1.1 Attrition in Fluidised Beds

Attrition in fluidised beds has been recognised as a threat to an efficient and environmentally responsible use of fluidised bed operations already at the start of the application of fluidised bed operations in the process industry. Forsythe and Hertwig (1949) devised a test based on a single fluidised bed jet to estimate the friability of fluid catalytic cracking powder, a material widely used in oil refineries. In a review of fluidised bed attrition studies, Bemrose and Bridgwater (1987) attribute the breakage of particles in fluidised beds mainly to particle-particle and particle-wall collisions. The mechanisms of particle breakage depend therefore mainly on the material response to hydrodynamic stresses in relation to geometrical constraints and operating conditions (Seville et al., 1992). Zenz (1971) identifies three areas in fluidised bed operations which contribute mainly to particle breakage, namely i) the exhaust separator cyclones, ii) the dense bulk, and iii) the jetting region above the distributor grid, where the latter two are confined within the bed.

Let us consider as an example a typical cracking unit operation, as illustrated in Figure 1, with a solids load of about 500 tonnes. The oil feed is introduced into the riser and joins the flow of regenerated catalyst up into the reactor. The endothermic cracking reactions already take place in the high velocity (about 20 m s\(^{-1}\)) riser, with a solids flux of about 50 tonnes per minute. In the design in Figure 1, the reactor is a dilute phase fluidised bed. Many other designs of FCC units include a fast fluidisation reactor riser, featuring a terminator, where particles may impact at velocities of about 20 m s\(^{-1}\) to be subsequently deflected into the cyclones (Sesti Osséo, 1996). After the reactor, the particles move back to the regenerator, where the coke deposit on the particle surface is burnt off. The regeneration process thus generates the heat of reaction for the cracking process. While moving from the reactor to the regenerator, the particles may pass through cyclones and, depending on the design of the unit, may pass through a grid before entering the dense fluidised bed.
of the regenerator. Before reaching this grid, the particles may encounter an impact plate, intended to enhance particle distribution in the area below the grid. Typical particle velocities here are about 25 m s\(^{-1}\), and velocities are about 50 m s\(^{-1}\) at the grid holes of a few centimetre in size, spaced at a triangular pitch of about 0.5 m. Apart from suffering mechanical stresses, the particles also participate in chemical reactions and undergo a certain degree of surface sintering. An average daily amount of 4-8 tonnes of fines is lost mainly due to attrition in a unit of this type.

![Figure 1. Fluid Catalytic Cracking Unit.](image)

Other examples of processes in which particle attrition may be significant are fluidised bed drying (see e.g. Deiva Venkatesh et al., 1996), combustion (see e.g. Chirone et al., 1991) and desulphurisation units (see e.g. Lee et al., 1993). Fluidised jet mills are commonly used in the pharmaceutical industry in comminution processes (Alpine, 1991; Bentham et al., 1996). In these units very often opposing air jets impinge on the fluidised suspension, or gas-accelerated particles impinge on a fixed target wall (BMHB, 1987).
1.2 Jets in Fluidised Beds

Within the confines of the fluidised bed, most of the breakage will take place in the jetting region, as particles are subjected here to large velocity and solids concentration gradients. The jet generally consists of a fast moving gas stream penetrating the dense phase and drawing in particles from the surrounding dense phase, which are subsequently accelerated. The jet may be intermittent, and unstable, as bubbles depart from its top, and it may sway from side to side, especially in two-dimensional configurations (Caram et al., 1984). There is widespread controversy in the literature on the nature and appearance of fluidised bed jets (Grace and Lim, 1987; Yates et al., 1986). Studies of jet hydrodynamics have concentrated mainly on the jet geometry, i.e. the jet penetration length (Yates et al., 1986; Merry, 1975) and the angle of divergence into the dense phase (Vaccaro, 1997; Massimilla, 1985; Cleaver et al., 1995). Recently, X-ray studies of jets in fluidised beds have been carried out, but so far only qualitative classifications have resulted from this, which are not yet related to ranges of operating conditions (Gilbertson and Yates, 1998).

In the present study, it will be shown that the structure of the jet is of crucial importance to the particle breakage mechanism. For this purpose, a comprehensive study of jet hydrodynamics has been carried out in this work, based largely on image analysis techniques, concentrating on the measurement of jet angles, particle velocities and solids concentration profiles in the jet. Part of this data is used as input to the hydrodynamic model of fluidised bed jets, devised by Massimilla and Donsk and co-workers (see e.g. Massimilla, 1985). This hydrodynamic jet model has been used by Ghadiri and co-workers in a model of attrition in fluidised beds (Ghadiri et al., 1992). The second component of this model, characterising the breakage, is given by a model of single particle impact breakage, in which particles are accelerated and impact singly on a rigid target at a known velocity. The underlying assumption is that particles, as they flow into the jet, accelerate freely in the dilute core of the jet until they hit the dense phase on top of the jet. Thus, the breakage mechanism in a jet is similar to that occurring in a single particle impact and the amount of breakage can be characterised conveniently on a lab-scale by
single particle impact tests. A large part of the present work concentrates on the verification of this assumption, the characterisation of the solids concentration profiles and divergent angles of the jet and, more importantly, the mere existence of the dilute jet core.

The jet attrition model incorporates a model of single particle impact breakage. This presupposes the similarity of the impact breakage mechanism in the jet and a single impact on a rigid target. Early observations of the jet have indicated that the existence of a dilute jet core depends on a number of conditions, such as the orifice-to-particle size ratio, the orifice gas velocity and the background fluidisation level.

The jet attrition model is described more extensively in Appendix A, but its main features will be summarised here. As mentioned above, attrition in the jet involves the entrainment of solids into the dilute jet core at a rate $W_s$, and the particles will then accelerate to a velocity $u_p$, at which they collide with the dense phase on top of the jet. Similar to all other models of attrition in jets, as shown in Table 1 of Appendix A, the model assumes that the dependence of the attrition rate in the jet on the main design parameters, the orifice gas velocity, $u_o$, and the orifice diameter, $d_o$, can be expressed in a power law:

$$ R \propto u_o^n \, d_o^h $$

Similar power law expressions can be obtained for the relevant hydrodynamic parameters, $W_s$ and $u_p$:

$$ W_s \propto u_o^k $$

$$ W_s \propto d_o^s $$

$$ u_p \propto u_o^l $$

$$ u_p \propto d_o^t $$

The model of jet hydrodynamics of Massimilla and co-workers (1985) can predict values for $W_s$ and $u_p$ for given $u_o$ and $d_o$. We now assume that the jet attrition rate $R$ is linearly related to the entrainment rate, $W_s$, and the single particle impact attrition rate, $R_i$, which is a function of the particle velocity $u_p$, related via a power law
expression with exponent $m$. Therefore we arrive at the following overall expressions for the jet attrition rate as a function of $u_o$ and $d_o$ by substituting Eqs 2-5 into Eq. 1:

$$R \propto W_s R_i \propto u_o^k u_p^m \propto u_o^k (u_o^l)^m \propto u_o^k u_o^l u_o^m \propto u_o^n$$

where $n = k + l m$. The same approach may be followed for the effect of the orifice diameter, $d_o$, and after normalisation of $W_s$ with the gas mass flow rate $W_g$:

$$R \propto \left(\frac{W_s}{W_g}\right) R_i \propto d_o^s u_p^m \propto d_o^s (d_o^l)^m \propto d_o^s d_o^l d_o^m \propto d_o^h$$

where $h = s + t m$. Figure 2 illustrates the interactions of the above equations. The values of $k$, $l$, $s$, and $t$ can be obtained from the hydrodynamic jet model, whereas the value of $m$ is supplied by the analysis of the breakage behaviour of single particles.

\[\text{Figure 2. Schematic of the jet attrition model.}\]
1.3 Single Particle Impact Breakage

The single particle impact model, mentioned above and used in the jet attrition model, has been developed by Ghadiri and co-workers (Zhang, 1994; Papadopoulos and Ghadiri, 1996). Based on the assumption that the particles fail in a semi-brittle failure mode, where cracks form around a plastic deformation zone and subsequently propagate through the particle as subsurface lateral or median vent cracks, this has led to the following model equation relating the single particle breakage rate to the impact velocity and the mechanical properties of the particle that determine its attrition propensity:

$$\xi \propto \frac{\rho_p \nu^2 d_p H}{K_c^2}$$  \hspace{1cm} (8)

where $\nu$ is the impact velocity, $\rho_p$ the particle density, $d_p$ the particle size, and $H$ and $K_c$ the hardness and fracture toughness, respectively. The latter two mechanical properties are not easily measured for small particles, especially for agglomerates and composite structures. A special nano-indentation technique has been developed to characterise these parameters (Arteaga et al., 1996), but it has not yet been applied to FCC. It has recently been shown that Equation 8 successfully describes the attrition propensity of a wide range of materials, e.g. FCC (Duo et al., 1996), lactose and paracetamol (Bentham et al., 1997, 1998), porous silica and ammonium nitrate prills (Papadopoulos, 1998) in different size ranges.

In addition to common salt, the main focus in the present work has been on FCC particles, which are produced by spray-drying. A large portion of these particles is non-spherical, as they assume ginger-root shapes due to secondary agglomeration in the spray-drying process. The breakage mode of agglomerates is a function of both the internal structure and external shape, and is not clearly understood. In the present work, single particle impact tests have been carried out to characterise the breakage mode of FCC particles, and also to supply the necessary quantitative input for the jet attrition model. Furthermore, in order to enhance our understanding of agglomerate behaviour, the experimental programme of impact tests has been accompanied by simulations using the Distinct Element Method, as reported by Ning et al. (1997).
The simulations are not directly related to the FCC behaviour, as they address the breakage of very weak particles. Nevertheless, the simulations show the important effect of structure on the failure mode of agglomerates.

### 1.4 Current Focus

In the following, the interactions between single particle properties, such as particle size, structure and surface topology, and the geometry and hydrodynamics of fluidised beds and jets, will be addressed in order to gain a fundamental understanding of breakage processes in fluidised environments.

The work outlined in the previous sections has resulted in a number of publications, most of which are appended to this dissertation. Rather than reiterating all the findings already presented elsewhere in these appendices, a general discussion of the matters presented in these papers, and a critical review will be presented, as agreed by the University of Surrey. Table 1 presents an overview of the papers in the appendices, highlighting the aspects dealt with in each of them. A number of phenomena have been characterised in this work, namely (i) the influence of the orifice-to-particle size ratio on the jet hydrodynamics and on the breakage mechanism in the jet. This was done in order to elucidate the relevance of the Forsythe and Hertwig (1949) test to assess the attrition propensity of fine powders, following the observation of the absence of a dilute jet core in the case of small orifice-to-particle size ratios; (ii) the interaction between the particle structure and strength and the breakage mechanism in a fluidised bed jet and in single particle impact. This topic received attention following the observation of the susceptibility of the fresh FCC to surface abrasion and of the used FCC to necking in fluidised bed jets with small orifice-to-particle size ratios.

In this dissertation, a number of aspects, which have not been addressed in the published papers, will be discussed. These are the effects of time on fluidised bed attrition tests and the use of the elutriate as the main indicator of the extent of
attrition. Also, some further attention will be paid to the use of the jet hydrodynamic model and especially to the specification of the many input parameters which are required. The hydrodynamic model can benefit from a recent publication by Vaccaro (1997) on the jet angle. The analysis carried out by Vaccaro (1997) will be discussed for its relevance to the effect of the ratio of orifice-to-particle size. Finally, a more in-depth discussion of the role of the single particle impact model within the jet attrition model will be presented.
Table 1. Overview of appendices and brief summary of subjects and arguments.

<table>
<thead>
<tr>
<th>App.</th>
<th>Paper Title, Publication</th>
<th>Subjects</th>
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<tbody>
<tr>
<td>A</td>
<td>A Model of Attrition in the Jetting Region of Fluidised Beds <em>KONA Powder and Particle</em>, Vol. 14, pp. 5-15, 1996</td>
<td>Summary of attrition model concept, which comprises a model of the single particle breakage, which is coupled with a hydrodynamic model of the jet. Together, this enables the prediction of the dependence of the attrition rate on operating parameters such as the orifice gas velocity and the orifice size.</td>
</tr>
<tr>
<td>C</td>
<td>Impact Attrition of Fluid Cracking Catalyst <em>Proc. 5th Int. Conf. on Multiphase Flow in Industrial Plants</em>, pp. 170-179, 1996</td>
<td>Effect of particle size and impact velocity on single and repeated impact breakage of FCC particles. Both fresh and used particles follow the dependencies on particle size and impact velocity as predicted by the model of Ghadiri and Zhang for semi-brittle failure. However, the breakage mechanism is termed “necking” which is only remotely similar to semi-brittle failure. There is little effect of repeated impacts on fresh FCC particles.</td>
</tr>
<tr>
<td>D</td>
<td>Distinct Element Simulation of Impact Breakage of Lactose Agglomerates <em>Adv. Powder Technol.</em>, Vol. 8, No. 1, pp. 15-37, 1997</td>
<td>Distinct element simulation of weak agglomerates, investigation of the failure mode of composite materials. The breakage propensity of the agglomerates follows the predicted dependence on impact velocity, but the mechanism is not semi-brittle, but more similar to ductile failure with extensive plastic deformation.</td>
</tr>
<tr>
<td>E</td>
<td>Effects of Particle Size and Bond Strength on Impact Breakage of Weak Agglomerates <em>Powders and Grains '97</em>, pp. 127-130, 1997</td>
<td>Investigation of failure mode of agglomerates with multiple primary particle sizes (core/shell) and different levels of surface energy. The effect of structural differences on the breakage propensity of the agglomerates is highly complex.</td>
</tr>
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</table>
F. Disintegration of Weak Lactose Inhalation Drug Excipient Agglomerates  
Submitted to Int. J. Pharmaceutics, 1998  
Investigation of the effect of agglomerate size and ambient humidity on the extent and mechanism of impact damage. In the case of these loose agglomerates, an increase in the agglomerate size results in a decrease in the extent of breakage. This may be related to the dry tumbling process. In the case of increased humidity, the particles acquire a glassy shell, which causes a change in breakage mode from ductile towards brittle, leading to less breakage.

G. High Speed Video Image Analysis of Flow of Fine Particles in Fluidised Bed Jets  
Description of the use of optical techniques and digital image analysis for determination of hydrodynamic properties of fluidised bed jets. Digital high speed video cameras and photographic cameras have been used to track particles in fluidised bed jets. A novel technique of image analysis, based on the relation between the back scatter luminance and the solids concentration, has been used to measure the voidage gradient in a fluidised bed jet.

H. The Effect of Orifice Size on the Breakage of Fluid Cracking Catalyst Particles in Fluidised Bed Jets  
accepted for World Congress on Particle Technology 3, 1998  
Identification of the interaction between particle properties and jet hydrodynamics in the case of small and large orifices. It is shown that the attrition model works well for large orifice-to-particle size ratios. In the case of small orifice-to-particle size ratios, the success of the model depends on the particle resistance to surface abrasion.

I. Interactive Processes of Sorbent Attrition and Chemical Reaction During Fluidised Bed Sulphurisation  
accepted for World Congress on Particle Technology 3, 1998  
Identification of the effects of chemical reaction on the structure and strength of limestone particles within the fluidised bed reactor at high temperatures and in single particle impact tests at low temperatures.

J. Time-resolved Voidage Profiles at the Bed Surface and in the Freeboard of Shallow Fluidised Beds  
accepted for World Congress on Particle Technology 3, 1998  
Characterisation of the effect of changes in the particle size distribution, specifically the fines content, on the expansion behaviour and voidage profile along the height of the bed and in the freeboard. Significant effects of the fines content and shifts in mean particle size on the voidage profile and on the bubbling behaviour have been identified and measured.

K. Analysis of ISO Fluidised Bed Jet Test for Attrition of Fluid Cracking Catalyst Particles  
accepted for Fluidization IX, 1998  
Analysis of a single jet attrition test, using the jet model to describe the flow patterns. A comparison with experimental data shows that the ISO test is not suitable for particles which are prone to surface abrasion, as the breakage mechanism will differ from that occurring in other geometries. This poses a limit on the applicability of the test.
2. **Hydrodynamic Jet Model**

The hydrodynamic jet model of Massimilla and co-workers (1985) calculates the particle flow patterns in the jet assuming a straightforward jet geometry, as illustrated in Figure 3. The jet is modelled as a dilute phase penetrating the dense bulk phase, diverging at a constant angle $\theta$. The model assumes that there are no horizontal solids concentration gradients within the jet and that the solids concentration changes rapidly from the dilute jet core up to the bulk phase concentration within a fraction of the jet width, which is termed the boundary layer. For simplicity, it also assumes that the pressure gradient within the jet is that of the bulk phase.

![Figure 3. Schematic of the jet.](image)

### 2.1 Input Parameters

In addition to particle properties such as the particle density and size, a number of geometrical and boundary conditions of the jet also have to be provided as input for the model:

- the divergent jet half angle, $\theta$
- the jet penetration length, $L_m$
• the relative portion of the jet which belongs to the boundary layer, $\xi$
• the non-dimensional rate of lateral gas entrainment, $\omega_g/(\rho_g u_o)$
• the initial gas velocity at the nozzle, $u_o$
• the bulk phase solids concentration,
• the background fluidisation superficial gas velocity,
• the initial solids velocity at the jet nozzle, $u_{p,i}$
• the initial solids concentration at the jet nozzle, $\gamma_s,i$.

The jet penetration length is of great importance for the design of bed internals, such as heat exchangers. For this reason, this parameter has received considerable attention in the literature, as summarised in a recent review (Kimura et al., 1995). Very little is known, however, about the gas and solids entrainment rates in fluidised bed jets (Massimilla, 1985). Even harder to measure are values for solids concentrations in the jet core, the boundary layer and the surrounding bulk phase. Some data is available on the jet angle (Cleaver et al., 1995; Vaccaro, 1997), but the exact interactions between all these parameters are to date poorly understood.

2.2 Verification of Hydrodynamic Model Predictions for Present Systems

In the present work, some values of solids concentration profiles, jet angles and particle velocities have been measured to verify the validity of the predictions of the jet model and its use in the attrition model. The results are given in Appendices B and G. Two systems have been studied here in a two-dimensional fluidised bed with background fluidisation and a central slot jet nozzle, 425-600 $\mu$m common salt particles (Appendix G), and 75-106 $\mu$m FCC particles (Appendix B). In both systems, particles entrained into the jet have been tracked with high speed video imaging and analysis. With the FCC particles, also the jet angle and solids concentration profiles have been measured (Appendix B). The techniques used to perform the analysis of the flow patterns are described in more detail in Appendix G.
The analysis of particle velocities at two positions in the jet, just at the nozzle exit and higher up into the jet, shown in Appendix G, has revealed an interesting phenomenon. Comparing the velocities at the two positions, it appears that there is virtually no acceleration along the jet height taking place if the orifice is less than about 10 times the particle size. A possible explanation for this was found in the absence of the dilute jet core in the case of low orifice-to-particle size ratios. In the case of small orifices, the jet is reduced to a point source piercing through the dense phase, which does not open up, inhibiting particle entrainment and free acceleration into the dilute jet core. Appendix B shows the results of an extended study of the flow patterns of FCC in the jet, including a measurement of the full solids concentration profile in the entire jet area. From this, the jet angle, the initial solids concentration at the jet nozzle exit and the solids concentration in the boundary layer could be derived and used as input into the jet hydrodynamic model. It was found that the initial solids concentration at the nozzle exit varies with the orifice gas velocity, but little variation with the orifice size was detected. The jet angle was shown to increase with an increase in the orifice size and a decrease in the orifice gas velocity. Using the measured data, a good comparison between the measured and predicted particle velocities has been obtained.

2.2.1 Analysis of model sensitivity to input parameter variations

It is difficult to quantify experimentally a number of input variables, such as the relative boundary layer thickness, $\xi^*$, the initial values for particle velocity, $u_{p,i}$, and concentration, $\gamma_i$, in the jet core, and the non-dimensional lateral solids entrainment rate, $w_{g\|}/(\rho_g u_o)$. A sensitivity analysis of these parameters may help to establish an understanding of their importance in the predictions and whether endeavours should be made to measure them experimentally. The sensitivity analysis has been carried out for a model material with the properties of paracetamol, one of the materials recently included in a parallel study of fluidised bed jet breakage of pharmaceutical powders (Bentham et al., 1998). The material has a density of 1290 kg m\(^{-3}\) and an average particle size of 423 μm and the orifice size is 1 mm. The model exhibits a
higher sensitivity to parameter variations with paracetamol than with materials of finer sizes, such as FCC, especially where the entrainment rate is concerned, hence the use of this material as an example here.

The sensitivity analysis is performed by varying the above four parameters over the ranges indicated in Table 2 for three different orifice velocities, 50 m s⁻¹, 150 m s⁻¹, and 250 m s⁻¹.

<table>
<thead>
<tr>
<th>Table 2. Parameter ranges</th>
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<tr>
<td>( \xi^* )</td>
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<tr>
<td>(-/-)</td>
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<tr>
<td>0.70</td>
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<tr>
<td>0.75</td>
</tr>
<tr>
<td>0.80</td>
</tr>
<tr>
<td>0.85</td>
</tr>
<tr>
<td>0.90</td>
</tr>
<tr>
<td>0.95</td>
</tr>
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</table>

The relative boundary layer width, \( \xi^* \), is defined here as:

\[
\xi^* = \frac{y - b}{y}
\]  

(9)

where \( y \) is the half width of the jet and \( b \) the width of the boundary layer. One parameter was varied at a time, using the values given for each parameter in Table 2, and the remaining parameters were set to the following values: \( \xi^* = 0.9; u_{p,i} = 0.1 \) m s⁻¹; \( \gamma_{s,i} = 0.1; w_{p}(\rho_s u_o) = 0.001 \). Therefore it should be noted that the values, given in Table 2 in rows, have not been used in these combinations in the calculations.
Figure 4 shows typical profiles of the gas and particle velocities along the central axis of the jet. The particle velocity reaches a maximum and then stabilises at a slightly lower value, whereas the gas velocity decays monotonously along the entire jet length. The solids concentration exhibits a local minimum about where the particle velocity has its maximum, which in the following will be denoted as $u_p$. There is a point of inflection in the solids concentration, approximately at $x = 0.005$ m, which is of great importance for the stability of the solution. When the parameters have been wrongly chosen, so that the program becomes unstable, the local minimum will not occur but the solids concentration will continue to decay and may even become negative. This is illustrated in Figure 5, where the instability of the program is caused by setting $\varepsilon^* = 0.70$, instead of $\varepsilon^* = 0.90$, as in Figure 4. Figure 5 also shows that the particle velocity does not remain stable, but exhibits a second, run-away rise towards the top of the jet at $x = 0.007$ m.
Figure 5. Unstable profiles of gas (---) and particle (—) velocities and solids concentration (---) along the jet central axis ($u_0 = 150 \text{ m s}^{-1}$, $x$ is the vertical distance from the nozzle exit).

The jet height in the case shown in Figure 5 is 0.071 m (Figure 4), but the program halts after 0.009 m because of the instability.

Figures 6a through to 6d show the dependence of the maximum particle velocity, occurring in the jet, on the four parameters in the ranges defined in Table 2.
From Figure 6a, there appears to exist a lower limit of $\xi^*$, below which the solution becomes unstable. This is shown by negative values of the solids concentration in these cases. Moreover, the limiting value of $\xi^*$ varies with $u_0$ and appears to be roughly proportional with $u_0$. From Figures 6b and 6c, there appears to be no effect of the starting values for solids concentration and particle velocity at the bottom of the jet central axis, right at the nozzle exit, except when $u_{p,i}$ assumes values of the same order of magnitude as $u_0$ (Figure 6c). From Figure 6d, finally, there appears to be an upper limit to the dimensionless lateral gas entrainment rate, $w_g/(\rho_g u_0)$, above which the particle velocity starts to run away. This limiting value varies with $u_0$. 

Figure 6a-d. Dependence of the particle velocity on model parameters.

$\bullet$: $u_0 = 50 \text{ m s}^{-1}$; $\square$: $u_0 = 150 \text{ m s}^{-1}$; $\diamond$: $u_0 = 250 \text{ m s}^{-1}$. 

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Values reported by Massimilla (1985) for various particle-fluid systems are of the same order of magnitude as those presented here and e.g. for air ($\rho_g = 1$ kg m$^{-3}$) and a jet velocity of 100 m s$^{-1}$ results in an entrainment velocity ($w_{gl}/\rho_g$) of about 0.1 m s$^{-1}$, which is of the same order of magnitude as the minimum fluidisation velocity.

For the model, the effect of parameter variations on the single particle velocities are of importance only when the effect on the trend of $u_p$ versus $u_o$ is considered. This is reflected in the value of the exponent $l$ (Equation 4), which is eventually affected by the program instability or the use of physically unrealistic values. Figures 7a and 7b show the dependence of the exponent $l$ on $\xi^*$ and $w_{gl}/(\rho_g u_o)$. The other two parameters, $u_{p,i}$ and $\gamma_{s,i}$ have been omitted here as they were found not to affect the trends of $u_p$, unless very large values of $u_{p,i}$ were used (Figures 6b and 6c).

![Graph](image1)

**Figure 7a-b.** Dependence of velocity exponent $l$ on parameter variations.

Figures 7a and 7b show that variations of 50% can be achieved by the choice of $\xi^*$ and of 100% by the choice of $w_{gl}/(\rho_g u_o)$. In the particular case of this model material, values of $\xi^* \geq 0.9$ and $w_{gl}/(\rho_g u_o) \leq 0.002$ should be used to provide a stable solution for the present range of $u_o$. The upper limit to $\xi^*$ is consistent with experimental observations that the jet boundary layer usually extends to no more than a few particle diameters (see also Appendices A and G). No evidence has been found in the literature with regard to the upper limit of $w_{gl}/(\rho_g u_o)$.
2.3 Forsythe and Hertwig Submerged Jet Fluidised Bed Attrition Test

Forsythe and Hertwig (1949) were among the first to recognise the importance of attrition in fluid catalytic cracking units. They devised a simple fluidised bed test to assess the resistance of the FCC particles to the mechanical stresses arising in fluidised beds. The test is based on a single jet of 0.397 mm, operated at an orifice gas velocity of just over 300 m s⁻¹. The importance of this test for the process industry can hardly be overestimated. It is commonly used not only in the oil industry for FCC, but also in the detergent industry for sodium perborates and by boric oxide suppliers (Hayat i, 1996). In fact, the ISO 5937 standard test of sodium perborate dustiness, formerly BS 5688, is based on this test. Other investigators have used it to assess the strength of iron molybdate catalysts (Cairati et al., 1980). Gwyn (1969) used an experimental technique based on the Forsythe and Hertwig test to propose his well-known model describing the time dependence of the attrition rate of catalyst particles.

2.4 Existence of the Jet Core

An important underlying assumption of the jet attrition model is the existence of an empty jet core, into which the particles become entrained. Once entrained, the particles accelerate and finally collide with the dense phase on top of the jet.

In the present study, observations have been made with high speed cameras which have enabled verification of this model assumption in a range of test conditions. In two-dimensional fluidised beds, the jet is highly unstable and sways from one side to another. Furthermore, the detachment of bubbles from the top causes the jet top to fluctuate strongly. These effects become strongly enhanced when observed at microscopic scale. Figure 8 shows the top of a jet of ambient air, recorded in the set-
orifice slot width of 5.0 mm. The particles in this case are 425-600 μm Salt. The image is recorded at 13,500 frames per second with a Kodak HS 4540 Motion Analyzer. The dense phase on top of the jet is shown brighter than the dilute jet core, which contains very few particles. The real size of the image is about 5.0 mm square. Particles at this point in the jet collide with the dense phase above the jet. Some particles collide with the sides of the jet, and may become re-entrained upon rebound, but this was observed very infrequently. After collision with particles in the boundary layer, the jet particles have lost most of their momentum and subsequent collisions with neighbouring particles occur at moderate velocities compared to the first impact. Collisions in the boundary layer take place at velocities which are closely comparable to those in the dense phase but at a rate which is comparable to the collision rate in the jet core, which is far below the rate in the dense phase where continuous particle contacting occurs. Similar observations have been reported by Seville et al. (1992). The first impact is sometimes observed to result in chipping of particles, in good agreement with the observations taken from single particle impact tests.

Figure 8. Dense phase on top of the dilute jet.

Clearly for the case presented in Figure 8 the dilute jet core exists. It has been observed, however, that the dilute jet core does not exist under all conditions. Figures 17a and 17b of Appendix G clearly distinguish between two cases where the dilute jet core ceases to exist in the region just above the distributor. The absence of the dilute jet core in this region is observed specifically when the jet operates at a low
orifice-to-particle size ratio. A quantitative measure of the existence of the dilute core is the extent to which particles that become entrained into the jet are able to accelerate. This is in fact the original indicator for the absence of the dilute jet core. For small orifice-to-particle size ratios, the particle velocities high up in the jet are found to be only marginally different from the particle velocities at the point of entrainment, as mentioned in section 2.2.
3. APPLICATION OF THE JET ATTRITION MODEL TO THE F&H TEST

An analysis of the behaviour of FCC particles in the Forsythe and Hertwig test and the modelling thereof with the present jet attrition model is presented in Appendices B, H and K. Appendix B describes a parametric study, in a two-dimensional fluidised bed, which verifies and validates the predictions of the hydrodynamic jet model of Massimilla and co-workers (1985) under conditions which are to some extent similar to those of the Forsythe and Hertwig test, assessing also some important input parameters for the jet model, such as the solids concentration profile and the divergent jet angle. It appears that the hydrodynamic model correctly predicts the particle velocities in the jet.

In the following comparison between the results from an experimental study of the attrition in the Forsythe and Hertwig test and the predictions of the jet attrition model, use is made of the single particle impact attrition propensity of fresh and used FCC, characterised in an extensive study of single particle impact breakage, reported in Appendix C.

In Appendix H, the effect of varying orifice-to-particle size ratios has been investigated for both fresh and used FCC, for one particle size and various orifice sizes. For used FCC, the attrition model is very well capable of predicting the attrition exponent for the dependence on the orifice size, $d_o$, expressed as $h$ in Eq. 7, for orifice-to-particle size ratios ranging from 1.5 to 16. For fresh FCC, on the other hand, a very different value of $h$ has been found in the experiments from the one predicted by the model (see Table 3 of Appendix H).

In Appendix K, the dependence of the attrition rate on the orifice gas velocity for a given orifice size, i.e. the Forsythe and Hertwig standard orifice size of 0.4 mm, has been investigated. Two materials, fresh and used FCC, have been used in different particle size ranges from 75 μm to 125 μm. It was found that the predictions for $n$, as defined in Eq. 6 using the jet attrition model compared very well to the experimental
values for used FCC, but for fresh FCC a large difference was found (see Table 2 of Appendix K).

It appears therefore that the jet attrition model correctly predicts the behaviour of used FCC, but not of fresh FCC in the range of orifice gas velocities (50 m s⁻¹ to 300 m s⁻¹), particle sizes (75 µm to 125 µm) and orifice sizes (0.175 mm to 1.5 mm) tested. In the following, several features of the hydrodynamics of the jet and their interactions with the structural properties of the particles will be discussed which cause this discrepancy. These issues also determine the applicability of the Forsythe and Hertwig test to other geometries and process conditions. A few examples are the process time during which particles are subjected to the test, the hydrodynamic regime imposed by the orifice velocity and orifice size, and the effect of particle structure.

3.1 Interaction between Particle Structure and Hydrodynamic Regime

The difference in the responses of fresh and used FCC to apparently similar hydrodynamic conditions in tests of the Forsythe and Hertwig (1949) type, as reported in Appendices H and K, has been very revealing with respect to the breakage mechanism of these samples in the hydrodynamic regime as prescribed by Forsythe and Hertwig (1949).

In the single particle impact test, both fresh and used FCC samples, regardless of size, failed following a necking mechanism, i.e. the agglomerated micro-spheroids disintegrated upon impact. This is similar to the observations of Forsythe and Hertwig (1949) for microspheroidal catalysts in their fluidised jet test. They note that during the attrition test, the agglomerates broke down and the resulting finer fractions were composed almost completely of individual spheres. Gwyn (1969) notes that the average diameter of the attrition fines is a function of the agglomerate structure of the catalyst. Contractor et al. (1989) note that any burrs break off in the Forsythe and Hertwig test. They developed a catalyst with an abrasion resistant shell, which
showed a very different performance in the Forsythe and Hertwig test from its performance in a commercial unit, and note that, for meaningful results, an attrition test should cause attrition by the same mechanisms that dominate the attrition process in the industrial reactor. Bass and Ritter (1977) show that catalyst used in a Roller test, based on a combination of a jet impingement and elutriation process (Allen, 1990), has been fractured, rather than worn, and note that fracture of the microspheres may be caused by the more severe environment encountered by the particles in the laboratory test when compared to the use in fluid catalytic cracking units. Ray et al. (1987) note that the dominant breakage mechanism in a fluidised bed is abrasion, and that cases of splitting are rare. At the same time, however, they note that the materials undergoing attrition, produce fines with a “natural grain size”, which is assumed to be a characteristic of the structure of the material, e.g. the primary particle size or the size of the microspheroids. This points at two phenomena, namely that the dependence of the breakage mechanism on the particle structure is evident in the resulting debris size distribution and that within the same tests, whether Roller or Forsythe and Hertwig, and at the same conditions, different breakage mechanisms may prevail depending on the particle structure.

3.2 Orifice-to-Particle Size Ratio and Breakage Mechanism

The orifice-to-particle size ratio has a strong effect on the mechanism of entrainment of solids into the jet core. Figure 9 shows schematically the fluidised bed above and around a jet nozzle of two different dimensions, as it was observed on high magnification video recordings using high speed cameras, as reported in Appendix G.
The jet depicted in Figure 9a has a large orifice size, compared to the particle size. Particles flowing in from the side suffer no resistance at all from the surrounding dense phase, and are entrained into the empty jet core and accelerate freely until they collide with particles in the dense phase covering and surrounding the jet.

The jet depicted in Figure 9b has a small orifice-to-particle size ratio. In this case, the surrounding dense phase completely engulfs the jet and a jet core is virtually non-existent. Particles flowing into the jet region and becoming entrained from the sides have to struggle and find their way through the dense jet region, where the solids concentration is only just below the dense phase value. However, the gas velocity is very high here, and the particles undergo a violent shearing action as they flow in from the sides and hit each other and slide and tumble against each other. The apex of the jet core is not, as is often assumed in the modelling of the jet penetration (Merry, 1975), situated inside the nozzle, some distance below the nozzle exit, but above the nozzle exit. The jet does open up, usually at a distance a few times the nozzle diameter away from the nozzle exit. The solids concentration in the more dilute region that is created further up is always well above the solids concentration in the dilute jet core of a large orifice-to-particle size ratio jet. This may be related to the fact that the solids blocking the nozzle exit enforce rapid dissipation of the gas momentum. The particle velocities are not very high, as the solids concentration is high throughout the jet penetration length and the efficiency of the momentum transfer is consequently low.
The effect that the above difference in entrainment mechanism has on the breakage mechanism is evident. In the case of a large orifice-to-particle size ratio, the particles are accelerated by the drag of an otherwise empty gas jet, and undergo a high speed collision, as determined only by the rate of momentum transfer from the gas issued from the jet nozzle and the weight of the particle. The particles, having reached their ultimate velocity, thus suffer damage in a way analogous to a single impact on a rigid target. In the case of the small orifice-to-particle size ratio, particles are subjected to intense and rapid shearing in a dense flow, and the process is more similar to autogenous grinding. Numerous collisions and sliding cause particle breakage in this case.

The breakage mechanism in the case of a small orifice-to-particle size ratio was found to be very different from that occurring in a jet with a large orifice-to-particle size ratio. The response depends very much on the susceptibility of the material to surface abrasion. This is illustrated in Figures 4 and 5 of Appendix H. Four cases have to be distinguished here, for large and small orifice-to-particle size ratios with either fresh or used FCC particles. In the case of fresh FCC, at an orifice-to-particle size ratio of about 2, the debris consists of very small fines, as shown in Figure 4a of Appendix H. In the case of a large orifice-to-particle size ratio of about 16, the debris consists of large fragments as well as a large quantity of fines, as illustrated in Figure 4b of Appendix H. For used FCC, with a small orifice-to-particle size ratio of about 2, the debris, shown in Figure 5a of Appendix H, consists of very few fragments and a small quantity of fines. For a large orifice-to-particle size ratio, the debris of used FCC, shown in Figure 5b of Appendix H, consists only of broken fragments, i.e. necked burrs.

Let us concentrate in the first instance on the first and the fourth case, of fresh FCC with a small orifice-to-particle size ratio, and of used FCC with a large orifice-to-particle size ratio, as they most clearly demonstrate the effect of the orifice-to-particle size ratio on the breakage mechanism. In a situation as sketched in Figure 9b, in the presence of a rapid shearing flow, the fresh FCC particles will suffer extensive
abrasive damage, resulting in the generation of a large amount of fines, as illustrated in Figure 10a. Fresh FCC is very susceptible to surface abrasion, as it has a very high surface roughness. In the case of a large orifice-to-particle size ratio, as sketched in Figure 9a, the used FCC will acquire a high velocity and impact on the dense phase covering and surrounding the jet, which may result in the necking fracture of weak microspheroids, as illustrated in Figure 10d. Secondary collisions at far lower impact velocity may not cause significant surface abrasion as the resistance to surface abrasion of used FCC is very high. Used FCC has survived many events of contact damage in the FCC unit operation at high temperatures and has reached the equilibrium state with respect to attrition: the edges are rounded off and the surface roughness has diminished because of the numerous times it has been recycled and regenerated.

The remaining two cases are affected by mixed processes. In the case of a large orifice-to-particle size ratio, fresh FCC will fracture in a way similar to a single impact on a rigid target, as illustrated in Figure 10c. However, secondary collisions will result in the generation of a significant amount of fines, which may adhere to the particle surface and act as a buffer to collisions and limit the damage. The adhesion of fines to large fragments is illustrated in Figure 4b of Appendix H. In the case of a small orifice-to-particle size ratio, the rapid shearing flow will cause the generation of only a limited amount of fines in the case of used FCC, as its resistance to surface abrasion is very high. Collisions higher up in the jet, at a higher velocity and in a more dilute environment, will cause some fracture of burrs from the surface, as illustrated in Figure 10b. The total amount of breakage in the latter case is of course several orders of magnitude below the extent of breakage observed with fresh FCC for a similar hydrodynamic case. This is illustrated in Figure 2 of Appendix K.


3.3 Applicability of the Jet Attrition Model

The jet attrition model correctly predicts the attrition dependence of used FCC on the operating parameters with both small and large orifice-to-particle size ratios, as this material is not susceptible to surface abrasion even with small orifice-to-particle size ratios. However, with fresh FCC the predictions are quite different from the measured values. A good comparison between the predictions from the jet hydrodynamic model and the particle velocities in the jet has been found for a wide range of orifice-to-particle size ratios covering both hydrodynamic regimes, provided that the correct boundary conditions are supplied. These have been measured for a range of orifice-to-particle size ratios for which the agreement between the model predictions and the data is good (see Appendices A and G). It must therefore be
assumed that the use of the single particle impact model is responsible for the failure of the model in the case of fresh FCC. The above observations with respect to the breakage mechanism provide a clear indication that the susceptibility of this material to surface abrasion makes it sensitive to the rapid shear flow occurring in the lower region of the jet. Surface abrasion rather than collisional damage is the dominant breakage mechanism here, and therefore an additional contribution from this mechanism should be added to the model in order to obtain valid predictions in the case of a small orifice-to-particle size ratio. Further work is required to characterise the threshold orifice-to-particle size ratio below which the surface abrasion becomes dominant, but there are several indications, which will be discussed below, that it is about 10.
4. BREAKAGE MECHANISM AND DEBRIS FEATURES

The investigations, described in Appendices H and K have been specifically targeted at identifying the dependence of the breakage mechanism on the hydrodynamic regime prevailing in the Forsythe and Hertwig test. The response of fresh and used FCC to a submerged jet test following the Forsythe and Hertwig recipe is illustrated in Figure 4 of Appendix K. There is a clear difference in the size of the debris for fresh and used particles. The debris of fresh FCC consists of micron-sized chips, whereas the debris of used FCC, produced under the same conditions, consists of microspheroids, previously belonging to larger agglomerates, as is clear from the presence of the rough necks, which indicate the place where the microspheroid was previously attached to the agglomerate. Apart from the difference in average size, it is worth noting that both samples are very uniform in size.

Bass and Ritter (1977) note that used FCC after recycling and regeneration has a sintered surface layer of approximately 0.2 \( \mu \text{m} \) thickness, which makes it far more resistant to surface abrasion. This is in a way similar to the progress of sulphation of limestone, discussed in Appendix I, which shows that the formation of a highly resistant sulphate shell significantly reduces the attrition rate. Contractor et al. (1989) also show a significant reduction of the attrition rate by the formation of a hard silica shell on FCC particles. Above a certain threshold velocity, however, the shell was shown to suffer catastrophic failure and the slope of the data trend of attrition vs orifice gas velocity changes at this point dramatically. A similar effect has also been observed in a computer simulation of agglomerate impact by the Distinct Element Method of the effects of the presence of a thin layer of small particles on an agglomerate with a core of large particles, as shown in Figure 1b of Appendix E. Figure 7 of Appendix E shows that the extent of breakage of the bimodal agglomerate is much lower than that of a uniform agglomerate, until a threshold velocity at approximately 4 m s\(^{-1}\) is exceeded, at which point the order switches and the bimodal agglomerate fails catastrophically, with the extent of breakage exceeding that of a uniform agglomerate. It is unclear at this stage, whether
this is due to a different breakage mechanism, resulting in a difference in the size distribution of the product, and further work is on-going to characterise this aspect.

The size distribution of a breakage product is of great importance in the determination of the breakage mechanism and the material response to hydrodynamic stresses. Visual examination of the FCC particles, shown in Figure 1 of Appendix C and in Figures 11 to 13 below, allows the identification of a number of different length scales in the particle structures, which are dominant, depending on the particle size and the process history of the material. When looking at the particles, shown in Figure 11, a ginger-root shape is observed, with multiple microspheroids of a few tens of microns in size, attached to a larger spheroid. Furthermore, the fresh FCC (Figure 11a) has a far more complex structure than the used FCC (Figure 11b). The latter has far less burrs on the surface and has a much smoother appearance. At intermediate magnification, shown in Figure 12, we observe less of the ginger-root structure, and the spheroid shape and size is more predominant. At higher magnification still, shown in Figure 13, a large difference in the length scales is observed between fresh (Figure 13a) and used (Figure 13b) FCC. The length scale of surface roughness of fresh FCC is much larger than that of used FCC. Bass and Ritter (1977) show that the internal pore structure is dominated by the size of the constituent kaolin platelets, aluminosilicate gel, and zeolite, which are all of the order of a micron in size. Also, debris from the Forsythe and Hertwig test of this powder, shown in Figure 4a of Appendix K, is predominantly of this size. On the other hand, debris from the Forsythe and Hertwig test of used FCC catalyst, shown in Figure 4b of Appendix K, is much larger. The used FCC has the same origin as the fresh FCC, but has been recycled in an FCC unit.
Figure 11. 75-90 μm fresh (a) and used (b) FCC at low magnification.

Figure 12. 75-90 μm fresh (a) and used (b) FCC at medium magnification.

Figure 13. 75-90 μm fresh (a) and used (b) FCC at high magnification.

Figure 14 shows an FCC particle and three different damage patterns for this type of FCC particle, of which a real sample is shown in Figure 11. The particle labelled I is the original particle with its ginger-root structure of secondary agglomerates. The
particle labelled II has suffered minor damage on the surface. This could occur at low velocities in a single particle impact or in a dense shear flow. At higher impact velocities, larger burrs may be broken off, as shown in case III, and at still higher velocities the particle may fragment into several pieces (IV). In dense phase flow, even at high compressive and shear loads, it is unlikely that particle fragmentation would occur. The debris features thus reflect the process that caused the damage.

![Image of particle damage patterns](image)

(a) Original particle  
(b) Surface chipping  
(c) Microspheroid fracture: necking  
(d) Fragmentation

**Figure 14.** FCC damage patterns.

The Distinct Element Analysis of an impact of a lactose agglomerate, described in Appendices D and E, has given some insight into the internal shear deformation mechanisms that cause failure of this type of binderless agglomerate. The primary particle size distribution used for these simulations is very uniform, almost monosized. Figure 11 of Appendix D shows the resulting product size distribution. It appears that the slope of the size distribution is almost constant for all impact velocities. The product size distribution is not nearly as uniform as the primary particle size distribution. The self-similarity of the product size distribution may, however, be significant. In a study of the impact behaviour of brittle spheres, Arbiter et al. (1969) show that the resulting size distribution can be represented by a set of
three straight lines in a log-log plot, one for each of three categories: coarse fragments (residual, or mother particle), fine fragments (complement) and dust. The slope of the line representing the complement was found to be constant with varying impact velocity. Papadopoulos *et al.* (1998) and Subero *et al.* (1998) report the same feature for the impact products of common salt and glass agglomerates, respectively. Merrick and Highley (1974) note that in fluidised bed combustion of coal particles, the size distribution of the elutriated fines was similar in all test conditions. Arena *et al.* (1983) measured the size distribution of carbon fines in a fluidised bed combustor. They found that there is only a slight change in the fines size distribution with a change in fluidising velocity, and suggest that this is related to the changing cut-off size for elutriation. They also note that the size of the inert bed material may affect the size of carbon fines generated. Based on these two studies and their own observations, Ray *et al.* (1987) suggest that the effects of particle size and gas velocity on the fine size distribution are not significant and that a basic size distribution exists. The size distribution of elutriated fines is strongly related to the hydrodynamic conditions at the outlet of the fluidised bed, which makes it hard to interpret this as a function of the breakage process conditions. In the other cases, e.g. single particle impact, establishing a relationship between the product size distribution and the breakage process is more straightforward and certainly deserves further investigation. The Distinct Element Analysis may provide a convenient way of investigating the relationship between the debris features and specific breakage mechanism, and to deconvolute combinations of breakage mechanisms, such as surface wear and fragmentation (Ghadiri and Ning, 1997).
5. THE USE OF THE IMPACT MODEL

5.1 Evaluation of the Impact Attrition Exponent

The role of the single particle impact model in the jet attrition model is of vital importance, as it is here that the breakage is accounted for. The breakage model produces the exponent $m$, which appears in the formulation of the dependence of the attrition rate on both the orifice gas velocity and the orifice size, as shown earlier. Papadopoulos (1998) has collected the values of $m$ for various materials, and has shown that a large number of materials show an exponent of $m = 2$. Examples of these materials are PMMA, NaCl, KCl, MgO, $\alpha$-lactose monohydrate, polystyrene, and both fresh and used FCC, shown in Appendix C, as well as the fresh Ignaberga and Massicci limestone samples, shown in Appendix I. These materials have very different properties and structures, and they may not all fail in a semi-brittle mode. Recent experiments with prills have shown that $m$ may strongly reflect the breakage mechanism as a function of the processing conditions, e.g. temperature. The value of $m$ changes dramatically, depending on whether the tests are carried out above or below the glass-transition temperature. However, there are cases, such as the lactose agglomerates, as reported in Appendix D, where the breakage mechanism is very different from the semi-brittle failure mode, but may still result in a value of $m$ equal to 2.

In previous sections, it has been argued that the single particle impact model can be used only if the breakage mechanism in the jet is the same as that occurring in the single particle impact test. If we restrict the breakage mechanisms to semi-brittle failure, the value should be 2, and even when the failure does not occur in semi-brittle mode the exponent may be 2. The discrepancy in behaviour of fresh and used FCC in the jet, and by contrast similar behaviour in single particle impact tests show that two very similar materials with the same exponent $m$ may behave totally differently in a fluidised bed jet. The use of the exponent alone may be too simplistic,
and is not sufficient to characterise the breakage occurring in a fluidised bed jet and ensure the applicability of the impact attrition model.

5.2 Jet Attrition Rate

The attrition rates of fresh and used FCC, reported in Appendix K for the Forsythe and Hertwig (1949) test, differ widely. The jet attrition model should be further developed to take account of differences in the material properties. The attrition exponent cannot express these differences alone, and the value of the proportionality constant needs to be determined to enhance the predictive capability of the model. In fact, the only proportionality constant which has so far received any attention has been that of Equation 8. Papadopoulos (1998) has found that it has a value between 1 and 10 for the materials tested in his work. Following the comprehensive analysis of Papadopoulos (1998), it can be assumed that the proportionality constant of the impact attrition rate is not a function of material properties. Further work is required to characterise and analyse the rate constants and their dependence on the hydrodynamic conditions prevailing in the jet, and on the impact attrition rate.

5.3 Alternative Impact Tests for Different Breakage Mechanisms

In previous sections, it has been shown that the single particle impact model cannot provide accurate predictions for rapid shearing flow when particles are susceptible to surface abrasion. The breakage mechanism following numerous low energy collisions and shear cannot be accurately represented by a single high velocity impact, when particles exhibit work hardening, fatigue, or have high surface roughness. This could be more suitably represented by repeated impact tests at low impact velocity, or by impacts at an inclined target. Some repeated impacts have been performed with fresh FCC, as reported in Appendix C. Very little work has been done so far on the assessment of the effects of multiple impacts and inclined targets on the attrition propensity of particulate solids (Papadopoulos, 1998; Salman et al., 1995; Uuemois and Kleis, 1975). An extensive investigation of the response
of the attrition propensity of particles and their properties on the number of impacts and the impact angle would be required to enhance our understanding and extend the application of the jet attrition model to the case of rapid shear flow.
6. **Jet Solids Concentration Profile and Angle**

A number of assumptions in the jet hydrodynamic model concern the profile of the solids concentration in the jet. In the present work, a novel method has been employed to measure the solids concentration profile in two-dimensional jets in fluidised beds. Figure 9 in Appendix G shows a typical example of a solids concentration profile obtained in this way. The solids concentration is shown here as a widening valley through a highlands landscape. The downward slopes are the boundary layers, where the solids concentration gradients are located and the dense phase borders on the jet. The vertical position is relative to the porous distributor plate, and the centre of the jet nozzle is located at a horizontal position of approximately 4.5 mm. Close observation shows that the gradients at the nozzle exit are sharper than at the top, indicating that the boundary layer broadens as the jet penetrates further into the bed. The assumption of a constant value for $\xi^*$, the boundary layer width (Eq. 9), with height therefore seems fair. The exact portion of the jet which is to be located in the boundary layer and the jet angle at which it extends are not easily assessed from this kind of presentation. Measurements of the jet angle have been carried out using the method also described in Appendix G for a number of conditions. The experimental measurements of the jet angle will be compared below to a dimensional analysis of the parameters controlling the jet angle, carried out recently by Vaccaro (1997).

6.1 **Dimensional Analysis of Jet Angle Parameters**

In a recent publication, Vaccaro (1997) presents a dimensional analysis of the jet angle, $\theta$, taking into account the following eight hydrodynamic parameters:

$$\theta = f(d_m, u_p, \rho_g, \rho_p, d_p, g, L_m)$$

(10)

where $L_m$ is the jet penetration length. According to Vaccaro (1997), a combination of these parameters leads to a proportionality expression for $\theta$ in the following form:
\[ \theta \propto \left( \frac{\rho_p - \rho_g}{\rho_g} \right) g \frac{d_p}{d_o} \left( \frac{L_m}{d_o} \right)^2 \]  

(11)

The first fraction on the right-hand side is the square of the inverse of a two-phase Froude number times a characteristic dimension. Vaccaro (1997) proposes two ways of expressing the Froude number, either on the basis of the particle size, or on the basis of the orifice size:

\[ Fr = \left( \frac{u_o^2}{g \frac{d_o}{d_p} \rho_p - \rho_g} \right)^{1/2} \]  

(12)

or

\[ Fr^* = \left( \frac{u_o^2}{g \frac{d_o}{d_p} \rho_p - \rho_g} \right)^{1/2} \]  

(13)

When the formulation of Equation 12 is used, the square of the inverse of this Froude number is multiplied by \( d_p \), and when Equation 13 is used, by \( d_o \), so that eventually the same product is found. The discrepancy between the two therefore appears to be of a rather hypothetical nature, as it is no longer found in the final expression of \( \theta \) (Eq. 11). Vaccaro (1997) nevertheless plots the jet half angle versus the inverse Froude numbers \( Fr \) and \( Fr^* \). He finds that for orifice-to-particle size ratios below 7.5 the jet half angle does not vary appreciably with both Froude numbers, whereas for orifice-to-particle size ratios above 7.5 there is a clear dependence of the jet half angle on the Froude number.

6.2 Comparison with Present Measurements - Froude Number

The measurements of jet angles in the present work as a function of the orifice size and orifice gas velocity, shown in Figure 6 of Appendix A, can be easily plotted against the Froude numbers as indicated above, to give some insight into the choice of \( Fr \) or \( Fr^* \). Figure 15 shows the dependence of the jet half angle on the Froude numbers, as given by Eqs 12 and 13. This provides a limited basis of comparison as the dependence of \( \theta \) on the Froude number has not been fully tested. Nevertheless, in both cases a reasonable degree of uniformity is achieved. If we look more closely at
the region where values of $1/\text{Fr}$ and $1/\text{Fr}^*$ are small, we observe that the slope of the data is lower than in the region where $1/\text{Fr}$ and $1/\text{Fr}^*$ are higher. The normalisation of the data with respect to the different orifice sizes results in a closer fit in this region for $1/\text{Fr}^*$ than for $1/\text{Fr}$, as the data sets of the orifices of 0.5 mm and 1.0 mm fully coincide in Figure 15b, whereas a significant difference is still observed in Figure 15a. It appears that there is a change of slope in the data at $1/Fr = 1$ and $1/Fr^* = 0.1$, clearer in the former case than in the latter. This is exactly the point where the orifice-to-particle size ratio exceeds the limit of 7.5. The change of slope is much harder to observe here than in other data in the literature, as correlated by Vaccaro (1997). Below these threshold values of the Froude numbers, still some dependence of the jet half angle on the Froude number is found, whereas Vaccaro (1997) found that the jet half angle showed no dependence on the Froude number at all.
6.3 Comparison with Present Measurements - Overall Analysis

Vaccaro (1997) does not present the final supporting evidence for his dimensional analysis, as he does not plot the jet angle against the function defined in the right-hand side of Equation 11. In order to calculate this function, an estimate of the jet penetration length, $L_m$, is required. This was obtained from a number of correlations.
for the jet penetration length. Most correlations include a two-phase Froude number, which means that their correlation with the jet half angle will not be straightforward and transparent. For the purpose of the present comparison, two correlations have been selected which have been found to give reliable predictions in various studies, as reviewed recently by Kimura et al. (1995). These are the correlations of Yang and Keaims (1979):

\[ L_m = 15.0 \, d_o \left( \frac{\rho_g}{\rho_p - \rho_g} \frac{u_o^2}{g \, d_o} \right)^{0.187} \]  

(14)

and Yates et al. (1986):

\[ L_m = 21.2 \, d_o \left( \frac{u_o^2}{g \, d_p} \right)^{0.37} \left( \frac{\rho_g \, d_p \, u_o}{\mu_g} \right)^{0.05} \left( \frac{\rho_g}{\rho_p} \right)^{0.68} \left( \frac{d_p}{d_o} \right)^{0.24} \]  

(15)

In both correlations the two-phase Froude number appears, incorporating either the orifice size (Yang and Keaims, 1979) or the particle size (Yates et al., 1986).

Figure 16 presents a plot of the jet half angle as a function of the r.h.s. expression in Equation 11 for various estimates of the jet penetration length, \( L_m \). In Figure 16, on the horizontal axis the functional form with \( Fr \) has been used:

\[ d_p \left( 1/ Fr \right)^2 \left( L_m/d_o \right)^2 \]  

(16)

On the basis of Equations 11 and 12 this is equivalent to:

\[ d_o \left( 1/ Fr^* \right)^2 \left( L_m/d_o \right)^2 \]  

(17)

where \( Fr^* \) has been included instead. The correlation for the jet penetration length of Yang and Keaims (1979), shown in Equation 14, is a function of the Froude number \( Fr \) based on the orifice size. Thus, comparing Figures 15a and 16b, only a change in the scales on the axis, but not in the data trend is observed. When using the correlation of Yates et al. (1986), on the other hand, the uniformity of the trend of the data is enhanced for the smallest orifice of 0.5 mm, but the comparison has deteriorated for the intermediate size of 1.0 mm, which appears to be anomalous. Hence there is clearly a strong interaction between the jet penetration length and the jet half angle, which cannot all be characterised by a single two-phase Froude number.
Figure 16. Jet half angle vs parameter from Vaccaro (1997).

Data from Appendix B.
6.4 Effect of Orifice-to-Particle Size Ratio

Vaccaro (1997) suggests that the sensitivity of the dependence of the jet half angle on the two-phase Froude number with respect to the threshold value of 7.5 may originate from a transition in entrainment mechanism or gas-solids momentum transfer inside the jet, and he refers to the present work for experimental support (Boerefijn and Ghadiri, 1996). Figure 17b in Appendix G shows that for a small orifice-to-particle size ratio, the jet boundaries close in on each other, so that the jet approaches a point source. Close to the nozzle exit, there is virtually no solids entrainment and there may be little gas exchange with the surrounding dense phase. This would in turn limit the sensitivity of the jet half angle to changes in the ratio $u_o^2/d_o$, which is the core variable of the two-phase Froude number when applied to a single gas-solids system.

The Froude number takes into account only one of two characteristic dimensions of the system, either the particle size, or the orifice size. It is therefore surprising that the Froude number can reflect the limit in $\theta$, which arises from the critical orifice-to-particle size ratio! The orifice-to-particle size ratio should influence the momentum transfer and solids entrainment, which cause a change in the dependence of not only the jet angle, but of all other jet parameters on the Froude number, such as the jet penetration length. For the case where $\theta$ approaches its limiting value at the threshold orifice-to-particle size ratio of 7.5, Vaccaro (1997) rightly remarks that perhaps this does not signify a limit in $\theta$, but a limit to the existence of the jet itself, where the analysis breaks down.

Grace and Lim (1987) re-analysed a large amount of literature data and confirmed the finding of Chandnani and Epstein (1984), that an orifice-to-particle size ratio of 25.4 is required for the formation of a stable jet, penetrating the bed. Above this ratio, the jet is unstable and consists of a series of bubbles which break away from the nozzle exit. This number is well above the present critical ratio of about 10, and in fact relates to a different phenomenon. Grace and Lim (1987) also plotted a Froude number, based on the orifice size, against the orifice-to-particle size ratio, and found
that they are totally uncorrelated. The analysis, presented in the previous section, showed that the overall parameter (Eq. 16) was not affected by including the jet length correlation of Yang and Keairns (1979). This correlation contains a Froude number based on the orifice size (Eq. 14). On the other hand, some effect of the jet length correlation of Yates et al. (1986) was observed (Fig. 16). The Froude number used in this correlation is based on the particle size (Eq. 15). These observations may aid in resolving the dilemma of Vaccaro (1997) on the choice of Froude number to model the jet angle, i.e. based on the particle size or the orifice size.
7. **Effect of Processing Time**

Forsythe and Hertwig (1949) report a strongly time-dependent response of fresh synthetic silica-alumina catalyst in their attrition test. Within the first 5 minutes the fines content increases from 3 to 19%, whereas during the last 5 minutes of the 60 minutes test period the increase is only from 51 to 53%. Forsythe and Hertwig (1949) attribute this effect to four factors:

- the initial rounding-off of the rough edges,
- the elimination of weaker particles,
- the increase in the content of fines, which act as cushions, and finally
- the decreased content of coarse particles.

When the fines content was screened from the product after a one hour test, and the same sample was again subjected to the test for another hour, the attrition rate was found to be approximately constant.

### 7.1 Equilibrium Attrition Rate

Cairati *et al.* (1980) have used the Forsythe and Hertwig test for the assessment of the mechanical behaviour of a newly developed iron-molybdate catalyst. They note that comparisons of catalysts on the basis of this test are relative and do not permit direct quantitative prediction of actual plant losses, unless the test procedure reproduces in a reasonable way the mechanism of particle breakdown in commercial units so that the results can be representative of commercial operation. They presume that the attrition in the jet is caused by the collisions of particles which have been rapidly accelerated by the air jet with slower moving particles, which is supposed to simulate the actual situation in the grid zone of a fluidised bed, as well as by collisions of the particles in the bulk of the fluidised bed. Their results show that during the last 20 minutes of the one-hour attrition test the fines content increases from 37% to 47% for a control catalyst and from 62% to 75% for their new catalyst. If the test is carried on for another 20 minutes, until 80 minutes total test time, the fines content further increases at a fixed rate for the control catalyst, but the increase in fines
content for the new catalyst is much lower, indicating that it has not reached steady state within the first hour. Contractor et al. (1989) show that the attrition rate of catalyst, expressed as the weight loss per hour, decreases gradually with time over a period of no less than 2600 hrs. It is therefore to be concluded that the time necessary to reach equilibrium, if this exists at all, very much depends on the material properties.

7.2 Time Effects in F&H Test

In the present work, a series of tests have been carried out to assess the time dependence of the attrition rate in the Forsythe and Hertwig test for the fresh FCC samples. As the scope of the present work included assessing the suitability of the Forsythe and Hertwig test, it was important to assess also the effect of the processing time used in that test, in order to see if the samples reached equilibrium rates well within the test period. Samples of fresh FCC, 75-90 μm, were subjected to fluidisation with an air jet in a Forsythe and Hertwig set-up for different lengths of time, and afterwards the attrition rate was assessed by gravimetric analysis of the products. The debris separation was achieved by using a sieve size of 53 μm, i.e. two BS410 standard sizes below the lower size limit of the original sample size range.

The results are shown in Figure 17 for tests of up to 5 hrs. The attrition rate, expressed as the fractional mass loss, increases with processing time. During the first hour, the rate drops quickly, after which it levels off, as is evident from Figure 18, which shows the rate of attrition as a function of the processing time. This indicates that during the first hour, the process of rounding off and elimination of weaker particles is dominant. As the equilibrium time depends on the material properties, it is clear that the one hour testing period in the Forsythe and Hertwig test has an important effect on the test results.
Figure 17. Time dependence of attrition rate of 75-90 μm fresh FCC in the Forsythe and Hertwig test.

Figure 18. Time rate of attrition of 75-90 μm fresh FCC in the Forsythe and Hertwig test.
7.3 *Time Effects and Chemical Reactions*

In the study of limestone attrition during fluidised bed desulphurisation, reported in Appendix I, the time-dependent attrition rate of two different types of limestone has been characterised. There appears to be a strong dependence of the attrition rate on the particle porosity and its resistance to thermal shock. The materials were processed via two different routes, either via calcination and subsequent sulphonation, or simultaneous calcination and sulphonation. The comparison of the resulting products showed that the impact attrition propensity of the materials had changed dramatically, depending on the process route. It was also observed that the formation of an abrasion resistant shell of sulphated lime around the calcined core protected the particle from extensive surface abrasion. The time required to reach equilibrium rates was just below two hours, as shown in Figures 3 and 4 of Appendix I, which is also the time required to complete conversion. There exists, therefore, a clear relationship between chemical reaction and equilibrium attrition rates.

7.4 *Equilibrium in Standard Tests*

The ISO standard test for dustiness of sodium perborates, ISO 5937, has been derived on the basis of the Forsythe and Hertwig test. The ISO test prescribes fluidisation using a single air jet at about 900 m s\(^{-1}\) for one hour. The total amount of fines below a certain size limit is taken as a measure of attrition. Two misconceptions can be identified in this approach, which were also evident in other investigations employing the Forsythe and Hertwig test. Firstly, that the samples will exhibit equilibrium attrition rates during the entire test period of one hour, or that non-equilibrium effects may be neglected. Forsythe and Hertwig (1949) clearly indicate that only the final stage of the test can be accepted as a significant measure of the equilibrium rate. Taking the total amount of fines produced during the one-hour test will incorporate the initial rounding-off and weaker particle elimination, which will exceed by far the final amount of fines collected towards the end of the test and will bias the test results. Experimental evidence of Cairati *et al.* (1980) and Contractor *et al.* (1989)
has shown that not even the final values of the attrition rate after one hour are acceptable as measures of the equilibrium rate. Secondly, that fluidisation with these small nozzles will cause breakage following the same breakage mechanism taking place in the actual process it is trying to simulate. The combined evidence from the present test results, shown in Figures 17 and 18 and in Figure 13 of Appendix A, and from the literature, indicates that great caution should be exercised when comparing fresh catalysts on the basis of their performance in these tests for one hour. With used catalyst it is an entirely different matter, as these particles are no longer susceptible to surface abrasion, as they have reached the equilibrium state and also because the breakage mechanism is more similar to that occurring in a real process. Therefore the attrition should be investigated as a function of time to get a full understanding of the various prevailing mechanisms.
8. ELUTRIATION RATE AS MEASUREMENT OF ATTRITION RATE

8.1 Fines in Bed Inventory

The ISO 5937 standard test for sodium perborates, like the Forsythe and Hertwig test, prescribes that the contents of fines in the exhaust filter be a measure of the attrition rate. For relatively coarse solids, like the char particles of several millimetres original size, used by Massimilla and co-workers in their studies of comminution of carbons in fluidised bed combustion (Chirone et al., 1991), or the limestone particles used recently in studies of attrition in desulphurisation processes, reported in Appendix I, this may be a valid approach, as the fines produced by abrasion can easily percolate through the coarse fluidised bed contents and elutriate. The gas velocity required to fluidise the coarse bed inventory is probably several times higher than the terminal velocity of the fines. Ayazi Shamlou et al. (1990) found that with 2 mm glass agglomerates, after 16 hr of fluidisation at up to 1.3 times the minimum fluidisation velocity no less than 28% of the fines had remained in the bed and had not become elutriated. In the present studies of the Forsythe and Hertwig test of FCC particles, a considerable amount of fines, between 30% and 60%, was found in the bed inventory, rather than in the exhaust filter. The exhaust filter consisted of a 0.2 μm absolute retention disc filter. The superficial gas velocity was high enough to entrain particles of a size up to about 30 μm, not far below the debris separation size limit. The debris of fresh FCC in the Forsythe and Hertwig test consisted mainly of small fines, of a few micrometer in size, which could easily elutriate from the bed at the prevailing superficial gas velocities. Nevertheless, a significant quantity always remained in the bed. Therefore it is essential to always analyse the bed inventory.
8.2 Incomplete Local Fluidisation and Fines Hold-up.

Studies of segregation of fine particles in packings of coarse particles have shown that there are conditions in which the fines are distributed evenly throughout the packing, to a level which is determined by the interstitial gas velocity. This is also called confined fluidisation (Donsi et al. 1988, 1990). In these systems, the minimum fluidisation velocity for the fines in the interstices is given by the product of the unconfined minimum fluidisation velocity and the voidage of the coarse packing. At moderate interstitial flow levels, where the coarse particles did not fluidise, not all fines migrated to the free bed surface. This indicates that in defluidised zones in a fluidised bed, only very limited fines percolation may occur. In bubbling fluidised beds, fines migrate mainly as a consequence of bubble motion (Hailu, 1992). At background fluidisation levels close to the incipient fluidisation condition, and even at higher fluidisation levels, it is well-known that the jet may draw gas from the dense phase into the jet core (Vaccaro, 1997). This may cause local defluidisation around the jet which in turn would inhibit the migration of fines, created by jet attrition, to the bed surface and fines may also migrate to dead zones, depending on the operating conditions. In the absence of background fluidisation, as in the Forsythe and Hertwig test, the migration of fines from the dead zones surrounding the central jet nozzle may be limited. Great caution should therefore be exercised when estimating the attrition rate by assuming that it is proportional or equal to the elutriation rate.
9. **Hydrodynamic Modelling of the Jetting Region**

The hydrodynamic model of Massimilla and co-workers (1985) has been found to give correct predictions for the particle velocities in the jet and its use in the jet attrition model has in many cases provided the correct input to predict the appropriate dependence of the overall attrition rate on the operating parameters.

A more comprehensive approach to solving the momentum and continuity balances for the gas and solids phase was employed by Gidaspow *et al.* (1983) for the entire bed, including the jet region. Although to a lesser extent than in the model of Massimilla and co-workers, as discussed in Chapter 2, this model also relies on assumptions on the geometry of the jet, as the fluidised bed is divided into two sections, a jet and a downcomer. However, the resulting predictions of solids concentration and velocity profiles could not be verified satisfactorily in experiments.

Given the nature of the present hydrodynamic jet model, which requires a large number of empirical input parameters, there are good reasons to look at different ways of modelling the jet region. Two different routes can be taken here, discrete element and granular kinetics modelling.

### 9.1 Kinetic Theories for Granular Flow

The continuum mechanics approach can be extended by including assumptions on the nature of the particle motion and the mechanism of energy dissipation in the dense phase, as in the case of granular kinetics simulations. The granular flow theory of Haff (1983), further developed and applied by Savage and co-workers (Lun *et al.*, 1984) and culminating in the computer simulations of Campbell (1990) have made great progress in the simulation of rapid shear flow in dense phase granular systems. In this theory, particle collisions at a certain root-mean-square (r.m.s.) velocity and rate per unit volume are used as dissipative mechanism. Different ways of modelling the particle collisions, either as elastic or inelastic with a certain coefficient of
restitution have been derived for different flow systems (Campbell, 1990; Lun et al., 1984). Haff (1983) identifies a number of important criteria which have to be satisfied before molecular dynamics can be extended to granular flow. One example of this is the continuum hypothesis: a cubic millimetre may hold $10^{19}$ molecules of water, but only a few grains of sand. Therefore velocity gradients in granular flow may not be easily linearised on the length scale of a few grains, as r.m.s. values have no significance here. Other length scales which affect the continuum hypothesis are the size of the container confining the system, which is usually only three orders of magnitude larger than the grain size, and the perturbation scale to which interparticle collisions will affect their neighbours. The latter can be considered small, as the inelastic collisions will dampen the perturbation of interparticle collisions and limit the perturbation to a few particle diameters only. These conditions are not easily satisfied in the jetting region.

In the case of a large orifice-to-particle size ratio, the jet may be considered a separate continuum from the dense phase, with sharp gradients of solids concentration and velocity over a distance of a few particle diameters. The size of the jet may extend from a few tens of particle diameters to several hundred particle diameters, so that the entrainment of a few particles will cause a large disturbance of the flow field.

Finally, the collision of entrained particles with the jet boundary may cause them to rebound over a considerable distance, causing perturbations of the flow field which may extend to the size of the jet, rather than the single particle. In the case of a small orifice-to-particle diameter, this perturbation may be limited to a few particle diameters, so that the condition of the continuum hypothesis may still be satisfied, and also the assumption that the separation distance between the particles is of the order of the particle diameter may be justified, as illustrated in Figure 9b.

The dilute pocket, existing above the dense zone at the nozzle exit in the case of a small orifice-to-particle size (Figure 9b), cannot be easily included in this type of model, as here the separation distance between the particles well exceeds the particle
size. For these discontinuities it may be appropriate to put a moving boundary around the dilute pocket, or the empty jet core in the case of a large orifice-to-particle size. The moving boundary can be fixed by iterative solution of the continuum balance for the dense phase and the dilute phase. The transport of solids across this boundary would be a considerable complication. As the shape of the jet would have to be defined a priori in order to obtain a starting value for the boundary conditions, this model would be subject to the same weakness as the hydrodynamic model of Massimilla and co-workers (1985). Moreover, Lun et al. (1984) mention that the current state of granular dynamics modelling, which assumes instantaneous binary collisions, does not yet include rate-dependent stress contributions resulting from enduring contacts between particles at high concentration, as would occur in the dense region above the nozzle exit in case of a small orifice-to-particle size ratio.

Campbell (1990) notes that the above granular methods wrongly rely on the assumption that the particle motion and collisions can be described by independent spatial distribution and probability density functions, which breaks down at high solids concentration. These methods do provide quantitative information on the collision rate and impact velocity of interparticle collisions, and an extension of these models to provide predictions on the rate of attrition in suitable systems is possible. The mechanism of collision is, however, binary and instantaneous, and true shear is not taken into account. This is due to the fact that interparticle collisions are a means of energy dissipation in these models, which have been devised to describe granular flow and no interest is taken in the effects of breakage. The material properties have no bearing on the interparticle collisions and usually a constant coefficient of restitution is assumed, which is not a function of impact velocity. Plastic and frictional dissipation processes are not considered in these macroscopic models. Campbell (1990) notes that a correction for the mechanical properties of the particles would probably be intractable in any theoretical model or computer simulation, but given time the computing power may develop sufficiently to allow this.
The Distinct Element Method (DEM) has recently been employed to model jets in fluidised beds (Tsuji et al., 1993; Hoomans et al., 1996). In this method, particles are individually tracked and interparticle forces are calculated using contact mechanics and then used to determine the motion of the individual particles by integration of the Newtonian equations of motion. Tsuji and co-workers have applied this method in the simulation of fluidised beds (Tsuji et al., 1993). The results of simulations of a jet in a two-dimensional fluidised bed appear very convincing, but the values used for the contact stiffness are unrealistically low, which allows for large calculation time steps, but does not represent realistic contact mechanics. More recently, Hoomans et al. (1996) presented similar DEM simulation results partly based on a hard-sphere granular dynamics approach. The hard-sphere approach assumes that particles exchange momentum only by means of instantaneous and inelastic collisions. Furthermore particles are assumed to be rigid spheres which cannot be deformed. On the other hand, soft-sphere DEM approaches use more realistic contact deformation models allowing for elastic and plastic deformation as well as tangential tractions due to friction (Thornton et al., 1996) in order to calculate the interparticle contact force. So far, however, no Distinct Element Analysis has been carried out of a soft-sphere system in the presence of gas flow using the comprehensive contact mechanics models developed in the literature (see e.g. Johnson et al., 1973; Ning and Thornton, 1993). With the current computing power it is still not feasible to model a system of a million particles with a soft-sphere approach, which would be necessary to realistically model the breakage process in a fluidised bed jet. The DEM has, however, the potential to predict the breakage of individual particles in a complex hydrodynamic flow pattern with large velocity and solids concentration gradients. With a rapid increase in computer speed and memory, this should be resolved in the next few years.
CONCLUSIONS

Close observations by means of specially developed imaging facilities have enabled the identification of the effect of the orifice-to-particle size ratio on the hydrodynamics of fluidised bed jets, which has great relevance for the study of breakage of particulates in common unit operations, such as fluid catalytic cracking units, fluidised bed dryers, and jet milling operations. The interactions between the structure and mechanical properties of particles and the hydrodynamic regime in fluidised bed jets determine which mechanism dominates the breakage process. The particle structure and its performance in a unit operation is greatly affected by its process history of production, storage and handling.

In the case of a large orifice-to-particle size ratio, particles get entrained in a wide, dilute jet core. The breakage in this case is purely by collision, and can be simulated by single particle impact tests. In the case of a small orifice-to-particle size ratio, below 7.5-10, particles are subjected to a rapid shearing flow at the nozzle exit and the particles do not accelerate to high impact velocities at the top of the jet. The breakage in this case depends on the particle’s propensity to surface abrasion. In the case of particles with a low propensity to surface abrasion, the overall extent of breakage is very low. In the case of particles with a high propensity to surface abrasion, the extent of breakage is very high.

Attrition tests, such as the Forsythe and Hertwig (1949) test and the ISO test method for sodium perborates, which try to simulate the actual process conditions are found to have serious shortcomings. They are unsuitable for assessment of particle attrition, as they depend on the specific geometry and hydrodynamic conditions used in these tests, and do not allow for extrapolation to different geometries and types of mechanical stress.

Within certain limits of orifice-to-particle size ratio, the jet attrition model presents a suitable method for the prediction of particle breakage behaviour in different geometries and hydrodynamic conditions. Different breakage
mechanisms can be easily represented by incorporating the appropriate single particle test method. The single particle impact method is a suitable test for the assessment of the resistance of a particle to collisional damage and the breakage mechanism arising from that. For rapid shear flow, further work is required to develop a single particle test which can represent this mechanism. This work is on-going, and the possibilities include a single particle wear test and a shear cell test, repeated single particle impact tests and impact tests at an inclined target.

The Distinct Element Method has been of great use in elucidating the mechanisms by which agglomerate structures fail. This method has great potential in the application to dynamic systems, such as fluidised beds, and it is certainly worthwhile to develop this method further to predict the particle breakage in fluidised bed jets.

The dimensional analysis of the jet angle by Vaccaro (1997) has aided in the identification of the threshold value of the orifice-to-particle size ratio below which the jet dilute core at the nozzle exit ceases to exist. The threshold value appears to lie between 7.5 and 10. Further analysis has shown that the particle Froude number reflects the changing jet hydrodynamics better than the orifice Froude number.

There is still only a very limited understanding of the time-dependent rate effects, such as conversion by chemical reaction and surface abrasion, on attrition. The use of attrition rate constants has not been supported sufficiently by modelling theory to explain their physical meaning.

The use of the rate of elutriation as an indicator of the attrition rate must be applied with great care as there exist hydrodynamic conditions in which fines generated by attrition do not elutriate.

The identification of the breakage mechanism in particulate processing can benefit from analysis of the size distribution of the debris. The product size distribution
may have a single characteristic which is representative of a typical material response to the prevailing range of hydrodynamic conditions. An example of this characteristic is a constant slope of the trend line of the complement part of the particle size distribution, which has been found under a range of conditions.
FUTURE WORK

In order to move away from the present model and its substantial dependence on experimental input data, and to obtain more generally applicable and predictive models, a concerted effort should be made to incorporate fluid dynamics into the soft-sphere DEM approach of Thornton et al. (1996) and to use powerful computing facilities to realistically simulate the detailed, i.e. single particle, material response to hydrodynamic stresses.

In order to extend the present modelling approach to dense phase rapid shear flow, attempts should be made to include data from multiple and inclined single particle impact tests into the jet attrition model, instead of the present data from a single normal particle impact on a flat target, and to compare the predictions with experimental data from fresh FCC.

The level of understanding of breakage processes and the development of better predictive models should be enhanced. The use of techniques like the nano-indentation method for measurement of Young's modulus, fracture toughness, and hardness should become more widely used to generate a database that will allow correlation of a large number of material properties for the verification of mechanical models of solids fracture.

An important part of the study of single particle properties must be directed towards identification of the influence of the particle structure, such as crystal habit for crystalline solids, pore structure for porous solids, anisotropy of the composition, core-shell structure in agglomerates and granulates, surface roughness, agglomerate primary particle size distribution, and flaw distributions, on the breakage mechanism in different stress environments.

Further study of the parameters controlling the jet penetration length and the jet divergent angle should enable the clarification of the dependence of the jet geometry on the operating conditions.
There is some indication that there is a significant effect of particle shape on the entrainment rate of solids into the jet and the consequent breakage rates (Bentham, 1998). Further investigations should be directed towards identification of the dependence of the jet hydrodynamics on particle shape and ways of including this in models.

The interest in fundamental research in dense fluidised beds should be revamped in order to gain a better understanding of the interactions between the particle size and shape distribution of the fluidised assembly, or simply the fines content, and the fluidisation characteristics, such as voidage fluctuations, bubble structure, and fines elutriation. This will enable the verification of the use of the elutriate as a measure of the attrition rate, which is often not merely convenient but dictated by the limited possibilities to analyse the full bed inventory.

New test methods should be considered to replace the inadequate standard tests such as the Forsythe and Hertwig (1949), or the ISO 5937 test. Tests should not only be able to compare the performance of different materials, but should also represent the hydrodynamic conditions of the actual process in which these materials are going to be used. Alternatively, a suitable single particle test method must be used in conjunction with a hydrodynamic model to predict the assembly behaviour in the application.
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APPENDICES
Appendix A

A Model of Attrition in the Jetting Region of Fluidised Beds

A Model of Attrition in the Jetting Region of Fluidised Beds

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Abstract

Attrition causes material loss and environmental hazards in powder processing. In fluidised beds, the jetting region is the main contributor to attrition. The present paper reviews the recent investigations of the effects of the interaction between single particle properties and jet hydrodynamics. A model of attrition in the jetting region of fluidised beds is presented, based on the impact attrition propensity of single particles and on modelling of the particle flow patterns in the jetting region. The experimental work, carried out for the purpose of evaluation of the model, focused especially on the effects of orifice gas velocity and diameter. The experiments involve measurements of impact attrition of single particles and measurements of particle velocities and solids concentrations in fluidised bed jets. The test materials are fluid cracking catalyst and common salt, both representatives of widely used classes of composite and crystalline materials, respectively. Significant effects of orifice gas velocity and diameter were predicted, which corroborated the experimental data. Hence, the model successfully establishes a link between single particle properties and bulk behaviour in a fluidised jet.

1. Introduction

Attrition, the unintentional breakage of particulate solids during processing, handling and storage, causes material loss and environmental hazards. Fluidised bed operations have become increasingly popular in industrial particle processing, for the ease of solids handling, mixing and high rates of heat and mass transfer. These features are desirable for a number of processes, such as drying and reactor systems, e.g. fluid catalytic cracking and combustion. Unfortunately, the intensive particle motion in fluidised beds causes attrition at the same time. For example, material loss in fluidised
catalytic cracking units can amount to several tonnes per day! Zenz and Kelleher [1] have identified the distributor region and the cyclone as important sites for attrition due to the presence of high local velocity gradients. The distributor region provides by far the largest contribution to attrition in fluidised beds, when compared to the bubbling bed and the freeboard above it. Several studies have been undertaken in the past to characterise the attrition propensity of particles in fluidised beds [2], but few take account of the material properties of particles, or attempt to decouple the interacting hydrodynamic parameters. The dependence of the attrition rate in the jet region on the design and operating parameters, such as the distributor orifice size and gas velocity, and particle size and properties has so far not been established satisfactorily.

A number of empirical correlations for the rate of attrition in fluidised beds, $R_a$, have been reported in the literature. These are summarised in Table 1. Focusing on two main design and operation parameters, the orifice size, $d_{or}$, and the gas velocity, $u$, the correlations are generally written in the form of a power law:

$$R_a \propto u^n d_{or}^h$$

(1)

where $u$ can be the orifice gas velocity ($u_o$), or the superficial ($u_s$) or excess gas velocity ($u_s - u_{mg}$), with values of $n$ ranging from 0.66 to 5.8 and of $h$ from 0 to 1.11, as reported by different authors. The exponential correlation of Lin et al. [6] appears to be an exception to this general form, but replotting their data shows that there is no need for an exponential correlation, and that a good fit is also achieved with a linear regression. Notwithstanding this basic similarity, each of these correlations has been obtained for different materials, experimental set-up and operating conditions, making it difficult to compare them.

In this work, the attrition of common salt and fluid cracking catalyst (FCC) is considered, focusing in particular on the link between single particle properties and bulk attrition behaviour. These two material types, in addition to their own significance, may be considered as representatives of large classes of widely used materials, i.e. crystalline and composite structures. In this paper, the modelling approach will first be described. The experimental work supporting the development of the model is then presented.
2. **Modelling Approach**

In the present modelling approach, the impact breakage of single particles is coupled with a hydrodynamic model to predict the rate of attrition. The structure of the approach is represented schematically in Figure 1.

For impact attrition, Zhang and Ghadiri [16] have proposed the following correlation for the extent of attrition upon impact, $R_i$, based on the fracture mechanics of lateral crack formation:

$$ R_i = \alpha \frac{\rho_p u_i^2 H d}{K_c^2} $$

(2)

where $\rho_p$ is the particle density, $u_i$ the impact velocity, $H$ the hardness, $K_c$ the fracture toughness, $d$ a linear dimension of the particle, and $\alpha$ is a proportionality constant to be determined experimentally. In practice, the value of the power index of $u_i$ may differ slightly from 2, depending on the complexity of the particle structure. It is therefore more general to consider:

$$ R_i \propto u_i^m $$

(3)

where the power index $m$ may be obtained from particle impact tests.

In a fluidised bed with a jetting distributor region, the contribution of the bubbling zone to the attrition rate is usually very small compared to that of the jetting region [13]. The attrition mechanism in a fluidised bed jet involves the entrainment of particles into a dilute jet core, followed by the acceleration of the particles, whereafter they impact on the dense phase on top of the jet. Intense interparticle collisions are considered to cause attrition, at a rate that can be estimated from impacts of single particles on a rigid target, at an impact velocity, $u_i$, corresponding to the particle velocity, $u_p$, in the jet.

The number of particles engaged in the attrition process scales with the rate at which solids become entrained from the bulk into the dilute jet core. A hydrodynamic model may be used to obtain the dependence of $W_s$ and $u_p$ on the orifice gas velocity. This can be done by power-law correlations [12]:

$$ u_p \propto u_o^l $$

(4)
Ghadi\textit{r}i \textit{et al.} \cite{12} proposed that the attrition rate in the jetting region is linearly related to the single particle impact attrition and the solids entrainment rate. Thus, substituting Eq. (4) into Eq. (3), and multiplying by the solids entrainment rate, $W_s$, the attrition rate in a single jet may be given as:

$$R_a \propto W_s \propto u_o^k u_p^n \propto u_o^{k+1/n}$$

In this way, a descriptive and predictive model is established, which incorporates the single particle attrition characteristics. The results of this approach will be presented in section 4.

The dependence of the attrition rate $R_a$ on the orifice size, as reported in the literature, is inconsistent. The power index $h$ in Eq. 1 given by Kono \cite{8} and Ghadi\textit{r}i \textit{et al.} \cite{15} is of order unity, between 0.44 and 1.11. For comparison with other correlations that are expressed in different dimensions, \textit{e.g.} those of Werther and Xi \cite{11}, and Zenz and Kelleher \cite{1}, it is necessary to normalise the correlations, \textit{e.g.} by dividing the rate of attrition by the mass flow rate of fluidising gas. Following this approach, the correlations of Werther and Xi \cite{11} and Zenz and Kelleher \cite{1} indicate that the attrition rate does not depend on the orifice size.

For the analysis of the effect of the orifice size on the attrition rate $R_a$, Ghadi\textit{r}i \textit{et al.} \cite{15} have employed a similar approach as described above for the effect of orifice gas velocity. Normalising the solids entrainment rate with respect to the gas flow rate, $W_g$, its dependence on the orifice diameter, $d_{or}$, can be expressed in a power law \cite{15}:

$$W_s / W_g \propto d_{or}^t$$

and similarly for the dependence of the particle velocity:

$$u_p \propto d_{or}^t$$

Following the same approach as for the effect of velocity (Eq. 6), the attrition rate in the jet can expressed as:

$$R_a \propto \left(W_s / W_g\right) R_i \propto d_{or}^t u_p^n \propto d_{or}^{t+1/n}$$

Thus, the dependence of the attrition rate on the orifice size can be established by quantifying the power indices. This is described in section 4.
Independent studies of the hydrodynamics of fluidised bed jets by Massimilla and co-workers, firstly introduced by De Michele et al. [17], have provided comprehensive models of particle flow patterns in the jetting region [18]. With these hydrodynamic models, particle velocities and solids entrainment rates can be readily obtained for different orifice velocities and sizes. However, the application of these models requires knowledge of the relevant input parameters, describing the jet geometry, such as the jet penetration length, the jet divergent angle, and the solids concentration in the jet. These have to be obtained from separate experiments, which are described in sections 3.2 and 3.3.

3. Experimental

Several tests have been employed in the evaluation of the jet attrition model. The experiments involve firstly single particle impact tests for the determination of the impact attrition of single particles as a function of the impact velocity, as shown in Eq. 2. This is described in section 3.1. Secondly, the attrition rate in a fluidised bed with several gas distributors has been measured in order to compare these results with predictions from the attrition model. This is described in section 3.2. In order to quantify the power indices given in equations (6) and (9), it is first necessary to specify a number of hydrodynamic parameters, such as the jet angle. This is described in section 3.3.

3.1 Single particle impact test

Single particle impact tests have been carried out in an impact test rig developed previously [19] and shown in Fig. 2. It consists of a funnel-shaped inlet section guiding the particles into an eductor tube, which ends in a collection chamber, where particles impact on a rigid horizontal target plate made of sapphire. The particle velocity before impact is measured with dual light diodes or with a laser Doppler velocimeter.

The impact product is analysed gravimetrically. As a criterion for attrition, the mass of debris passing through a sieve, with a size of two BS410 sieve sizes below the lower limit of the original size, is chosen. In the low range of impact velocities, where
mainly surface damage (chipping) occurs, the fines produced will be far smaller than the original mother particles. Cleaver et al. [20] have shown that the attrition results are in this case insensitive to the specific criterion applied, as long as the sieve size, used for the separation of debris from the mother particles, lies in between the particle size distributions of the Fine Product and Coarse Product, as shown schematically in Fig. 3. The attrition rate is then simply defined as the ratio of the mass of fine product to the initial sample mass of mother particles:

\[ R_i = \frac{M_{\text{fine product}}}{M_{\text{mother particles}}} \]  

(10)

3.2 Fluidised bed attrition test

Fluidised bed attrition tests have been carried out for the determination of the variation of attrition with orifice gas velocity and size.

Two materials have been investigated, Pure Dried Vacuum (PDV) NaCl salt, produced by ICI plc, mean particle size \(d_p = 418 \, \mu\text{m}\), with a density of 2180 kg m\(^{-3}\), and FCC, \(d_p = 106 \, \mu\text{m}\), with a density of 1500 kg m\(^{-3}\). For FCC, the data of Werther and Xi [11] were used. The experimental set-up used for fluidised bed attrition tests of salt is shown in Fig. 4. A detailed description can be found elsewhere [13]. Three different perforated plate distributors were used, with 73, 110, and 175 holes of 1 mm diameter, i.e. each with a different free area. This enabled the decoupling of the effect of superficial and orifice gas velocity on the attrition rate, since the bed could be operated at the same superficial velocity for three different orifice gas velocities. According to the correlation of Fakhimi and Harrison [21], all grid holes were active in all conditions, so that good distribution of air across the distributor was ensured.

The contributions of the jetting region and of the bubbling part of the bed to the attrition rate have been decoupled by operating the bed at different loading, yielding different bed heights. By plotting the attrition rate against bed height and extrapolating to a height equal to the jet height, the contribution of the jetting region to attrition has been quantified.

The effect of distributor orifice diameter on the attrition rate has been investigated by Ghadiri et al. [15] using salt as the test material in a 58 mm diameter cylindrical Perspex bed of 0.9 m height, otherwise similar to the set-up shown in Fig. 4. The results are presented in section 4.
3.3 Measurement of jet parameters

In the hydrodynamic jet model of De Michele et al. [17], which is employed here, the particle flow patterns in the jet are calculated on the basis of Schlichting similarity profiles, following the turbulent jet theory of Abramovich [22]. These profiles scale with the actual geometry of the jet, as it extends axially and expands radially into the bulk of the bed. To establish these profiles for a given orifice size and velocity, several parameters, such as the jet angle and height, and the initial solids concentration at the jet exit need to be specified. All these parameters are difficult to measure, especially in a three-dimensional bed. A number of measurements have been carried out in two-dimensional and three-dimensional (cylindrical) fluidised beds, using various measurement techniques [12-15, 23-25]. The measurements with two-dimensional beds are only used for input in the two-dimensional version of the hydrodynamic model. However, comparison of the model predictions with the experimental data from two-dimensional configurations can be used to check the validity of the current approach.

Measurements of jet angles in the cylindrical fluidised bed, described in section 3.2, have been carried out using an X-ray facility using salt, $d_p = 418 \mu m$, and alumina particles, $d_p = 107 \mu m$ [25].

Digital analysis of video images taken from a two-dimensional fluidised bed, shown in Fig. 5, has been used for measurements of jet half angles with FCC catalyst, $d_p = 90 \mu m$. In this apparatus, the gas jet is produced by slots, sandwiched between two porous plates for background fluidisation [23].

In this two-dimensional set-up, a strong dependence of the jet angle on orifice gas velocity has been found, even with a narrow orifice of 0.5 mm, where the jet half angle decreases in a linear fashion from $11.5^\circ (u_o = 9.4 \text{ m s}^{-1})$ to $7^\circ (u_o = 54 \text{ m s}^{-1})$. The trends for different orifices behave like tangents to a single hyperbolical curve, as shown in Fig. 6. There is, however, a certain overlap in the gas velocity range, where the trendlines for neighbouring jet sizes cross over each other, indicating a distinct effect of the orifice size, rather than just a velocity effect. Similar effects, but to a different extent, were observed in the three-dimensional set-up [25].

Measurements of the initial solids concentration in the jet have been made using the two-dimensional fluidised bed, shown in Fig. 5, which was equipped with optical glass walls for the specific purpose of video recordings with a high resolution camera.
Results of these measurements are shown in Fig. 7. The solids concentration at the nozzle exit appears to increase with decreasing orifice size. This is supported by our visual observations [24] that with wide orifices, the jet core is clear of solids, whereas with narrow orifices the jet core is occupied by a significant amount of solids, even at high $u_o$.

In order to compare the hydrodynamic model predictions with the actual solids flow patterns, measurements of particle velocities have been carried out in the two-dimensional bed equipped with optical glass walls, for the FCC and the NaCl salt. The results are shown in Figs 8 and 9 [23, 24]. These measurements have been obtained, using digital image analysis of video images of the particle flow, recorded with a Kodak high speed video camera at frame rates up to 40500 frames per second.

For FCC (Fig. 9), a good agreement is shown between experimental data and predictions from the hydrodynamic model. For NaCl (Fig. 8), the agreement is fair, but the data are more closely matched for large orifice diameters. Considering the observation that NaCl particles do not accelerate significantly along the jet height for narrow orifices, because of the increased solids concentration, this feature suggests that there is a distinct effect of the ratio of particle-to-orifice diameter on the flow pattern, which is currently not accounted for in the model.

4. Attrition model application and evaluation

4.1 Effect of velocity on single particle impact attrition

The first step in the application of the attrition model is the establishment of the power index $m$ in Eq. (3). The single particle impact tests show a slight variation from the theoretical value of 2 for some materials. Table 2 shows measured values of $m$ for melt-grown and solution grown NaCl crystals, and for FCC. The solution grown NaCl crystals contain polycrystals and crevasses, which contributes to the deviation of $m$ from 2. The index $m$ is sensitive to internal and surface defects and structure, the presence of polycrystals, work-hardening and fatigue [13, 14, 16, 26].
4.2 Effect of distributor orifice size and velocity on fluidised jet attrition

The measurement of the fluidised bed jet attrition as a function of orifice velocity and size involved decoupling of a number of concurrent processes, such as the attrition in the jetting region and in the bubbling part of the bed. As described in section 3.2, experiments with different bed heights have been carried out to enable the establishment of a correlation between the bed height and the attrition rate, from which the attrition in the jetting region may be inferred. The result of this operation is shown in Fig. 10, where the data points are accompanied by a trendline of a best fit with a power index yielding a value of \( n \) in Eq. (6), as reported in Table 3.

The entrainment rate and particle velocity in the jet, as predicted by the hydrodynamic jet model, are shown in Figs 11 and 12 for NaCl for a 1.0 mm orifice diameter. From the curve fits, values of the power indices \( k \) and \( l \), for the particle velocity and the solids entrainment rate, respectively, as given in Eqs (4) and (5), may be obtained. These are given in Table 3, together with the value of \( m \), from Eq. (3), for the NaCl particles. The overall attrition index \( n \) (Eq. 6) is then calculated and is given in Table 3.

The predicted results compare favourably with experimental data for NaCl and FCC, also shown in Table 3. The experimental value of \( n \) for FCC is that given by Werther and Xi [11]. Zenz and Kelleher [1] report a slightly lower value of 2.5 for larger FCC particles. Values of \( k \) and \( l \) for FCC have been obtained using the experimental parameters given by Werther and Xi [11] and \( m \) has been obtained by single particle impact testing [14] using the same material as used by Werther and Xi [11].

The predicted dependence of the attrition rate on the orifice diameter is shown in Table 4, where the values of \( s \), \( t \) and \( h \) have been given (see Eqs (7)-(9)). The values of \( h \) compare reasonably well with the experimental values of Kono [8] and Ghadiri et al. [15], but contradict with the absence of any effect of the orifice size, as indicated by Werther and Xi [11] and Zenz and Kelleher [1]. Variations in the level of background fluidisation may be responsible for this difference (see Table 1).
5. Discussion

The model of jet attrition combines a model of particle breakage with a hydrodynamic model of particle flow in a jet in order to predict the rate of attrition. The experimental observations of particle velocity and concentration profiles suggest that there is an influence of the orifice-to-particle size ratio, and this is currently not taken into account in the hydrodynamic model.

For orifices much larger than the particle size, the core of the jet is almost entirely clear of particles, whereas for orifices of size comparable to or smaller than the particle size the solids concentration in the jet was observed to be high. This strongly influences the particle-particle interactions. For the latter, at high orifice velocities, the particles will shear against each other, causing abrasion, whereas for the former, particles may accelerate freely, causing more integral damage, thus increasing the probability of fragmentation. Further refinement of the model is necessary to incorporate the effect of the ratio of orifice diameter to particle size.

There are several simplifying assumptions in the attrition model, whose validity may have to be assessed for particles of interest. These are as follows.

i) It is assumed that the dependence of interparticle impact damage on impact velocity follows the same trend as the impact of a particle on a rigid target. The latter may provide a more extensive damage as the target is rigid. However, the effect of impact velocity is not expected to be different from interparticle collisions.

Generally, the collision frequency in a fluidised bed is very high. The high speed video recordings of NaCl particles showed that particles in fact accelerate along the jet axis, which is almost clear of particles, and subsequently impact on the dense phase on top of the jet. The particles often scour the jet boundary as if they had been launched into a pin ball machine. However, most of the momentum is dissipated during the first impact, and the subsequent collisions take place at a velocity similar to the recirculation velocity of particles in the bulk. Therefore, the actual process of attrition will be contained in the first particle-bulk collision, and the similarity between the single impact and the jet impact is preserved.

ii) The influence of time is shown in Fig. 13 for NaCl particles. This effect has not been considered in several investigations, where tests have been performed for one hour only [3, 27], or even shorter periods [10]. Cairati et al. [28] have shown that
molybdate catalyst particles do not reach steady state equilibrium within one hour in
the Forsythe-and-Hertwig test [27], and the present NaCl particles reach steady state
only after five hours.

iii) Impact breakage studies have shown that repeated impacts at the same velocity
may progressively either weaken or strengthen the particles, causing the attrition rate
to vary with the number of impacts [29]. Plastic deformation of semi-brittle particles
may cause work-hardening, which eventually leads to an increase in the attrition rate.
On the other hand, the first few impacts may cause weaker particles to break, whence
the remainder would appear to be more resistant to attrition.

iv) In the analysis of attrition, Ghadiri et al. [12-15] have carried out a complete
analysis of the bed inventory, accounting for the debris in the bed. In a number of
previous investigations, the attrition rate is quantified by the amount of fines elutriated
from the bed. This process ignores the quantity of the debris [30] which is in dynamic
equilibrium in the bed [31]. This may be a source for discrepancies in the trends
reported in the literature.

The power index $m$ for single particle impact damage does not vary widely from
its theoretical value of 2 for FCC and different types of NaCl. However, the overall
attrition index varies from 3 to about 5! It is interesting to note that the present
approach is capable of identifying the relevant hydrodynamic parameters to predict the
actual power index for attrition in the processing environment.

6. Conclusions

The model of attrition in fluidised bed jets provides a realistic prediction of the effect
of a number of important design and operating parameters such as orifice size and gas
velocity. The model takes account of the particle properties and hydrodynamic
conditions of the jet. The procedure established to estimate the attrition rate in the
jetting region of fluidised beds is to obtain a measure of the attrition propensity of
single particles by impact testing and to couple this process with the rate of solids
entrainment into the jetting region by the use of hydrodynamic modelling. The model
has been successfully applied to two very different types of material, FCC powder and
NaCl crystals. Further testing with materials such as weakly-bonded agglomerates and resins would be useful to establish the range of the applicability of the model.

7. Acknowledgement

Financial support from the University of Surrey and Shell Research B.V. is gratefully acknowledged. The authors are grateful to EPSRC and Mr Peter Goodyer for providing quick access to the high speed video and photography facilities from EPSRC's equipment pool. Dr W. Duo is thanked for his valuable comments on the manuscript.

8. Nomenclature

\begin{align*}
C & \quad \text{Constant} \quad \text{(see Table 1)} \\
d & \quad \text{linear particle dimension} \quad \text{(m)} \\
d_{or} & \quad \text{distributor orifice size} \quad \text{(m)} \\
d_p & \quad \text{mean particle size} \quad \text{(m)} \\
d_{p0} & \quad \text{initial mean particle size} \quad \text{(m)} \\
f & \quad \text{distributor free area} \quad \text{(-/-)} \\
h & \quad \text{power index (Eq. 1)} \quad \text{(-/-)} \\
H & \quad \text{hardness} \quad \text{(Pa)} \\
HGI & \quad \text{Hardgrove Grindability Index} \quad \text{(kg)} \\
k & \quad \text{power index (Eq. 5)} \quad \text{(-/-)} \\
K_c & \quad \text{fracture toughness} \quad \text{(N m}^{3/2}\text{)} \\
l & \quad \text{power index (Eq. 4)} \quad \text{(-/-)} \\
m & \quad \text{power index (Eq. 3)} \quad \text{(-/-)} \\
M & \quad \text{mass} \quad \text{(kg)} \\
M_b & \quad \text{total bed weight} \quad \text{(kg)} \\
n & \quad \text{power index (Eq. 1)} \quad \text{(-/-)} \\
Q & \quad \text{volumetric orifice gas flow rate} \quad \text{(m}^3\text{ s}^{-1}\text{)} \\
R_a & \quad \text{fluidised bed jet attrition} \quad \text{(-/-)} \\
R_{bed} & \quad \text{fluidised bed bulk attrition} \quad \text{(-/-)}
\end{align*}
\[ R_i \] single particle impact attrition
\[ s \] power index (Eq. 7)
\[ t \] power index (Eq. 8)
\[ u_i \] particle impact velocity
\[ u_{mf} \] minimum fluidisation velocity
\[ u_o \] orifice gas velocity
\[ u_p \] particle velocity in the jet
\[ u_s \] superficial gas velocity
\[ W \] residue bed weight
\[ W_c \] carbon loading
\[ W_s \] solids entrainment rate
\[ W_g \] gas mass flow rate
\[ z \] number of grid jet holes
\[ \beta \] correction factor
\[ \rho_f \] fluid density
\[ \rho_p \] solids density
\[ \sigma_{br} \] crushing strength
\[ \tau \] processing time
\[ \phi_s \] particle sphericity
\[ \phi_{so} \] initial particle sphericity

9. References

3) Blinichev, V.N., V.V. Strel'tsov and E.S. Lebedeva: Intern. Chem. Engng, 8 (4), 615 (1968)


29) Zhang, Z., PhD Thesis, Univ. of Surrey, 1994


<table>
<thead>
<tr>
<th>Authors</th>
<th>Modelling Equation</th>
<th>Dim. of Attrition Rate</th>
<th>Material</th>
<th>Time of Operation (hr)</th>
<th>Range of ( u_r ) (m s(^{-1}))</th>
<th>Range of ( u_o ) (m s(^{-1}))</th>
<th>Grid Type</th>
<th>Background Fluidisation</th>
<th>Attrition Debris Criterion</th>
</tr>
</thead>
<tbody>
<tr>
<td>Blinichev et al. [3]</td>
<td>( R_a = \frac{90 \varphi_s u_s^{5.8} \rho_f^{2.6} 10^{0.21 d_o/d_p-18 f}}{d_p^{1.9} \sigma_{br}^{1.3} \rho_p^{1.5}} )</td>
<td>s(^{-1})</td>
<td>3-5 mm NaCl, 3-5 mm nitroforsk, 3-5 mm silica gel</td>
<td>1</td>
<td>0-11</td>
<td>20-240</td>
<td>1-8 mm holes in perforated plate</td>
<td>None</td>
<td>Elutriate</td>
</tr>
<tr>
<td>Merrick and Highley [4]</td>
<td>( R_a = C M_b \left( u_s - u_{mf} \right) )</td>
<td>kg s(^{-1})</td>
<td>3 types of coal 1.59 and 3.18 mm diameter</td>
<td>100</td>
<td>0.67-2.67</td>
<td>not indicated</td>
<td>not indicated</td>
<td>0.67-2.67 m s(^{-1})</td>
<td>Elutriate (smaller than 63 ( \mu )m)</td>
</tr>
<tr>
<td>Chen et al. [5]</td>
<td>( R_a = \frac{C \rho_f Q \left( \beta u_o \right)^2 f(d_p, \varphi_{so})}{W d_p \rho_p} )</td>
<td>s(^{-1})</td>
<td>115-274 ( \mu )m siderite iron ore and 210 ( \mu )m lignite char</td>
<td>up to 24</td>
<td>0.2-0.5</td>
<td>up to 300</td>
<td>1.47-3.18 mm single jet in porous plate</td>
<td>Variable</td>
<td>Elutriate (smaller than 75 ( \mu )m)</td>
</tr>
<tr>
<td>Lin et al. [6]</td>
<td>( R_a = C \exp \left( 0.162 \left( u_s - u_{mf} \right) \right) )</td>
<td>s(^{-1})</td>
<td>mixtures of 133-354 ( \mu )m Silica sand and 10-113 ( \mu )m char</td>
<td>5</td>
<td>0.1-0.32</td>
<td>up to 53.5</td>
<td>not indicated</td>
<td>None</td>
<td>Elutriate</td>
</tr>
<tr>
<td>Zenz and Kelleher [1]</td>
<td>( R_a = C \left( u_o \sqrt{\rho_f} \right)^{2.5} \frac{\pi d_{pr}^2}{4} )</td>
<td>kg s(^{-1})</td>
<td>200 ( \mu )m FCC</td>
<td>12-80</td>
<td>33-303</td>
<td>492 holes in triangular pitch</td>
<td>Downward pipe holes 0.8-19 mm wide</td>
<td>Variable</td>
<td>Elutriate</td>
</tr>
<tr>
<td>Author(s)</td>
<td>Equation</td>
<td>Units</td>
<td>Description</td>
<td>Details</td>
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<tr>
<td>Donsi et al. [7]</td>
<td>( R_a = C \left( u_s - u_{mf} \right) \frac{W_c}{d_p} )</td>
<td>kg s(^{-1})</td>
<td>Coal in pipe holes</td>
<td>0.4-3.0 mm not indicated, 0.55-1.3 82-135 76 upward pipe holes, 1.5 mm wide</td>
<td></td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>Kono [8]</td>
<td>( R_a = C \rho_f u_o^3 d_{or}^{0.55} ) for ( u_o \leq 3.6 ) m s(^{-1}) ( R_a = C \rho_f u_o^2 d_{or}^{0.55} ) for ( u_o &gt; 3.6 ) m s(^{-1})</td>
<td>s(^{-1})</td>
<td>0.97-4.00 mm Mullite Alumina-Silicates 8-12 0.45-8.0 0.5-30 3.6-15.5 cm single tapered jet None</td>
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</tr>
<tr>
<td>Sishtla et al. [9]</td>
<td>( R_{bed} = 1.6 \cdot 10^{-3} \left( u_s - u_{mf} \right)^{0.66} (HGI)^{-0.05} ) ( R_a = 2.8 \cdot 10^{-12} \left( \rho_f^{1.56} u_o^{3.12} \right) (HGI)^{-2.15} )</td>
<td>s(^{-1})</td>
<td>500-841 ( \mu )m char 14 0.3-0.6 67-134 6 bubble caps with 3 orifices, 0.24 cm wide, 30° downward None</td>
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<td></td>
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<tr>
<td>Seville et al. [10]</td>
<td>( R_a = C \left( u_s - u_{mf} \right) )</td>
<td>kg s(^{-1})</td>
<td>1.18-2.80 mm sand agglomerates up to 0.25 0.15-1.15 74-111 139 holes, 1.5 mm wide on 12 mm triangular pitch Variable</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Werther and Xi [11]</td>
<td>( R_a = C \rho_f d_{or}^{2} u_o^{3} )</td>
<td>kg s(^{-1})</td>
<td>106 ( \mu )m spent FCC, 125 ( \mu )m fresh FCC up to 260 0.2 25-100 0.5-2.0 mm single jet in porous plate Elutriate (smaller than 23-35 ( \mu )m)</td>
<td></td>
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<td></td>
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<tr>
<td>Ghadiri et al. [12-15]</td>
<td>( R_a = C u_o^n d_{or}^h ) with ( n = ) {5.1 for NaCl 3.31 for FCC} and ( h = ) {0.44 - 1.11 for NaCl 0.6 - 0.76 for FCC}</td>
<td>hr(^{-1})</td>
<td>425-600 ( \mu )m NaCl, 90-106 ( \mu )m FCC 10-22 0.85 25-125 73, 110 and 175 3.0 mm holes in triangular pitch None</td>
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</table>
Table 2. Impact attrition power indices

<table>
<thead>
<tr>
<th>Material</th>
<th>$m$</th>
</tr>
</thead>
<tbody>
<tr>
<td>NaCl melt-grown</td>
<td>2.0</td>
</tr>
<tr>
<td>NaCl solution-grown</td>
<td>2.6</td>
</tr>
<tr>
<td>FCC</td>
<td>2.34</td>
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</table>

Table 3. Fluidised bed hydrodynamic and attrition power indices

<table>
<thead>
<tr>
<th>Parameter</th>
<th>NaCl</th>
<th>FCC</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k$</td>
<td>1.9</td>
<td>1.3</td>
</tr>
<tr>
<td>$l$</td>
<td>0.9</td>
<td>0.9</td>
</tr>
<tr>
<td>$m$</td>
<td>2.6</td>
<td>2.3</td>
</tr>
<tr>
<td>$n$</td>
<td>4.3</td>
<td>3.3</td>
</tr>
<tr>
<td>$n$ (exp.)</td>
<td>4.6</td>
<td>3.0 [11]</td>
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</tbody>
</table>

Table 4. Hydrodynamic and attrition parameters for the effect of the orifice diameter

<table>
<thead>
<tr>
<th>$u_o$</th>
<th>FCC $h = s + t \cdot m$</th>
<th>NaCl $h = s + t \cdot m$</th>
</tr>
</thead>
<tbody>
<tr>
<td>25</td>
<td>-0.006 0.41 0.95</td>
<td>-0.004 0.44 1.11</td>
</tr>
<tr>
<td>75</td>
<td>0.06 0.3 0.76</td>
<td>0.11 0.28 0.84</td>
</tr>
<tr>
<td>125</td>
<td>0.17 0.182 0.6</td>
<td>0.18 0.1 0.44</td>
</tr>
</tbody>
</table>
Figure 1. Schematic representation of the modelling approach.

Figure 2. Air eductor single particle impact rig.
Figure 3. Particle size distribution for chipping attrition.

Figure 4. Fluidised bed used for attrition tests.
Figure 5. Two-dimensional fluidised bed for high-speed video imaging of jets.

Figure 6. Jet angles in a fluidised bed jet of FCC \(d_p = 90 \mu m\) as a function of \(u_o\) for different orifices.
Figure 7. Solids concentration in a fluidised bed of FCC ($d_p = 90 \mu m$) at the orifice exit as a function of $u_o$ for different orifices.

Figure 8. Comparison of numerical predictions (lines) and experimental measurements (markers) of particle velocities of NaCl particles in jets from different nozzles.

$\times$, $-$ $-$ $d_{or} = 8.0$ mm; $\bigcirc$, $-$ $-$ $d_{or} = 5.0$ mm;
$\blacktriangle$, $-$ $-$ $d_{or} = 0.5$ mm; $\square$, $-$ $-$ $d_{or} = 0.2$ mm.
Figure 9. FCC particle velocity in the jet from a 0.5 mm jet nozzle.

Figure 10. Variation of the attrition per jet with jet velocity for the jetting region of a fluidised bed containing PDV salt, calculated at a jet height estimated from the Yang and Keairns [32] correlation.
Figure 11. Predicted variation of the solids entrainment rate $W_s$ in a fluidised bed of NaCl with the orifice gas velocity from a 1.0 mm orifice.

Figure 12. Prediction of particle velocity as a function of orifice velocity from a 1.0 mm orifice in a fluidised bed of NaCl.
Figure 13. Time dependent attrition rate of common PDV salt in a jetting fluidised bed [13].
Appendix B

Attrition of Fluid Cracking Catalyst in Fluidised Beds

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ATTRITION OF FLUID CRACKING CATALYST IN FLUIDISED BEDS

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ABSTRACT

Particle attrition in fluid catalytic cracking units causes loss of catalyst, which could amount to a few tonnes per day! The dependence of attrition on the process conditions and catalyst properties is therefore of great industrial interest, but it is however not well established at present.

The process of attrition in the jetting area of fluidised beds is addressed and the attrition test method of Forsythe & Hertwig is analysed in this paper. This method is used commonly to assess the attrition propensity of FCC powder, whereby the attrition rate in a single jet at very high orifice velocity (300 m s⁻¹) is measured. There has been some concern on the relevance of this method to attrition in FCC units. Therefore, a previously-developed model of attrition in the jetting region is employed in an attempt to establish a solid basis of interpretation of the Forsythe & Hertwig test and its application as an industrial standard test. The model consists of two parts. The first part predicts the solids flow patterns in the jet region, simulating numerically the Forsythe & Hertwig test. The second part models the breakage of single particles upon impact. Combining these two models, thus linking single particle mechanical properties to macroscopic flow phenomena, results in the modelling of the attrition rate of particles entrained into a single high speed jet. High speed video recordings are made of a single jet in a two-dimensional fluidised bed, at up to 40500 frames per second, in order to quantify some of the model parameters. Digital analysis of the video images yields values for particle velocities and entrainment rates in the jet, which can be compared to model predictions.
INTRODUCTION

The mechanism of attrition in fluidised bed operations is affected by a number of parameters, which cannot easily be investigated independently. Zenz and Kelleher (1980) have identified the grid jet and the cyclone as the main sites of importance for their large contribution to attrition due to high local velocity gradients. The grid jet provides by far the largest contribution to attrition inside the fluidised bed, when compared to the bubbling bulk and the freeboard above it, which contribute to attrition mainly via bubble entrainment and ejection, respectively (Bemrose and Bridgwater, 1987). Several studies have been undertaken in the past to characterise the attrition propensity of FCC. Forsythe and Hertwig (1949) - hereafter referred to as F&H - have developed a test to estimate the friability of FCC particles, which should in principle match the hydrodynamics of an actual fluidised bed operation. Their method prescribes that a sample of particles be subjected to fluidisation by a single jet of 1/64 in diameter in a cylindrical bed, operated at an orifice velocity of 300 m s$^{-1}$. This test has been adopted as a standard test for attrition of FCC and many other materials. In fact, an ISO standard (ISO 5937, formerly BS 5688, part 26) has been derived from this test for sodium perborate particles, used e.g. in soap powders.

During fluidisation, particles loose their angularity due to interparticle collisions, which results in a high initial attrition rate, decreasing rapidly to an equilibrium rate after approximately 10-24 hours. The duration of the F&H test is only one hour. This is very short when compared to the life-cycle of FCC in industrial operation, which is usually up to three years, and when compared to the time necessary for the attrition rate to reach equilibrium. Cairati et al. (1980) employed the F&H test to assess the friability of iron molybdate based catalysts. They found that the catalyst does not reach equilibrium within the prescribed test duration, yielding a comparison of the equilibrium attrition behaviour of different materials on the strict basis of this test almost impossible. Gwyn (1969) used a modified F&H apparatus, with three grid jets in stead of a single one, fluidising FCC for up to 20 hrs, for the derivation of his well-known exponential time law of attrition.
In the recent past, Ghadiri et al. (1992) have developed a model for attrition in the jetting region of fluidised beds. This model combines experimental measurements of the attrition propensity of single particles upon impact with a hydrodynamic simulation of the particle flow in the jetting region, developed by Donsl et al. (1980) based on turbulent jet theory (Abramovich, 1963). This model has been employed successfully to explain the trend in attrition measurements of FCC particles in fluidised beds with a grid jet region (Werther and Xi, 1993; Ghadiri et al., 1994). This important result establishes the usefulness of the single impact test as an indicator of particle failure mode and friability in pneumatic conveying processes based on single particle properties.

The present study sets out to elucidate the hydrodynamics of the F&H test, to assess its usefulness for general application to industrial processes. The hydrodynamic jet model of Donsl et al. (1980) has been used to simulate the conditions of the F&H test, to predict the particle flow patterns. An experimental study in a two-dimensional fluidised bed has been carried out to verify the usage of this model. Jet angles and solids concentration profiles have been measured to serve as input, and particle velocities have been measured to be compared with output of the hydrodynamic model. In the following, results of the experimental study will be presented first, together with the model predictions for the applied experimental conditions. Thereafter, the model predictions of the F&H test will be presented and discussed in the light of the application of this test to industrial processes.

**Experimental**

The experimental set-up is shown in Figure 1. The lower section of the fluidised bed is made of a 12.5 mm thick optical glass sheet, 1.5 m high and 0.25 m wide. The internal bed width is 12.5 mm. A central aluminium nozzle is set flush in between two brass porous plates, which can be slid sideways in order to accommodate a range of differently sized nozzles. The nozzle is set in chamber, made of Perspex, below the porous plates, which provide an even distribution of the flow for background fluidisation. The chamber and the nozzle are both fed with compressed air from separate rotameters.
nozzle has the shape of a slot across the width of the bed, and the dead zone at each side of the slot is 0.5 mm because of its wall thickness. The slots of the nozzles used in this work were 0.5, 1.0, 5.0 and 8.0 mm wide. Orifice velocities ranged up to 54 m s⁻¹. The upper section of the bed is made of 12.5 mm thick Perspex sheet, on the inside flush with the lower section, and 0.43 m high. Entrained solids in the particle size range 30-60 μm are returned to the bed by a cyclone with a horizontal vortex, smaller particles are caught by a 0.2 μm absolute retention Sartopure¹ pleated paper filter. Fresh FCC particles, \( d_p = 90 \, \mu m, \rho_p = 850 \, kg \, m^{-3} \) have been used. The particles are fairly angular.

![Figure 1. Experimental apparatus.](image1)

![Figure 2. 3D Luminance Table.](image2)

A Kodak High Speed Motion Analyser HS 4540 video camera was used to record particle motion in the jet, at frame rates up to 40500 frames per second. A particle can be identified in subsequent images. Its position can be determined and at a given frame rate the particle velocity can be calculated. With a viewport of calibrated size of 0.5 mm, particle velocities up to 22 m s⁻¹ can be measured. Video images are downloaded to a PC equipped with an Optimas² automatic image analysis system.

¹Sartopure is a registered trademark of Sartorius AG, Goettingen Germany
²Optimas is a registered trademark of Optimas Corp., Seattle USA
For measurement of the divergent jet angle and the solids concentration in the jet, a Sony DXC930 3CCD video camera was used to capture macro images of the jet. Firstly, an image is taken of the still bed, used as a reference. Using Optimas, the jet image is subtracted from the background image. The jet region has a lower grayscale value, i.e. shows darker, than the bulk, so that the result of the subtraction from the background image is a totally black image with an illuminated jet area. The luminance or grayscale value is assumed to be proportional to $(e-e_{mf})$, i.e. the voidage taken relative to the bulk voidage, as supported by experimental work of Poletto et al. (1995). Figure 2 shows an example of the 3D luminance table of the jet area just above a 0.5 mm jet orifice, in which the solids concentration has been set out for an area of only $3.0 \times 3.0$ mm.

The hydrodynamic jet model of De Michele et al. (1976) represents the two-dimensional jet in a fluidised bed of fine particles in a similar fashion to the model of Donsi et al. (1980) for three-dimensional beds. These models have been used here for the 2D and 3D simulations respectively, using the same particle properties as in the experimental study. The models require input of the initial solids concentration of particles as an initial condition, and of the jet half angle as a geometrical parameter (see Figure 3). The jet height, another input parameter, is calculated with the correlation of Yates et al. (1986).

Figure 4 shows the results of measurements of the initial solids concentration as a function of $u_{or}$ for different orifice sizes. Measurements of the jet angle are reported in Figure 5. The trends for different orifices behave like tangents to a single hyperbolic curve. There is, however, a certain overlap in the gas velocity range, where the trend lines for different jet sizes cross over each other, indicating a distinct effect of the orifice size, rather than just a velocity effect. This pertains to the interaction of jet angle, size and velocity, indicated by Ghadiri et al (1995) and Cleaver et al. (1995). Jet angles measured by Cleaver et al. (1995), using an X-ray technique in a three-dimensional fluidised bed of spent FCC of 90-125 μm and orifices of 0.5 and 1.0 mm do not show any dependence on $u_{or}$ in the range of 20-60 m s$^{-1}$, but are almost constant, about $7^\circ$. In two dimensions, a strong dependence of jet angle on orifice gas velocity is found, even
with narrow orifice slots of 0.5 mm where the jet half angle decreases in a linear fashion from 11.5° ($u_{or} = 9.4 \text{ m s}^{-1}$) to 7° ($u_{or} = 54 \text{ m s}^{-1}$).

Apart from the jet half angle and the solids concentration, also particle velocities have been measured with the 0.5 mm orifice, which matches closely the orifice size of the F&H test. Figure 6 presents the results of these measurements together with the predictions from the hydrodynamic jet model. The predictions compare very well to the measured values, both with respect to the values and with respect to the trend.

In our experimental set-up, higher orifice velocities up to the Forsythe and Hertwig test conditions could not be achieved. We can at present only juxtapose, however, that the model provides as good a prediction in those conditions as it does in the conditions verified in the present experiments.

**NUMERICAL MODELLING OF THE F&H TEST**

Assuming that the hydrodynamic model gives correct predictions when the right input parameters are provided, the problem remains that for the F&H test conditions

![Figure 4. Solids Concentration as a function of $u_{or}$ for different orifices.](image)

![Figure 5. Jet angles as a function of $u_{or}$ for different orifices.](image)

![Figure 6. Particle Velocity in the Jet from a 0.5 mm Jet Nozzle](image)
the solids concentration and the jet angle have not been measured. We have therefore carried out simulations for a number of jet angles, as shown in Figures 7 and 8, using the model of Donsi et al. (1980). We have assumed a value of 0.05 for the initial solids concentration, in line with our experimental observations.

As the jet angle narrows down, less momentum from the jet fluid is dissipated, resulting e.g. in less particle entrainment, and the particles that do become entrained are accelerated to a greater extent, achieving higher particle velocities in the jet. With the experimental evidence, shown in Figure 5, we may safely assume that the jet angle for an orifice velocity of 300 m s\(^{-1}\), as in the F&H test, will be close to 5\(^{\circ}\). Therefore the particle velocities will reach about 100 m s\(^{-1}\) in the jet, indicating that the F&H test is an extreme test case for particulate processing. On increasing the interparticle collision velocity, there is a change in the attrition mechanism from chipping, involving only superficial damage, to fragmentation with integral damage (Ghadiri et al., 1994). Considering this dependence of the attrition mechanism on the particle velocity, it is unlikely that the F&H test represents the conditions causing attrition in the actual fluidised bed operation.

**CONCLUSIONS**

A novel experimental technique, based on digital analysis of high speed video images, has been successfully employed to verify hydrodynamic jet model simulations in two-
dimensions. With a similar model for three dimensional fluidised bed jets, predictions have been obtained for the F&H test conditions. From this, it appears that the F&H test is an uncompromisingly harsh test case for lean phase particulate processing, hardly representative of more common practical industrial conditions. The F&H test must therefore be applied with great caution for the assessment of the friability of particles in actual process conditions.

ACKNOWLEDGMENT

Financial support from the University of Surrey, EPSRC and Shell Research B.V. is gratefully acknowledged. The authors wish to express their thanks to Mr Peter Goodyer for providing quick access to the EPSRC's high-speed video camera and Shell Research B.V. for supplying the catalyst.

NOTATION

\[ u_{or} \quad \text{Orifice gas velocity} \quad (\text{m s}^{-1}) \]
\[ \varepsilon \quad \text{Voidage} \quad (-/-) \]
\[ \varepsilon_{mf} \quad \text{Voidage at minimum fluidisation level} \quad (-/-) \]
\[ \theta \quad \text{Jet half angle} \quad (\circ) \]

LITERATURE CITED


Appendix C

Impact Attrition of Fluid Cracking Catalyst

Proc. 5th Int. Conf. on Multiphase Flow in Industrial Plants, pp. 170-179, 1996
Abstract

Fluid catalytic cracking of heavy oil is an important multiphase process in the oil refining industry. One of the concerns in such a process is particle attrition which leads to material loss. An experimental investigation has been carried out using a single particle impact attrition test to examine the attrition behaviour of FCC particles. The results show that the attrition rate increases linearly with the particle size and with the square of the impact velocity. The attrition propensity of the fresh catalyst studied is 4 to 5 times greater than that of the corresponding equilibrium catalyst. Attrition of fresh catalyst is shown to decrease slightly with an increase in the number of impacts. Scanning electron micrographs show that the original particles have a large number of surface protrusions and necks, whereas the debris produced are similar in size and shape to the burs on the surface, suggesting that attrition occurs by breakage of one or more of the burs from the rest of the particle.

1. INTRODUCTION

Fluid catalytic cracking (FCC) is used in the oil refining industry to convert heavy fractions of petroleum oil to lighter products such as transportation fuels. The catalyst used in the FCC process is produced in the form of fine powder of particle size usually below 180 μm by spray drying of a slurry. It comprises 5-40% zeolite dispersed in a matrix of synthetic silica-alumina, semi-synthetic clay-derived gel, or natural clay [1]. In their study of the microstructure of FCC catalyst, Bass and Ritter
[2] have described in detail the chemical composition and morphology of recently
developed catalysts, which are a combination of gel, clay and zeolite.

An FCC unit consists of a reactor and a regenerator which are both fluidised beds
operating around 500 °C and 700 °C, respectively. The catalyst deactivates rapidly
due to coke deposition. Consequently, a high recirculation rate between the reactor
and regenerator, around 50 tonnes per minute, is necessary to regenerate the catalyst
by burning the coke, which in turn provides the heat required for the endothermic
catalytic reactions. Because of the large scale of solids transport and fluidisation at
high velocities (up to about 50 m s⁻¹), attrition of particles can be a serious problem,
giving rise to the elutriation of fines from the unit. Consequently, this could result in
a substantial loss of fines, a few tonnes per day for a typical FCC unit, leading to
additional operating costs. Thus, the mitigation of attrition of FCC powder is of
great significance.

In a fluidised bed, attrition is mainly due to particle-wall and particle-particle
collisions. Zenz and Kelleher [3] reported that the distributor region of a fluidised
bed and the cyclones are the main sites where severe attrition occurs due to the
presence of high velocity gradients. For an FCC unit, substantial attrition may also
occur in the riser between the regenerator and the reactor, where the velocities of the
gas solids mixture can reach high values, and on baffles, which are designed to
enhance the distribution of the gas/solids mixture.

Previous work on attrition has been reviewed by Bemrose and Bridgwater [4]. The
approaches employed for the analysis are largely empirical and the current
understanding of the mechanism of attrition in fluidised beds is poor. In a recent
study, Ghadiri and Boerefijn [5] presented a compilation of the models available in
the literature for the description of the attrition in fluidised beds. Most of these
models correlate the attrition of certain materials to one or more operating
parameters, such as the gas velocity, and their applicability is therefore limited.
There is little reliable information on the dependence of attrition on the properties of
individual particles, such as particle size, shape, porosity, surface morphology,
internal structure, and mechanical properties, and on the operating parameters, such
as distributor orifice size, free area, and the gas superficial velocities.
In many investigations of the attrition propensity of FCC catalyst, the method of Forsythe and Hertwig [6] has been commonly adopted in its original or modified form [7, 8]. This method employs a submerged air jet at a very high orifice gas velocity of 300 m s\(^{-1}\) as the source of attrition. Contractor et al. [8] reported that the attrition behaviour of catalyst particles in a riser reactor was not comparable to the attrition behaviour in the Forsythe and Hertwig [6] test. Experimental studies and numerical modelling of fluidised jet hydrodynamics [9] indicate that the orifice gas velocity, used in this test, is beyond the range of practical conditions applied in an FCC unit. Moreover, the specific geometry of the prescribed fluidisation apparatus may cause the attrition mechanism to differ significantly from that arising in more realistic geometries [10]. A Roller classifier, utilising air impingement at a nozzle exit velocity of about 140 m s\(^{-1}\), was used by Bass and Ritter [2] to quantify the attrition propensity of catalysts with different microstructures. They observed a difference in attrition behaviour between fresh and used catalyst. The jet fluidisation test and the Roller test are both suitable methods for comparison of the attrition behaviour of different materials within the limits of the test conditions. These conditions are, however, different from those in FCC units, and therefore it is uncertain at present how the results of these tests can be correlated with the actual attrition behaviour in a plant.

As an alternative, the method of single particle impact has been developed to investigate the mechanism of impact attrition [11, 12]. In this method, the particles are all impacted on a target at a specified velocity, and therefore the test conditions are well defined. This test has proved to be a suitable method to evaluate the role of material properties. In the present study, the single particle impact test is employed to investigate the mechanism of breakdown of FCC particles. The dependence of the attrition propensity on impact velocity and particle size is evaluated. The attrition of fresh and equilibrium catalysts is compared. In the following, the experimental method and results are presented.
2. EXPERIMENTAL

2.1 Materials

Samples of fresh (F-cat) and equilibrium (E-cat) catalyst from the same origin were supplied by Shell International Oil Products B.V. The particle size distribution of the powder was measured using laser diffraction (Malvern Mastersizer). The volumetric mean size of F-cat and E-cat was 55 and 65 μm, respectively. Tests have been carried out with samples of particles from several single sieve cuts according to BS410.

Figure 1 shows scanning electron micrographs of some samples of the tested materials. Table 1 lists the size ranges tested for both types of material. Figs 1a and 1b show F-cat of 63-75 μm and 106-125 μm, respectively. The structure of the material is complex, akin to a ginger root, but more spherical than this. Comparing Figs 1a and 1b, the structure of the sample with the larger size is more complex, with a large number of burs on the surface of the particles. This type of structure has also been reported in previous studies.
E-cat, shown in Fig. 1c, has a far smoother surface. Some burs are still visible, but generally the particles are more spherical.

2.2 Experimental Apparatus

The experimental set-up is shown schematically in Fig. 2. Only a brief description is given here, more details can be found elsewhere [13, 14]. The test rig consists of a funnel-shaped inlet section guiding the particles into an eductor tube which ends in a collection chamber. A target plate of sapphire is positioned below the eductor tube. Particles fed from a vibrating tray are sucked into the funnel and then accelerated downward in the eductor tube by the entraining air flow. After exiting from the tube, the particles impact on the target. The impact products are retained in the collection chamber on a paper filter which is supported by a brass porous plate. The entraining air leaves the collection chamber through the porous plate into a vacuum system.

The pressure in the collection chamber is monitored by a micromanometer and kept at a slight vacuum to prevent back flow of air upward through the funnel at the top of the eductor tube. The particle velocity at the target is measured by means of laser Doppler velocimetry (Thermo Systems Inc.). For the small particles concerned, the particle velocity is solely determined by the gas velocity, and the effect of particle size and density on the particle velocity was found to be negligible in the present set-up.

2.3 Experimental Procedure

An impact test involves the following steps. A sample of material of a single sieve cut size is prepared by manual sieving. The sample, typically 2 to 3 g, is then fed into the test rig and collected from the chamber after the impact. The impact products are then sieved. For the purpose of gravimetric analysis, the mass of the particle sample at various stages of the test is recorded: the feed particles ($M_f$), which are weighed immediately before a test; the collected particles after the impact ($M_c$), the mother particles retained on the sieve ($M_m$), and the debris passing through the sieve ($M_d$). As a criterion for the size of the debris, a sieve with a size which is two BS410 sieve sizes below the lower limit of the particle size cut was chosen. Previous studies have shown that this criterion is insensitive to the breakage product
size distribution, if the breakage product is much smaller than the original particles [14].

Table 1 also lists the sieve size for the debris of the samples tested.

FCC powder, particularly fresh FCC, strongly adsorbs moisture due to a high porosity and a large surface area. In order to minimise the errors of the mass measurements caused by moisture adsorption or desorption, the samples were exposed to ambient air for a long time to ensure that equilibrium with the atmosphere had been reached prior to tests.

Great care was taken in handling the samples of fine particles to minimise handling losses. However, for fine particles, handling losses are inevitable at almost every stage of the test, and are variable due to e.g. electrostatic charge, ad- or desorption of moisture, and/or incomplete collection of particles. The overall handling loss is defined as:

$$H_L = (1 - \frac{M_m + M_d}{M_f}) \times 100\%$$  \hspace{1cm} (1)

Taking account of the handling loss, the attrition may be calculated in two ways. If the handling loss is considered to be debris, the attrition is overestimated. Therefore, by attributing the handling loss to the debris, an upper limit of the attrition, denoted by $\xi^+$, is defined:

Table 1. Tested materials and size ranges.

<table>
<thead>
<tr>
<th>Material</th>
<th>Size range (µm)</th>
<th>Debris separation sieve size (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>F-cat</td>
<td>63-75</td>
<td>45</td>
</tr>
<tr>
<td>F-cat</td>
<td>75-90</td>
<td>53</td>
</tr>
<tr>
<td>F-cat</td>
<td>90-106</td>
<td>63</td>
</tr>
<tr>
<td>F-cat</td>
<td>106-125</td>
<td>75</td>
</tr>
<tr>
<td>E-cat</td>
<td>75-90</td>
<td>53</td>
</tr>
<tr>
<td>E-cat</td>
<td>106-125</td>
<td>75</td>
</tr>
</tbody>
</table>
\[ \xi^+ = \left(1 - \frac{M_m}{M_f}\right) \times 100\% \]  

On the other hand, if the handling loss is attributed to the mother particles, the attrition would be underestimated, and based on this a lower limit, denoted by \( \xi^- \), may be defined:

\[ \xi^- = \frac{M_d}{M_m + M_d} \times 100\% \]  

It may be expected that the actual attrition rate is between \( \xi^+ \) and \( \xi^- \). However, duplicated tests showed that the lower limit of the attrition rate measured for the present systems was consistently more reproducible than the upper limit. The poor reproducibility of \( \xi^+ \) was mainly a result of varied magnitudes of the handling loss. It can be shown that \( \xi^+ \equiv \xi^- + H_L \) for \( M_d \ll M_m \), which was always the case. As the value of \( \xi^- \) was found to be insensitive to handling losses, only the lower limit will be presented in the following as an indication of the level of particle attrition.

The particles were fed in a single array from the vibrating tray at a low feeding rate. This resulted in a highly diluted entrained flow within which collisions between particles before or at the target will be negligible. Thus, single-particle impact conditions were maintained.

Scanning Electron Microscopy (SEM) was used as a visualisation method for qualitative characterisation of the original particles and the impact products.

### Table 2. Coefficient C and power index n of trendlines of attrition data vs impact velocity, shown in Figs 3 and 4.

<table>
<thead>
<tr>
<th>Material</th>
<th>Size range (μm)</th>
<th>C</th>
<th>n</th>
</tr>
</thead>
<tbody>
<tr>
<td>F-cat</td>
<td>75-90</td>
<td>5 \times 10^{-4}</td>
<td>1.93</td>
</tr>
<tr>
<td>F-cat</td>
<td>90-106</td>
<td>2.4 \times 10^{-3}</td>
<td>1.79</td>
</tr>
<tr>
<td>F-cat</td>
<td>106-125</td>
<td>1.3 \times 10^{-3}</td>
<td>2.06</td>
</tr>
<tr>
<td>E-cat</td>
<td>75-90</td>
<td>7.0 \times 10^{-5}</td>
<td>2.03</td>
</tr>
<tr>
<td>E-cat</td>
<td>106-125</td>
<td>8.0 \times 10^{-4}</td>
<td>1.79</td>
</tr>
</tbody>
</table>

3. **RESULTS**

3.1 **Effect of impact velocity**

Figures 3 and 4 show the attrition as the fractional mass loss upon impact, defined in Eq. 3, for F-cat and E-cat respectively. It is shown that the
fractional mass loss increases with impact velocity for both types of material for different size cuts. The dependence of the attrition on the impact velocity can be described by a power law:

\[ \xi = C v^n \] (4)

where values of constant \( C \) and power index \( n \) have been obtained by regression analysis, and are shown in Table 2. The best fit curves are shown in Figs 3 and 4.

### 3.2 Effect of particle size

Figures 3 and 4 also show that for a given particle velocity, the fractional loss increases with particle size for the size range between 63 and 125 \( \mu \text{m} \). The effect of particle size for F-cat may be better illustrated by replotting the attrition data against the arithmetic mean particle size. This is shown in Figure 5. It appears that a linear relationship exists between the fractional loss and the particle size. The slope of the straight lines increases with impact velocity.

Figure 3. Attrition of F-cat as a function of impact velocity. \( \triangle \) 63-75 \( \mu \text{m} \); \( \bigcirc \) 75-90 \( \mu \text{m} \); \( \square \) 90-106 \( \mu \text{m} \); \( \blacksquare \) 106-125 \( \mu \text{m} \).

Figure 4. Attrition of E-cat as a function of impact velocity. \( \bigcirc \) 75-90 \( \mu \text{m} \); \( \blacksquare \) 106-125 \( \mu \text{m} \).

Figure 5. Attrition of F-cat as a function of particle size. \( \triangle \) 14.7 m s\(^{-1}\); \( \bigcirc \) 23.4 m s\(^{-1}\); \( \square \) 33.1 m s\(^{-1}\); \( \blacksquare \) 40.5 m s\(^{-1}\).
velocity.

3.3 Fresh and equilibrium catalyst

Comparison of Figs 3 and 4 shows that the attrition propensity of particles of the fresh catalyst is substantially larger than that of the equilibrium catalyst. The attrition of F-cat is 4 to 5 times greater than that of E-cat for both size cuts of 75-90 μm and 106-125 μm.

3.4 Effect of the number of impacts

A catalyst particle experiences numerous collisions during its service life in the FCC unit. Attrition may occur at each collision, and the instantaneous attrition rate may depend on the impact history. A limited number of repeated impact tests has been undertaken in the present study. For F-cat of particle size 106-125 μm, the attrition appears to decrease slightly with an increase in the number of impacts, as shown in Fig. 6 for a particle velocity of 15 m s\(^{-1}\).

4. DISCUSSION

4.1 Morphology

Figure 7 shows a scanning electron micrograph of the debris of 106-125 μm E-cat. The debris appears to consist of burs, micro-spheroids detached from the parent agglomerate particle. The burs appear to have broken at solid bridges, or necks, formed between the microspheroids during spray drying.

For silica-alumina catalyst particles, tested in a modified Forsythe and Hertwig [6] device, Gwyn [7] reported that complete or partial wearing of protruding particles,
i.e. burs, from the surface of the spray-dried agglomerates is the main cause of attrition. Bass and Ritter [2] compared the behaviour of a typical, relatively soft catalyst, and a special, more resistant catalyst. They report that, compared to the latter, the former sample lost twice as much fines during attrition testing in the Roller device. The typical catalyst appeared to undergo attrition by breakage of surface protrusions, whereas the more resistant catalyst showed fractured parts of microspheroids.

Comparison of Figs 1b and 1c shows that E-cat has a very smooth surface when compared to F-cat. Therefore a relatively slow erosion mechanism must be operating on the FCC catalyst. Also, as indicated by the arrow in Figure 1c, the breakage of protruding particles from the surface of the agglomerate has left depressions in the surface. Bass and Ritter [2] show similar depressions in the surface of used catalyst. Necking of protrusions therefore seems to be the common cause for breakage in both the impact test and the actual cracking unit operation. Furthermore, the extent of the breakage appears to be determined by the irregularity of the agglomerate shape, i.e. the number and size of protrusions present. Further work is required to develop a model of attrition based on the necking mechanism.

Ghadiri and Zhang [15] proposed a model to describe the attrition by chipping of particles in the semi-brittle failure mode. The model predicts that the attrition rate increases linearly with the size of the particles and to a power of 2 with the impact velocity. These predictions are numerically in agreement with the present results, as shown in Figs 3, 4 and 5, with the power indices of the attrition rate with respect to
impact velocity in all cases being close to 2, as shown in Table 2. Despite possible differences in the breakage mechanisms, the agreement may be explained by recognising that the breakage is proportional to the incident energy of the particle upon impact, and it should therefore scale with the square of the impact velocity.

4.2 Effect of particle size

The increase in attrition rate with particle size may be attributed to i) a higher impact energy (proportional to \(d_p^3\)) for a given velocity, and ii) a more complex particle shape, \(i.e.\) more and possibly larger burs agglomerated on a larger particle. The latter may be seen by comparing Figs 1a and 1b, \(i.e.\) smaller particles are more regularly shaped than larger particles. If the best fit lines in Fig. 5 are extrapolated to intersect with the horizontal axis, then an intercept particle size, \(d_i\), is obtained for each velocity. As shown in Table 3, \(d_i\) is relatively constant over a large range of impact velocities. This indicates that there is a limiting particle size, below which the particles do not break according to the proposed mechanism, and suggests that the breakage may be due to a different mechanism from necking, perhaps leading to more superficial breakage than would be involved in necking of protrusions.

4.3 Behaviour of fresh and equilibrium catalyst

The difference in attrition behaviour between F-cat and E-cat particles, as shown in Figs 3 and 4, may be explained by the following considerations. A catalyst taken from an FCC unit is assumed to have reached the steady state attrition rate after a long time of service during which the particles have experienced a large number of impacts and abrasive interactions. It has also been treated thermally and chemically with severe conditions in the reactor and regenerator for numerous times, and is therefore referred to as equilibrium catalyst. As a result, E-cat particles have a smoother surface and a more spherical shape with fewer protrusions than F-cat particles, as shown in Figs 1b and 1c. This is illustrated by the apparent decrease, albeit

<table>
<thead>
<tr>
<th>Impact velocity (m s(^{-1}))</th>
<th>(d_i) ((\mu)m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>14.7</td>
<td>73.8</td>
</tr>
<tr>
<td>23.4</td>
<td>73.5</td>
</tr>
<tr>
<td>33.1</td>
<td>72.5</td>
</tr>
<tr>
<td>40.5</td>
<td>70.0</td>
</tr>
</tbody>
</table>
slight, in attrition of F-cat with the number of impacts, as shown in Fig. 6.

Bass and Ritter [2] observed a densified skin of used catalyst particles, which is less than 0.2 μm thick. This was attributed to sintering and coke deposition. However, the interior of the used particles was shown to remain open-structured. Similar features have also been observed in the present study for E-cat particles, as shown in Fig. 8. As for F-cat particles, no significant difference was observed between the morphologies of the interior and the surface. Contractor et al. [8] systematically modified the surface composition of the catalyst, by adding sub-colloidal particles to the slurry, which migrated to the agglomerate surface during spray drying, resulting in a tougher catalyst. Therefore, it appears that the surface texture and the regularity of the agglomerate shape are the important parameters determining the attrition propensity of the catalyst. Both parameters can be controlled during the spray drying process. Great attention should therefore be paid to this stage of the catalyst preparation.

5. CONCLUSIONS

An experimental investigation has been carried out using a single particle impact rig to examine the attrition behaviour of FCC catalyst particles. It has been observed that the attrition rate increases with impact velocity for both fresh and equilibrium catalysts of different particle sizes. The velocity dependence follows a second order power law. For a given particle velocity, the attrition rate increases linearly with particle size. The attrition propensity of the fresh catalyst is 4 to 5 times greater than that of the equilibrium catalyst. A limited number of repeated impact tests has shown that the attrition decreases gradually with an increase in the number of impacts.

Based on SEM observations of the original particles and debris, a necking mechanism of FCC catalyst attrition has been proposed. The original particles look like ginger roots with a large number of surface protrusions and necks, whereas the debris produced are similar in size and shape to the burs, suggesting that necking separates one or more of the burs from the rest of the particle. The lower attrition rate of the equilibrium catalyst may be attributed to a smoother surface, a more
spherical shape with fewer protrusions, and a different surface morphology when compared to the fresh catalyst. These results imply that the method and conditions applied for production of the powder have significant effects on the attrition propensity of the product through particle shape and structure.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge financial support from the EPSRC under the ROPA Programme and Shell International Oil Products B.V., Amsterdam.

NOMENCLATURE

\[ C \quad \text{Constant} \quad \text{(see Eq. 4)} \]

\[ d_i \quad \text{Intercept particle size} \quad \text{(m)} \]

\[ H_L \quad \text{Handling Loss} \quad \text{(kg)} \]

\[ M_c \quad \text{Mass of collected particles} \quad \text{(kg)} \]

\[ M_d \quad \text{Mass of debris} \quad \text{(kg)} \]

\[ M_f \quad \text{Mass of particles to be impacted} \quad \text{(kg)} \]

\[ n \quad \text{Power index} \quad \text{(see Eq. 4)} \]

\[ \xi \quad \text{Fractional mass loss} \quad (-/-) \]

\[ \xi^- \quad \text{Lower limit of fractional mass loss} \quad (-/-) \]

\[ \xi^+ \quad \text{Upper limit of fractional mass loss} \quad (-/-) \]

\[ v \quad \text{Impact velocity} \quad \text{(m s\(^{-1}\))} \]

REFERENCES


Appendix D

Distinct Element Simulation of Impact Breakage of Lactose Agglomerates
Distinct Element Simulation of Impact Breakage of Lactose Agglomerates

Z. Ning, R. Boerefijn, M. Ghadiri, and C. Thornton

Abstract - Traditional theoretical and experimental investigations of the mechanical behaviour of particulate solids are restricted by the limited quantitative information about what actually happens inside particulate assemblies. This paper presents computer simulation results of the breakage of lactose agglomerates due to impact on a target plate using the distinct element analysis. The agglomerates of interest here are generally weak and easy to disintegrate as no binder other than weak surface forces are holding the primary particles together. Particle interaction laws in the simulation code are based on theoretical contact mechanics, where adhesive interface energy determines the bond strength between individual particles of the assembly. Experimental investigations have been conducted to validate the computer simulation results, using a simple air-eductor where particles are accelerated to the required velocity by an air flow and impacted against a rigid target plate. Computer graphics of the simulation results of agglomerate breakdown are compared with the images obtained by high speed video recording of the impact events. A good agreement has been found between the simulation results and experimental measurements. Dynamic features and loading compliance of weak agglomerates are found to be distinctly different from those of high strength agglomerates and solid particles.

NOMENCLATURE

\( A \) Local area [-]
\( E \) Young's modulus of the primary lactose particles [N/m²]
\( F \) Contact force [N]
\( K_c \) Fracture toughness [N/m\(^{3/2}\)]
\( t \) Time [s]
\( \Delta t \) Time step [s]
\( V \) Impact velocity [m/s]
We

Mass of mother particles and debris collected after impact, before separation by sieving [kg]

$W_d$ Mass of debris after collection and separation by sieving [kg]

$W_f$ Mass of particles fed into the eductor [kg]

$W_m$ Mass of mother particles after collection and separation by sieving [kg]

$\Gamma$ Interface adhesion energy [N/m]

$\mu$ Friction coefficient [-]

$\nu$ Poisson ratio of the material [-]

$\xi$ Attrition rate: percentage weight loss on impact [%]

$\xi^+$ Upper limit of attrition rate [%]

$\xi^-$ Lower limit of attrition rate [%]

$\xi^*$ Attrition data from computer simulation [%]

1. INTRODUCTION

Understanding and control of agglomerate strength is of interest to many processes involving powders, such as handling, mixing, pneumatic conveying, compaction, etc. In processing these materials, there are circumstances under which inter-agglomerate and agglomerate-wall collisions cause their breakdown. In some cases, such as dispersion and comminution, this is desirable, while in other cases such as attrition it is undesirable. Control of agglomerate strength is therefore crucial. On one hand, the agglomerates should be sufficiently strong to resist attrition in storage and transport, but on the other hand they should easily breakdown and disperse in the processing stage. There is therefore a narrow window of operation for which the agglomerate strength needs to be carefully tailored.

It is well recognised that the failure mode of particulate solids upon impact can be broadly classified as brittle, semi-brittle and ductile, depending on the deformation form [1,2, 3]. Brittle failure is caused by fracture with little or no plastic deformation. Impact under this failure mode usually produces diametrical cracks, splitting the particle into fragments, as the diametrical plane and equatorial circumference are under the greatest tensile stresses [1]. If plastic flow precedes
fracture, the process is termed semi-brittle. Here, the plastic zone produces compressive radial stresses and tensile hoop stresses. The latter type of stress propagates radial and median cracks, initiated from the plastic zone. When the load is removed, the residual tensile stresses, formed after the elastic unloading process, generate sub-surface lateral cracks. The characteristics of the semi-brittle failure mode are particle fragmentation due to the formation of median and radial cracks, and chipping due to the formation of lateral cracks [2, 3]. Ductile failure is dominated by extensive plastic flow which is responsible for the rupture of the material. Ploughing and cutting are the two main mechanisms of material removal for this failure mode [4].

For agglomerate breakage, however, the understanding of the failure mechanisms is very much limited as identified by previous experimental [5-8] and numerical [9-12] investigations. Particulate solids are complex and redundant systems. The difficulty in quantifying the failure behaviour of the agglomerates lies in the fact that the agglomerate strength is related to the microstructure and the bond strength between primary particles, which cannot be easily measured with the current experimental techniques. Most of the reported studies are experimental and the characterisation of the failure mode has followed the standard solid mechanics techniques using the concepts of brittle, semi-brittle, or ductile failure. For example, linear elastic fracture mechanics has been applied to investigate agglomerate strength and breakage [7, 8, 13]. It is assumed in this approach that particulate solids fail by the propagation of cracks and the conditions for failure are related to the energy requirement for crack propagation. Limitations for this solid mechanics analysis approach are the difficulties in experimentally measuring the crack length and accounting for energy dissipation processes. Furthermore, there is no physically reliable correlation between single bond rupture on the microscopic scale and macroscopic failure of the bulk material.

The distinct element analysis (DEA), first developed by Cundall [14,15], presents an alternative way to obtain insight into the particle systems and provides fundamental information such as microscopic structure, interparticle forces, particle
velocities, etc. Most importantly, this method makes it possible to relate the bulk mechanical behaviour of the assembly to individual particle properties. A detailed examination of the micromechanics, which determine the bond breaking and the internal microstructural deformation, can therefore be provided. For simulations in two dimensions (2-D), impact fracture of agglomerates was reported by Thornton et al. [9]. Kafui and Thornton [10, 11] recently presented a series of three dimensional (3-D) simulations in which a monodispersed spherical agglomerate consisting of 8000 primary particles in face-centred cubic arrangement was impacted against a wall. Simulated impact fracture of solid particles has been reported by Potapov and Campbell [12], although the detailed methodology is significantly different from the one used in this paper. In their simulations of impact fracture and fragmentation, rigid elements are assembled into a simulated solid particle by "glueing" the elements together with compliant boundaries. Fracture occurs when the tensile strength of the glued joints is exceeded, permitting cracks to propagate through the solid body. The fragmentation characteristics observed by experiments of Arbiter et al. [6] have also been demonstrated by computer simulations, with respect to the fragment size distribution [10, 11] and the fan shape fracture [9, 12].

Most previous studies in quantifying agglomerate failure appear to be related to high strength agglomerates which produce identifiable crack planes on failure. Weak agglomerates on the other hand do not produce well-defined fracture patterns, and have not been investigated extensively. However, their behaviour is of interest to many pharmaceutical and process industries. This paper presents a distinct element analysis of impact of lactose agglomerates on a rigid target surface and addresses the effect of impact velocity on the amount of breakage produced. The agglomerates of interest here are weak and easy to disintegrate as no binder other than weak surface forces are holding the primary particles together. For a study on agglomerate impacts, a very important aspect that needs to be clarified is how the breakage of the particulate assembly differs from that of solid particles. The failure mode, loading compliance, and dynamic features such as the evolution of the contact force and kinetic energy of the system, are examined and compared with the reported data of other types of agglomerates and solid particles in the literature.
2. DISTINCT ELEMENT ANALYSIS

Computer simulation of assemblies of contiguous solid particles was pioneered by Cundall [14, 15], who developed the distinct element analysis in which the interaction of particles is considered as a transient problem with states of equilibrium developing whenever the internal forces balance. In order to implement realistic laws of contact interactions into the code, extensive modifications have since been made by Thornton and co-workers [16, 17, 18]. For the case that adhesion is absent, the normal and tangential contact forces are based on Hertzian theory and the work of Mindlin and Deresiewicz [19]. In the presence of adhesion, the JKR model (Johnson et al. [20]) is used for the normal contact stiffness while the tangential behaviour is governed according to Savkoor and Briggs [21] and Thornton [16]. For single particle impacts, the simulation results have been confirmed by matching analytical solutions when available, as reported by Thornton and Yin [17] and Ning [18]. The computer program used for agglomerate impacts presented in this paper is based on the version of TRUBAL developed by Thornton et al. [9], in which planar walls and surface energy are included in order to perform computer simulations of agglomerate-wall impacts.

For the details of the computer code structure and implementations of theoretical contact interaction laws, reference is made to Cundall and Strack [22] and Ning [18]. A brief description of the methodology used in the distinct element analysis is presented in the following. Newton’s second law is applied in TRUBAL to govern the motion and displacement of particles in the simulated assembly, where the related parameters, such as displacements (normal and tangential), velocities (linear and rotational), and contact forces (normal and tangential), are all time dependent. The evolution of a dynamic process consists of a series of calculation cycles in which the state of the particle system is advanced over a small increment of time $\Delta t$.

At any time $t$, interparticle force increments are calculated at all contacts from the relative velocities of the contacting particles using an incremental force-displacement law. The interparticle forces are updated and, from the new out-of-balance force and moment of each particle, new particle accelerations (both linear and rotational) are
obtained using Newton’s second law. Numerical integration of the accelerations is then performed over the time step $\Delta t$ to give new velocities. Further numerical integration provides displacement increments from which the new particle coordinates are obtained. Having obtained new positions and velocities for all the particles, the program repeats the cycle of updating contact forces and particle locations. Checks are incorporated to identify new contacts and contacts that no longer exist. In each cycle, every particle in the assembly is treated in the manner described above and the calculation cycle is repeated until the end of simulation.

3. SAMPLE PREPARATION

An agglomerate sample consisting of 2000 primary particles was prepared for the computer simulation of the impact tests. The damage to individual primary particles is not considered in this study. The simulations are carried out for an agglomerate of lactose, whose constituent particle properties were assumed to be the same as those of large lactose crystals. For the latter, the mechanical properties have been characterised by the use of nano-indentation techniques [23] and are summarised in Table 1. The simulated assembly is a polydispersed system containing three different particle sizes. The particle radii and corresponding mass percentage of the assembly occupied are 9 $\mu$m, 25%; 10 $\mu$m, 50%; and 11 $\mu$m, 25%, respectively.

One of the most important parameters chosen for agglomerate collisions is the surface energy. Roberts [24] summarised reported values of the interface energy of lactose crystals, which varied widely from $\Gamma = 0.2$ to 20 J m$^{-2}$. This variation is considered to be mainly associated with the differences in material properties such as Young’s modulus and porosity, as well as the test methods used. To determine an appropriate value of the interface energy for the computer simulations we used indentation fracture mechanics to obtain the fracture surface energy from the fracture toughness $K_c$. It is implicit in this approach that the interface energy of the lactose primary particles is approximated by the fracture surface energy of lactose crystals. According to the theory of linear elastic fracture mechanics [25] for the case of plane strain, $\Gamma$ and $K_c$ are related by
\[ \Gamma = \frac{K_c^2(1-\nu^2)}{E} \]  

where \( E \) and \( \nu \) are Young's modulus and Poisson ratio of lactose, respectively. The measured value of \( K_c \) is in the range of 35 - 88 kPa m\(^{1/2}\) for solid lactose crystals \cite{23}, from which the interface energy can be estimated to be in the range of \( \Gamma = 0.348 - 2.2 \ J \ m^{-2} \) from Equation (1). If we take the interface energy \( \Gamma = 0.5 \ J \ m^{-2} \), which was used in the simulations, this corresponds to \( K_c = 41.9 \ kPa \ m^{1/2} \), which is a modest value within the range of the measured values of fracture toughness.

### Table 1

Particle and target properties used for simulation

<table>
<thead>
<tr>
<th></th>
<th>Lactose crystals</th>
<th>Stainless steel</th>
</tr>
</thead>
<tbody>
<tr>
<td>Young's modulus (GPa)</td>
<td>3.2</td>
<td>215</td>
</tr>
<tr>
<td>Poisson ratio</td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td>Density (kg m(^{-3}))</td>
<td>1550</td>
<td>7800</td>
</tr>
<tr>
<td>Yield stress (GPa)</td>
<td>0.21</td>
<td>3.04</td>
</tr>
<tr>
<td>Coefficient of friction</td>
<td>0.35</td>
<td>0.35</td>
</tr>
<tr>
<td>Fracture toughness (kPa m(^{1/2}))</td>
<td>41.9</td>
<td>not measured</td>
</tr>
<tr>
<td>Interface energy (J m(^{-2}))</td>
<td>0.5</td>
<td>0.2</td>
</tr>
</tbody>
</table>

The procedures of sample preparation are briefly described in the following. Within a given spherical region, primary particles are randomly generated. A centripetal gravity field is then introduced to bring the particles together. With the increase of the number of contacts, the density of the agglomerate system or solid fraction gradually increases. When the co-ordination number, defined as the average contact number for each individual particle, or solid fraction of the assembly, reaches the desired level, surface adhesion is introduced in very small increments. After the agglomerate is prepared, the centripetal gravity needs to be transformed into a one dimensional gravity field. In the impact direction, the gravity value should be -9.81 m s\(^{-2}\), while the other two components are set to zero. For an agglomerate-wall collision, the wall is first created and then moved to a new location within a very
small distance from the agglomerate. The gap between the wall and the agglomerate is so small that the effect of velocity increase due to gravity can be ignored. Before the impact velocity is specified, sufficient computational cycles must be carried out so that the kinetic energy of the primary particles is gradually dissipated by introducing global damping, allowing the agglomerate to reach a static equilibrium state.

Before the impact velocity is specified, the established contact number within the sample is 5439, equivalent to a co-ordination number of 5.4 for the system. The solid fraction of the agglomerate is 0.522 and the agglomerate diameter is about 300 μm.

4. SIMULATION RESULTS

4.1. Visual observation

A series of 3-D simulations of agglomerate-wall collisions has been conducted at modest low impact velocities in the range 1.0 - 4.5 m s⁻¹. As for high impact velocity impacts, a velocity of 10.0 m s⁻¹ was also tested. For each specified impact velocity, the number of broken contacts and the extent of agglomerate breakage may be examined during the impact process. Using computer graphics, the breakage process of the agglomerate can be observed by designating different levels of grey scale colouring to the primary particles according to the size of the cluster to which they belong. Visual observations of simulation results are shown in Figures 1(a-h) for eight impact velocities. In these figures, the dark grey particles are the particles which form the residual cluster; white particles are singlets; light grey particles are the clusters which contain 2-10 primary particles; and black particles are the clusters which contain 11-30 particles. Even though this is only one view of a three-dimensional agglomerate, it is clear that the number of particles separated from the parent agglomerate increases with an increase of impact velocity. Significant deformation of the agglomerate is visible in the impact region, without the development of a clear crack plane. Therefore, the failure of the agglomerate during
the impact process appears to be macroscopically "ductile", although microscopically the interparticle bond failure is by elastic rupture, i.e. brittle.

For an impact velocity of 4.0 m s\(^{-1}\), Figure 2 shows the evolution of the residual cluster and contact breakage diagram at different impact stages. The individual particles or small clusters are separated from the parent agglomerate, while the residual cluster still remains large after impact at this velocity (Figures 2(a-I)-(d-I)). At the end of the impact, most detached particles and small clusters are found in the impact area, as it can also been seen in Figure 1(g). The number and location of interparticle contacts broken within the assembly are illustrated in Figures 2(a-II)-(d-II) by the equivalent space lattice. To form this space lattice, a short straight line is drawn connecting the centres of the two particles between which contact has been broken. Contact breakage initiates from the impact area and then spreads along the longitudinal direction to the top surface of the assembly. This implies that spreading of the contact breakage might be associated with the force transmission through the packed assembly. However, the breakage pattern is different from "strong" agglomerates where clearly identifiable crack planes can be observed [9]. Here it appears that more contacts are broken in the latitudinal direction, as overlapping of the contact breakage lines forms thick barbs in this direction. Since Figures 2(a-II)-(d-II) are three dimensional diagrams, it is difficult to identify the local structural change of the particle system. A suggestion of the contact breakage pattern is illustrated by the simplified 2-D diagrams, shown in Figures 3(a) and (b), respectively. When the agglomerate is flattened by the impact force, contact breakage due to particle sliding occurs in a way illustrated in Figure 3(b). For this type of local structural arrangements, the top and bottom particles will force the two middle particles to separate from each other. When more and more contacts are deleted, this mechanism will ultimately lead to the latitudinally distributed field of the broken contacts (Figure 3(a)). This distribution is also visible in Figures 2(c-II) and (d-II). The results here imply that the agglomerate cannot withstand much elastic strain and the interparticle contacts can fail easily by sliding or rupture.

Agglomerate damage at a high impact velocity of 10.0 m s\(^{-1}\) is shown in Figures 4(a-c). Two stages of the impact process are illustrated in Figures 4(a) and (b) at
impact times of 10.6 \( \mu s \) and 17.7 \( \mu s \), respectively. Figure 4(c) shows the clusters resulting from the impact with singlets removed from the figure. For this high impact velocity the residual cluster no longer exists. It should be noted that, for the impact velocity of 10 m s\(^{-1}\), the total impact duration is much longer than that indicated in Figure 4, which only demonstrates the state at two different stages of the impact. For impact fracture of solid particles, experimental investigations [27] indicate that there is a transition velocity above which particles are fragmented and below which chipping commonly occurs. However, it appears that this trend does not prevail in the impact of weak agglomerates. In general, weak agglomerates considered here cannot store significant elastic strain energy before breaking as their compliance is very high. This may be an important factor in affecting crack formation and propagation in the system, and consequently differentiating the behaviour of weak agglomerates from other types of agglomerates or solid particles. Further work is needed to elucidate this difference.

4.2. Dynamic features of the agglomerate system

Using distinct element simulations, the contact number, kinetic energy of the system, and interparticle forces can be monitored at every stage of impact. To quantify the change in the microscopic structure of particulate material resulting from agglomerate collisions, a contact damage ratio is defined as the ratio of the number of contacts broken to the total number of contacts existing prior to impact [9]. For impact velocities of 3.0 and 4.0 m s\(^{-1}\), Figures 5 and 6 show the evolution of the number of contacts with the wall and the total force on the target wall, respectively. The number of wall contacts generally increases with an increase of impact time but with small fluctuations during the impact. However, large fluctuations of the total wall force are observed for both impact velocities. Comparing these two figures, it is found that, for example at \( V = 4.0 \) m s\(^{-1}\), the peaks and troughs in the fluctuations of the wall contact number and the wall force are generally correlated with each other. The singlets or small clusters separated from the original agglomerate will not carry large forces to the wall and therefore the residual cluster plays a major role in transferring forces to the surface. Corresponding to the impact, the maximum wall
forces exerted during impacts at velocities of 3.0 and 4.0 m s\(^{-1}\) are 1.59 and 3.47 mN, respectively. The static force due to the weight of the agglomerate itself is about 0.129 µN, which is even far less than the minimum force during the simulation. The kinetic energy at the end of the simulation is still high which implies that the fluctuations could continue for some time for the agglomerate to reach a new static equilibrium state.

For solid particle impacts, the impact force will have a symmetric force-time pulse curve if the impact is purely elastic. When plastic yield is exceeded, the curve is asymmetric due to plastic loading and elastic unloading behaviour. When the particle bounces off the target surface, the contact force drops to zero. However, it is seen from Figure 6 that neither case occurs to the impact force on the wall. The fall of the wall force to a zero value for the weak agglomerate is not observed for the impact period recorded here, i.e. similar to the trend of the kinetic energy. The same behaviour is observed for other impact velocities, suggesting that for the agglomerate to reach a static equilibrium state the process is very slow.

The relationship between the wall force and the extent of agglomerate flattening is shown in Figure 7 for the three impact velocities of \(V = 2.0\), 3.0 and 4.0 m s\(^{-1}\). The extent of flattening is quantified as the reduction in the size of the agglomerate in the impact direction. The data presented in the figure correspond to the very early stage of the impact at about 1 µs, i.e. the time from the first contact to the moment that the first local maximum wall force is attained. Referring to Figure 6, for \(V = 3.0\) m s\(^{-1}\), the local maximum force on the wall occurs at impact time \(t \approx 1.0\) µs. The force-deformation relationship of an impacting agglomerate, as shown in Figure 7, indicates a distinct difference in the mechanical behaviour between weak agglomerates and single solid particles. The initial loading curve here depends on impact velocity, i.e. the compliance of normal loading is sensitive to the rate of loading, a behaviour which is not observed for solid particle impacts in the elastic as well as elastic-perfectly plastic deformation regimes. As generally recognised for solid particles, the loading curve at early stages remains the same for all impact velocities [18]. The macroscopic behaviour of weak agglomerates therefore appears
to be more akin to solid particle impacts under plastic deformation accompanied by strain rate effects.

Evolution of the kinetic energy of the particle system and of the damage sustained by the agglomerate are shown in Figure 8 and 9 respectively, for three impact velocities of 2.0, 3.0 and 4.0 m s\(^{-1}\). In Figure 8, the kinetic energy of the system is normalised by dividing it by the initial kinetic energy before impact. The kinetic energy decreases monotonically with time. The trend here is different from solid particle impacts, where the initial kinetic energy is first transformed into elastic or elastoplastic strain energy and reduced to zero when the impact force approaches the maximum value. The stored elastic strain energy then furnishes the kinetic energy of rebound in the recovery stage. However, for the case of weak agglomerates simulated here, the centre of mass of the agglomerate continues moving downwards and agglomerate rebound never occurs.

It can be seen from Figure 9 that the contact damage ratio over the impact duration can be divided into two regions, the fast contact breakage period and slow contact breakage period. The transition point corresponds approximately to the moment at which the impact wall force approaches its maximum value. In computer simulated impacts, a question that has to be addressed is when the simulation should be stopped, because the agglomerate does not bounce off after hitting the target. Consequently, interactions between the particles and the target surface will not stop and further damage to the agglomerate can still occur, until all the kinetic energy of the system has been dissipated. Several simulation tests [9-11, 18] indicate that the agglomerate fracture in terms of cluster size distribution will not significantly change after the contact damage ratio reaches a constant value. The time when the damage ratio reaches a constant value can therefore be considered as the characteristic impact time during which the main damage occurs to the agglomerate. This trend is also observed here in Figure 9, and therefore further simulations beyond this period have not been carried out.
4.3. Extent of breakage and fracture

Simulation results of the extent of breakage at different impact velocities are shown in Figure 10. The extent of breakage is defined as the ratio of mass of clusters separated from the parent agglomerate (excluding the residual cluster) after impact and the mass of the original agglomerate. This procedure has been adopted in order to be able to compare the simulation results with the experimental data (see Section 5). For the computer generated agglomerate, an interface energy of $\Gamma = 0.5 \text{ J m}^{-2}$ was used to bond the primary particles. Other values of the surface energy have also been tested. However, it was found that $\Gamma = 0.5 \text{ J m}^{-2}$ provided the best fit with the experimental data.

Figure 11 shows the size distribution of the fragments obtained from the simulation. It is seen that, for low impact velocities at which residual clusters survive, the size distribution of singlets and small clusters generated by impact can be represented in a log-log plot by a straight line with a slope that does not vary appreciably with impact velocity. This behaviour is similar to the trend reported by Arbiter et al. [6] and Kafui and Thornton [10, 11]. At a high impact velocity of 10.0 m s$^{-1}$, however, the fraction undersize is much higher than that of low impact velocities and the largest fragment is only one tenth of the size of the original agglomerate, i.e. disintegration of the agglomerate occurs as shown in Figure 4. This behaviour can also be seen in the high speed video of the experimental work, which is presented in Section 5.

5. EXPERIMENTAL

Experimental tests of agglomerate impact have been carried out to compare with the results of the computer simulations. The samples of lactose agglomerates were prepared by Glaxo-Wellcome Research and Development. The experimental apparatus is shown in Figure 12. It consists of a funnel-shaped inlet section guiding the agglomerates into a tube where the agglomerates are accelerated to the required velocity. The tube enters a chamber in which the agglomerates are impacted on a
rigid flat target, and then collected on a filter. The agglomerates are fed gently in a single array into the funnel. Near the exit of the tube the agglomerates pass through two horizontal light beams. The blocking of the light beams is detected by two light sensors, positioned at a vertical distance of 16 mm apart. The sensors trigger a timer, which provides a transit time for determination of particle velocity.

Samples provided for testing had previously been kept under dry air conditions. The ambient air to which the agglomerates were exposed at the time of testing was at approximately $22 \pm 2^\circ C$ and relative humidity of about $37 \pm 2\%$. The size of the agglomerates is in the range of 250-355 μm, with primary particles from 1 to 10 μm. The range of velocities were from the free fall velocity from a height of about 1 m up to an air-assisted velocity of about $10 \text{ m s}^{-1}$. A small mass of agglomerates, about 2 g, was subjected to one impact only.

After impact, the agglomerates were collected in a brass tray, and then transferred into a small aluminium tray for weighing. First, the mass of the particles collected was weighed, then the particles were sieved using BS410 sieves, and finally the mass of the mother particles and the debris was weighed separately. Debris was defined as the particles that passed through a sieve with a size of two B.S. sieve sizes below the lower size limit of the original particles. Particles that did not pass this sieve were considered as the parent agglomerates or the residual clusters. Both the parent agglomerates and the debris were stored in separate vials for later optical analysis. Since the agglomerate size was in the range of 250-355 μm, the sieve size for separating the debris from the parent agglomerates was 180 μm. This procedure for separating debris from the parent agglomerates was deliberately adopted to minimise sieving in order to avoid further damage to the agglomerates. It is a particularly convenient method when the parent agglomerates remain relatively intact so that the fractional loss from the parent agglomerates can be accessed. In the low range of impact velocities, where surface damage mainly occurs, the debris are much smaller than the original agglomerate. Cleaver et al. [28] have shown that the quantity of debris in this case is insensitive to the choice of sieve size for the separation of debris.
as long as it lies in between the size distributions of the debris and the residual/original agglomerates.

A similar approach has been adopted in interpreting the simulation results, as described in Section 4.3. In computer simulations, the choice of the cluster size, below which clusters are regarded as debris, was based on the sieve size used for experimental tests. Referring to Figure 1, the clusters resulting from impact except the residual clusters were all less than 180 μm, and consequently they can be regarded as debris whose mass is used to calculate the loss per impact. Therefore, the simulation results are also insensitive to the choice of cluster size separating debris from the original agglomerate.

In the experimental tests, the extent of breakage is considered as the percentage loss from the parent agglomerates and is calculated using the mass of debris generated upon impact divided by the mass of original agglomerates. During handling, not all the material originally fed to the impact test device can be recovered after impact as material loss is inevitable. The material loss can occur by either the loss of parent agglomerates or debris. For the former, the losses cannot be attributed to the breakage. In describing the extent of breakage, it is therefore not possible to report a single value for it, and an assumption has to be made about the source of losses. It is appropriate to define an upper and lower limit of the breakage rate, depending on whether the losses are attributed to the debris or the parent agglomerates. The actual extent of breakage lies somewhere in between these two limits. The upper limit is given by $\xi^+$:

$$\xi^+ = \frac{W_f - W_m}{W_f}$$  \hspace{1cm} (2)

where $W_f$ denotes the mass of particles fed originally into the eductor device, and $W_m$ is the mass of parent agglomerates after collection and separation by sieving.

The lower limit of breakage is given by $\xi^-$:

$$\xi^- = \frac{W_d}{W_c}$$  \hspace{1cm} (3)
where $W_d$ denotes the mass of debris after collection and separation by sieving, and $W_e$ the mass of parent agglomerates and debris collected after impact, before separation by sieving. Experimentally measured values of the breakage extent of lactose agglomerates are also shown in Figure 10. Notwithstanding the considerable spread between the upper and lower limits of attrition rate, generally a good agreement with simulation results is shown.

Qualitative information has been obtained from the experimental test by optical microscopy, to determine the visual features of the samples before and after impact. It was observed that, at an impact velocity of $4.0 \text{ m s}^{-1}$, the debris consisted of clumps of uniformly fine grains and a large quantity of loose grains, while the surfaces of the parent agglomerates had been abraded, causing a decrease in sphericity. It was also seen that some of the fine debris stuck to the surfaces of the parent agglomerates due to the existence of high adhesion forces. For an impact velocity of $2.0 \text{ m s}^{-1}$, it was observed that there were some fragments with round edges and finer debris sticking to the parent agglomerate. For most parent agglomerates, it was hard to identify the damage location and only a small portion of the sample showed flattened surfaces.

More detailed qualitative information on the process of disintegration of the agglomerates upon impacts has been obtained from high speed digital video recordings. These have been made with a Kodak HS4540 Motion Analyzer video camera, which stores up to 50000 digital images in D-RAM memory, at acquisition frequencies up to 40500 frames per second. This camera has been equipped with a high magnification macro lens, yielding a close view of the particles on impact with a calibrated viewport size of $5.0 \text{ mm}$ wide by $2.5 \text{ mm}$ high. The captured images can subsequently be down-loaded to a video tape. A small sequence of images, acquired at $27000$ frames per second, is shown in Figures 13(a-g), for an agglomerate impact at a velocity of $10.0 \text{ m s}^{-1}$. The extent of deformation upon impact (Figures 13(a-c)) and large-scale disintegration (Figures 13(d-f)) can be clearly seen. Only the debris show a certain degree of rebound, assisted by the surrounding air flow. After the debris has been carried away by the air flow, a small and compacted part of the
agglomerate sticks to the target (Figure 13(g)). The similarity in the geometry of the agglomerate at different stages of impact to the simulation results in Figure 4 is clearly shown, i.e. extensive deformation of the agglomerate followed by the detachment of a large number of debris. It should be noted, however, that comparison with Figure 4 is only possible up to the stage shown in Figure 13(c), since the simulation has not been continued beyond this stage of impact. During the later stages of impact, the air flow, which is not modelled in the simulations, has a significant effect on the dispersion of the debris.

6. DISCUSSION

An early experimental investigation of agglomerate breakage was reported by Arbiter et al. [6], who conducted free-fall impact tests of sand-cement agglomerates and spherical glass balls. In their results, the fragments resulting from an impact can be classified into three categories: the coarse fragments (residual), the fine fragments (complement) and the "dust". In a logarithmic plot of the cumulative fraction (by weight) of undersize fragments against fragment size, each category of fragments is represented by one of three straight lines for a given impact velocity. The agglomerates of Arbiter et al. [6] failed in a brittle or semi-brittle mode, as planar crack surfaces were observed, indicative of crack propagation along tensile stress trajectories. The same behaviour has also been observed in computer simulations of agglomerate impact damage by Thornton [26], who used agglomerates consisting of approximately 8000 monodispersed primary particles in a face-centred cubic packing. In the present study, the small fragments can also be represented by one straight line in a similar plot as shown in Figure 11. However, no crack propagation has been observed in both experimental and simulation investigations presented in this paper. The dynamic features of the particle system reported by Kafui and Thornton [10, 11] are distinctly different from those in our study, as presented in Figures 5-8. The profile of the wall force and kinetic energy over the impact duration, reported by Kafui and Thornton, appeared to be similar to those of solid particle impacts. When the force on the wall reached its maximum value in the loading process, the kinetic energy of the particle system was approaching its
minimum value. This indicates that the kinetic energy is transformed into elastic strain energy during the loading process, and after the peak force is attained, the stored energy is then released to furnish agglomerate rebound. Such a behaviour also applies to solid particle impacts under elastic and elasto-plastic deformations [18]. In this study, however, the lactose agglomerates are so ductile that they undergo extensive "plastic deformation" even at low impact velocities, at which the residual cluster survives. At high impact velocities, disintegration of the agglomerate occurs. For the investigated range of impact velocities, singlets and small clusters are always separated from the original agglomerate, and the bulk agglomerate itself does not bounce back during the impact process. Consequently, the force on the wall never reaches zero and the kinetic energy of the agglomerate system continues to decrease until a final state of equilibrium is approached.

For the weak agglomerates investigated in this study, it is difficult to characterise the failure mode. Since extensive plastic deformation was observed during the impact process, the lactose agglomerates here appear to fail in a ductile mode according to the visual observations. Further work is needed in order to relate the bulk failure behaviour of the assembly to the fracture of each individual bond and to elucidate the effect of agglomerate size. There is little literature coverage on the breakdown of weak agglomerates. Several previous studies of the solid particle impact breakage in the semi-brittle failure mode have shown that the extent of breakage is proportional to the square of the impact velocity, \( \xi \propto V^2 \) [29-31]. This is valid only in the chipping regime, where sub-surface lateral crack propagation produces small debris detaching from the particle surfaces. The computer simulation results of Thornton [26] have shown a similar dependence for strong agglomerates which produce clear planar cracks on failure. In this study, a similar trend is observed for the dependence of the extent of breakage on impact velocity for the weak agglomerates albeit a different failure mode prevailing. This is shown in Figure 14, where a squared power law relationship applies to the experimental and simulation results. The above evidence suggests that there possibly exists a general dependence of impact breakage of particulate solids on impact velocity squared. This implies that the primary processes involved in the dissipation of energy are
dependent on interparticle friction, rather than the interface energy which may have an indirect effect in influencing the interfacial friction. As suggested in Figure 3, interparticle sliding could play a significant role in breaking contacts for this type of agglomerates. However, a detailed explanation of the dependence of breakage on impact velocity requires further investigations.

7. CONCLUSIONS

Breakage of weak lactose agglomerates has been investigated by computer simulations using the distinct element analysis and by experimental tests using air-accelerated agglomerates colliding on a target surface. Good agreement was found between the simulation and experimental results on both qualitative and quantitative grounds. It is therefore shown that by quantifying the single particle properties, it is possible to predict the behaviour of agglomerate breakage. The effect of impact velocity on agglomerate breakage was examined and it has been found that the extent of breakage varies roughly with the square of impact velocity, a behaviour which is in agreement with the results reported in the literature for solid particles as well as for strong agglomerates. However, significant differences have been found in the loading compliance and dynamic features of weak agglomerates, as compared with those observed in solid particles and brittle agglomerates during the process of impact. With respect to the characteristics of agglomerate failure, both simulation results and experimental observations show that the tested agglomerates exhibit macroscopically a highly "ductile" behaviour, i.e. extensive plastic deformation occurs around the impact area for all the investigated impact velocities. It is also observed that, for low impact velocities, residual clusters survive with a number of singlets and small clusters separated from the original agglomerates; for high impact velocities, disintegration occurs during the impact. Further work is required in order to characterise the failure behaviour and to explain the failure mechanisms of "weak" agglomerates.
Acknowledgements

The authors are grateful to Dr G.W. Hallworth, Glaxo-Wellcome Research and Development, for preparing the lactose agglomerates; to Mr P.A. Arteaga for providing the data on some of the mechanical properties of lactose crystals, measured by nano-indentation; and to Glaxo-Wellcome Research and Development for approving the publication of the experimental data presented in this paper.

REFERENCES


Figure Captions

Figure 1. Lactose agglomerates after impact: white - singlets; light grey - clusters containing 2 - 10 particles; black - clusters containing 11 - 30 particles; dark grey - residual clusters.

Figure 2. Residual cluster (I) and contact breakage diagram (II) at different impact times for an impact velocity 4.0 m s\(^{-1}\).

Figure 3. Schematic diagram of the distribution field of the broken contacts: (a) overall contact breakage pattern: dashed lines - direction of compression; solid line - orientation of broken contacts; (b) contact breakage in the local area A.

Figure 4. Progression of agglomerate breakage at 10 m s\(^{-1}\) with time: (a) \(t = 10.6\) \(\mu\)s; (b) \(t = 17.7\) \(\mu\)s; and (c) \(t = 17.7\) \(\mu\)s with the singlets removed (grey scale colouring as that indicated in Fig. 1).

Figure 5. Evolution of the number of particles in contact with the target wall.

Figure 6. Evolution of the total force on the wall.

Figure 7. Relationship between the wall force and the extent of flattening in the early stages of the agglomerate impact.

Figure 8. Evolution of kinetic energy with time within the agglomerate systems.

Figure 9. Evolution of damage ratio with time over the impact process.

Figure 10. Experimental and computer simulation results of the extent of breakage as a function of impact velocity: \(\xi^+\) and \(\xi^-\) - upper and lower limits of experimental attrition data; \(\xi^*\) - DEA simulation data.

Figure 11. Fragment size distribution under different impact velocities.

Figure 12. Experimental apparatus.

Figure 13. A sequence of images captured by high speed digital video recording at 27000 frames per second for an impact velocity 10.0 m s\(^{-1}\).

Figure 14. Curve fitting for a square power law relationship between the breakage extent and impact velocity with \(\xi^+\) and \(\xi^-\): upper and lower limits of experimental attrition data; \(\xi^*\): DEA simulation data.
Figure 1. Lactose agglomerates after impact: white - singlets; light grey - clusters contain 2 - 10 particles; black - clusters contain 11 - 30 particles; dark grey - residual clusters.
Figure 2. Residual clusters (I) and contact deletion diagrams (II) at different impact times for an impact velocity $4.0 \text{ m s}^{-1}$.

(a-I) $t = 1.714 \mu s$, 1978 particles; (a-II) $t = 1.714 \mu s$, 1473 deleted contacts;

(b-I) $t = 3.428 \mu s$, 1866 particles; (b-II) $t = 3.428 \mu s$, 3026 deleted contacts;

(c-I) $t = 6.856 \mu s$, 1675 particles; (c-II) $t = 6.856 \mu s$, 3265 deleted contacts;

(d-I) $t = 10.63 \mu s$, 1497 particles; (d-II) $t = 10.63 \mu s$, 3574 deleted contacts;
Figure 3. Schematic diagram of the distribution field of the broken contacts:
(a) overall contact breakage pattern: dashed lines - direction of compression; solid line - orientation of broken contacts; (b) contact breakage in the local area A.

Figure 4. Progression of agglomerate breakage at $V = 10$ m s$^{-1}$ with time: (a) $t = 10.6$ μs; (b) $t = 17.7$ μs; and (c) $t = 17.7$ μs with the singlets removed (colour coding as that indicated in Fig. 1).
Figure 5. Evolution of the number of particles in contact with the target wall.

Figure 6. Evolution of the total force on the wall.

Figure 7. Relationship between the wall force and the extent of flattening in the early stages of the agglomerate impact.
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Figure 14. Curve fitting for a square power law relationship between the breakage extent and impact velocity with $\xi^+$ and $\xi^-$: upper and lower limits of experimental attrition data; $\xi^*$: DEA simulation data.
Appendix E

Effects of Particle Size and Bond Strength on Impact Breakage of Weak Agglomerates

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Effects of particle size and bond strength on impact breakage of weak agglomerates

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ABSTRACT: Breakage of weak lactose agglomerates due to impact on a target plate has been investigated by computer simulation using the distinct element method (DEM). The agglomerates of interest here are generally weak and easy to disintegrate as no binder other than weak surface forces is holding the primary particles together. Particle interaction laws in the simulation code are based on theoretical contact mechanics, where van der Waals adhesion forces determine the bond strength between individual particles in the assembly. Experimental investigations have also been conducted to compare with the computer simulation results of lactose agglomerates. A good agreement has been found between the simulation results and experimental measurements. New types of agglomerate have been constructed by varying the size and bond strength of the constituent particles and the resulting mechanical behaviour has been characterised. For the simulation study, the particle size and bond strength have shown significant influences on the dynamics of agglomerates. Further experimental work is required to confirm this behaviour.

1. INTRODUCTION

The design of agglomerate structure for enhanced performance is of great industrial interest to many processes involving powders. This can be done most effectively by computer simulation using the distinct element method (DEM). The method has been developed and successfully applied to the modelling of granular solids in the
past two decades (Cundall & Strack 1979; Thornton & Yin 1991). By direct numerical simulation of the particle interactions, the bulk behaviour of the assembly can be related to the mechanical properties of individual particles. Recently, surface adhesion has been incorporated into the 3-D computer code TRUBAL (Thornton & Yin 1991), which makes it possible to investigate the mechanical behaviour of agglomerates by computer simulation. In this paper, we analyse the breakage of weak lactose agglomerates due to an impact on a target plate and examine the effects of particle size and bond strength of the constituent particles on the dynamics of the agglomerates. The results of computer simulations are compared with the extent of breakage measured by experiments. Computer graphics of the simulation results of agglomerate breakdown are also compared with the images obtained by high speed video recording of the impact events.

2. AGGLOMERATE ASSEMBLIES

The computer program used for agglomeration and impact testing presented in this paper is based on the version of TRUBAL developed by Thornton et al. (1996) and Ning et al. (1996). In the code, the interparticle adhesion is calculated according to the model of Johnson et al. (1971). Planar walls are included in order to perform computer simulations of agglomerate-wall impacts. The damage to individual primary particles is not considered in this study. The lactose agglomerates of interest here are relatively weak and easy to disintegrate as no binder other than weak surface forces is holding the primary particles together. The use of the model of Johnson et al. (1971) is therefore justified for this application. For the simulation, the mechanical properties of the constituent particles in a lactose agglomerate were assumed to be the same as those of large lactose crystals (500-700 μm). For the latter, the mechanical properties have been characterised by the use of nano-indentation techniques (Arteaga 1995) and are summarised in Table 1.
Table 1. Properties of the primary particles and the target used for simulation

<table>
<thead>
<tr>
<th></th>
<th>Lactose crystals</th>
<th>Stainless steel</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Young’s modulus</strong></td>
<td>3.2 GPa</td>
<td>215 GPa</td>
</tr>
<tr>
<td><strong>Poisson’s ratio</strong></td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td><strong>Density</strong></td>
<td>1550 kg m$^{-3}$</td>
<td>7800 kg m$^{-3}$</td>
</tr>
<tr>
<td><strong>Yield stress</strong></td>
<td>0.21 GPa</td>
<td>3.04 GPa</td>
</tr>
<tr>
<td><strong>Friction</strong></td>
<td>0.35</td>
<td>0.35</td>
</tr>
<tr>
<td><strong>Fracture toughness</strong></td>
<td>41.9 kPa m$^{1/2}$</td>
<td>not measured</td>
</tr>
<tr>
<td><strong>Interface energy</strong></td>
<td>0.5 J m$^{-2}$</td>
<td>not measured</td>
</tr>
</tbody>
</table>

Table 2. Properties of the three constructed assemblies

<table>
<thead>
<tr>
<th></th>
<th>Sample 1</th>
<th>Sample 2</th>
<th>Sample 3</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Particle number</strong></td>
<td>2000</td>
<td>1400</td>
<td>2000</td>
</tr>
<tr>
<td><strong>Co-ordination no.</strong></td>
<td>5.3</td>
<td>4.6</td>
<td>5.3</td>
</tr>
<tr>
<td><strong>Solid fraction</strong></td>
<td>0.522</td>
<td>0.45</td>
<td>0.522</td>
</tr>
<tr>
<td><strong>Agglomerate size</strong></td>
<td>300 μm</td>
<td>300 μm</td>
<td>300 μm</td>
</tr>
<tr>
<td><strong>Mean particle size</strong></td>
<td>10 μm</td>
<td>10$^{+}$, 25$^{*}$</td>
<td>10 μm</td>
</tr>
<tr>
<td><strong>Interface energy</strong></td>
<td>0.5 J m$^{-2}$</td>
<td>0.5 J m$^{-2}$</td>
<td>0.5$^{*}$, 2.5$^{+}$</td>
</tr>
</tbody>
</table>

+ outer layer particle radius or interface energy;
* inner layer particle radius or interface energy.
Three types of agglomerate have been constructed by varying the size and bond strength of the constituent particles within the assembly, see Figure 1. There are two agglomerates with a narrow particle size distribution, where for one assembly (Sample 1) the interface energy is the same for all the bonds, and for the other assembly (Sample 3) the strength of the bonds formed by the outer layer particles is about 5 times larger than that of internal particles. The agglomerate of Sample 3 can be envisaged as a surface "coated" agglomerate. The third type of agglomerate (Sample 2) consists of large primary particles surrounded by a few layers of smaller particles but with the same value of surface energy. The properties of the three constructed agglomerates are shown in Table 2. The interface energy of 0.5 $J m^{-2}$ corresponds to a realistic value for a weak lactose agglomerate which is well within the range of measured values reported in the literature (Arteaga 1995 and Roberts 1991).

3. VISUAL OBSERVATIONS

A series of 3-D simulations of agglomerate-wall collisions have been conducted for the three types of agglomerates. For Sample 1, the external appearance of the agglomerate is shown in Figures 2(a)-2(d) after impact at four impact velocities of 1.0, 3.0, 4.0 and 4.5 $m s^{-1}$, respectively. In these figures, the dark grey particles are the particles which form the residual cluster; white particles are singlets; light grey particles are the clusters which contain 2-10 primary particles; and black particles are
the clusters which contain 11-30 particles. It is clear that the number of particles separated from the parent agglomerate increases with an increase of impact velocity. Significant deformation of the agglomerate is visible in the impact region, without the development of a clear crack plane. Detailed inspection of the pattern of the interparticle bond failure indicates that the failure of the agglomerate during the impact process is macroscopically “ductile”, although microscopically the interparticle bond failure is by elastic rupture, i.e. brittle. Figures 3(a) and 3(b) show the simulation results at a high impact velocity of 10.0 m s$^{-1}$, where extensive disintegration is observed. Figure 3(c) shows the behaviour of a real lactose agglomerate of the same size and velocity as the simulated assembly. This has been observed by recording a sequence of images captured by a high speed digital video recorder (Ning et al. 1996). Both experiment and simulation show extensive disintegration of the assembly.

At an impact velocity of 4.5 m s$^{-1}$, Figures 4 and 5 show the cluster distribution of Sample 2 and Sample 3, respectively. For the agglomerate of Sample 2, two residual clusters have survived after impact (see Figure 4(d)). The appearance of the surface “coated” agglomerate (Sample 3) after impact is significantly different from that of the two other assemblies, though plastic deformation occurred to all the samples. No single particle has been separated from the surface of the “coated” agglomerate, since the bond strength of the outer layer particles is much higher than that of the inner layer particles. However, in Figure 5(b), small clusters from the internal part of the agglomerate are shown, suggesting that some of the internal bonds in the centre part of the assembly fail because the interparticle bonding force is weak there, but the separated clusters are contained by the outer layer.
Figure 2. Lactose agglomerate (Sample 1) after impact: white - singlets; light grey - clusters containing 2-10 particles; black - clusters containing 11-30 particles; dark grey - residual clusters.

Figure 3. Progression of agglomerate breakage at 10.0 m s$^{-1}$ with time: (a) simulation at $t = 10.6$ μs; (b) $t = 17.7$ μs (grey density as that in Fig. 2); (c) experiment at different stages of the impact (time shown in the figures is related to the contact time).
Figure 4. Cluster distribution of Sample 2 (agglomerate with a "core") after impact at 4.5 ms⁻¹: (a) all the clusters; (b) clusters with 1-5 particles; (c) clusters with 6 - 32 particles; (d) residual clusters (grey density: white - clusters containing 1 - 10 particles; light grey - clusters containing 32 and 116 particles; dark grey - cluster of 516 particles).

Figure 5. Cluster distribution of Sample 3 (surface "coated" agglomerate) at an impact velocity of 4.5 m s⁻¹: (a) all the clusters; (b) clusters with 1-5 particles (grey density as that in Fig. 2).

4. EXTENT OF BREAKAGE AND FRACTURE

For Sample 1, simulation results of the extent of breakage at different impact velocities are shown in Figure 6. The extent of breakage is defined as the ratio of mass of clusters separated from the parent agglomerate (excluding the residual cluster) after impact and the mass of the original agglomerate. This procedure has been adopted in order to be able to compare the simulation results with the experimental data (Ning et al. 1996), also shown in Figure 6. In the experimental tests, the extent of breakage is considered as the percentage loss from the parent agglomerates and is calculated using the mass of debris generated upon impact divided by the mass of the original agglomerates. During handling, not all the
material originally fed to the impact test device can be recovered after impact and material loss is inevitable. It is therefore appropriate to define upper, \( \xi^+ \), and lower, \( \xi^- \), limits of the breakage rate, depending on whether the losses are attributed to the debris or the parent agglomerates (see Ning et al. 1996). The actual extent of breakage lies somewhere in between these two limits. Generally, it is seen from Figure 6 that a good agreement with the simulation results is obtained.

Figure 6. Experimental and computer simulation results of the extent of breakage (from Ning et al. 1996).

Figure 7. Comparison of the breakage extent between the two assemblies. Figure 7 shows a comparison of the extent of impact breakage between Sample 1 and Sample 2. The significant influence of the primary particle size on agglomerate breakage is shown in the figure. When the impact velocity is less than 4.0 m s\(^{-1}\), the strength of Sample 2 is higher than that of the agglomerate with a narrow particle size distribution, \textit{i.e.} less breakage is observed. However, the extent of breakage increases sharply when the impact velocity is larger than 4.0 m s\(^{-1}\). The trend shown in Figure 7 for Sample 2 suggests that, for this structured agglomerate, there might exist a transition velocity under which the assembly sustains minor breakage and
above which disintegration occurs. This behaviour needs further investigations as it has significant industrial implications.

5. CONCLUSIONS

Computer simulation by the use of DEM provides an effective way to quantify impact breakage of agglomerates and examine the effects of the particle size and the bond strength distribution of the constituent particles. A good agreement in the extent of breakage has been found between the simulation and experimental results. The effects of the particle size and bond strength on the dynamics of agglomerates appear to be significant. Further experimental work is required to confirm this behaviour. The approach is particularly useful for a rapid assessment of the influence of the structure on the strength of agglomerates.

ACKNOWLEDGEMENT

The authors would like to thank Glaxo Wellcome R&D for preparing the samples of lactose agglomerates, Mr P. Arteaga for measuring the mechanical properties of lactose crystals, and the EPSRC for the financial support.

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Appendix F

Disintegration of Weak Lactose Inhalation Drug Excipient Agglomerates
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Disintegration of weak lactose agglomerates for inhalation applications.

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Abstract

Inhalation is a convenient and effective method of drug delivery for a number of common illnesses, many of which affect the respiratory system, such as asthma. Lactose is a commonly used material in the pharmaceutical industry as a carrier of drugs. The use of weak lactose agglomerates in dry powder inhalers depends very much on the way the agglomerates disintegrate. The disintegration of weak agglomerates depends on the bonding mechanism and the agglomerate structure, both of which are to date poorly understood. An experimental study of the disintegration of weak lactose agglomerates is reported here. The effects of agglomerate size and ambient humidity level on the extent and mechanism of breakage are investigated using a single agglomerate impact testing technique. The extent of disintegration is shown to scale with the square of the impact velocity. The dry-kept agglomerates are shown to fail in a mode similar to the ductile failure mode of solid materials, with large overall deformation and internal shearing, without the development of a clear crack plane. Agglomerates which have been kept in a humid environment exhibit a classical semi-brittle failure mode. The experimental results of impact tests with dry-kept agglomerates compare well to the results of numerical simulations using Distinct Element Analysis. The impact test appears to be a useful tool for the determination of the influence of material properties and structure and process conditions on the breakage behaviour.

Keywords: Inhalation, Agglomeration, Particle Technology, Lactose, Breakage
1. Introduction

Inhalation is a convenient and effective way of drug delivery for many illnesses. It has many advantages over alternative methods of drug delivery, such as injection and tablets. Inhaled drugs are particularly favoured for respiratory illnesses, as they are delivered directly to their site of action. Compared to injections and oral delivery, generally smaller doses are needed, thus reducing the risk of side effects, and the action of the medication is swifter, thus making them suitable for treating symptoms immediately. Furthermore, the method is particularly attractive if the medication is likely to get broken down in the digestive system when using tablets.

Considerable effort is being spent to package a variety of drugs for inhalation therapy. The current technology is based mainly on "metered dose" inhalers using propellants. For environmental reasons, however, there are serious concerns about the use of CFC based propellants. Furthermore, the presence of the propellant requires the use of additional ingredients to render the formulation of the drug successful in combination with the propellant. An additional problem is the control of the powder size during dispensing. These problems have led to the development of dry powder inhalers. In these devices, a dose of the drug is dispensed in a powdered form and the energy of the patient's breath intake is sufficient to deliver the dose. However, lack of control of the disintegration causes a significant proportion (up to 85-90%) of the drug to be deposited in the oro-pharynx (Merec Briefing, 1993). The precise delivery of the right dose in the right particle size is a challenging problem, the solution of which would make this technique suitable for use with a wide range of drugs, not only for asthma, but also for antibiotics, insulin for diabetes and medication for common influenza.
In *dry powder inhalers*, the drug (in fine particulate form), is mounted/coated on the large particles of an excipient such as lactose. For the process to be effective, the drug has to be in fine particulate form, typically in the micrometre range or smaller. However, for precise dosage and ease of dispensing, the fine particulates may be agglomerated on their own or with an excipient such as lactose into much larger particles. These agglomerates should disintegrate thoroughly after dispensing so that the fine particles can be inhaled deep into the lungs. The agglomerate must therefore be weak enough to disintegrate on a low energy impact and disperse for inhalation. On the other hand, the agglomerate should be sufficiently strong to withstand breakage during storage and transport. There is therefore a narrow window for which the strength of the agglomerate needs to be carefully tailored. A knowledge of the mechanical properties of the agglomerates, the disintegration characteristics of the agglomerate, and the environmental factors, such as humidity, is very important for reliable design and operation of dry powder inhalers. In the present study, the breakage of weak lactose agglomerates is quantified as a function of impact velocity using a single agglomerate impact testing technique. The mechanism of breakage is visualised using high speed video recordings of the impacts and the product of the breakage are examined by Scanning Electron Microscopy (SEM) and optical microscopy. Two different size ranges of the agglomerates are used to evaluate the effect of agglomerate size on the extent of breakage. The effect of humidity is also evaluated by testing a dry sample and a sample which has been exposed to a humid environment for a certain period of time. In the following, the test method is described first and the results are discussed and compared to numerical predictions of the breakage behaviour, obtained from Distinct Element Analysis.
2. Experimental

2.1 Materials

The materials used in the test are weak agglomerates of α-lactose, formed by dry tumbling of fine powder, and were prepared by Glaxo-Wellcome R&D (Hallworth, 1995). Figure 1 shows an optical micrograph of an agglomerate of about 700 μm. The agglomerate has a "fluffy" appearance because of the highly irregular surface formed by the adhesion of the primary particles, which are only a few microns in size. Figure 2 shows loose primaries which have detached from the agglomerate. They exhibit clear flat surfaces and high transparency. Figure 3 shows the primaries at the surface of the agglomerate. The primaries are bonded by surface adhesion forces only, and no material binder is present.

Pietsch (1991) gives a number of examples of surface adhesion forces in the absence of solid or liquid bridges and binders between primaries. They can consist of molecular (Van der Waals) forces, electrostatic forces, or free chemical bonds (valences). In the case of lactose agglomerates, the adhesion forces belong to the first category, which can attain very high values at extremely short distances, but they diminish quickly with increasing distance.

The internal structure of the agglomerate is determined largely by the number and nature of the points of (near) contact or co-ordination points, and depends on the production method. The presence of humidity may cause partial dissolution at the contact points, leading to the formation of liquid and solid bridges. The ambient humidity is therefore an important parameter in the breakage mechanism of weak lactose agglomerates.
Two different particle size ranges in single BS 410 sieve cuts were used, 250-355 µm and 600-710 µm. The samples were kept at two different humidity levels. One sample of both size ranges was kept dry in a desiccator with silica gel. Of the larger size range, one sample was kept in a humid environment. The humid environment was created in an airtight chamber, which contained a reservoir of a saturated solution of sodium carbonate monohydrate in distilled water. The temperature of the cupboard was kept at 24.5 °C. At this temperature, the relative humidity reaches a value of 87% (Wexler, 1965). The samples were left in this environment for 72 hours. The humidified samples were observed to have lost most of their "fluffy" appearance and obtained a shiny smooth surface.

2.2 Method

A batch of agglomerates, about 2 g, is fed gently in a single array into an air eductor, shown in Figure 4. The eductor consists of a funnel shaped inlet section, guiding the particles into the eductor tube. Near the exit of the tube, there are two photodiodes, 16 mm apart, providing triggers for a timer for measurements of the particle velocity. After leaving the tube, the particles enter a chamber, where they impact on a sapphire target plate. The impact velocities ranged from the free fall velocity up to 10 m s⁻¹. The air used to accelerate the particles is supplied by a compressor, and is withdrawn from the collection chamber through a brass porous plate. The porous plate is covered with a paper filter (Whatman). The impacts have been recorded using a digital high speed video camera, a Kodak HS 4540 Motion Analyzer, capable of recording rates up to 40,500 frames per second.

After impact, the products are collected from the chamber and classified using BS410 sieves. The particles are separated into two fractions. Debris is defined as the material that passes through a sieve of two standard sizes below the lower sieve
size of the original material. For the two size ranges used here, 250-355 µm and 600-710 µm, the sieve sizes for separating the debris are 180 µm and 425 µm, respectively.

The extent of breakage is defined as the ratio of the mass of debris created upon impact, $M_d$, to the mass of agglomerates originally fed to the impact rig, $M_f$. Handling losses are inevitable, especially with this material in view of its high adhesivity. This leads to errors in the determination of the extent of breakage. If the handling losses are all attributed to the mother particles, a lower limit of the extent of breakage can be defined as:

$$\xi^- = \left( \frac{M_d}{M_f} \right) \times 100\%$$  \hspace{1cm} (1)

If, on the other hand, all losses are attributed to the mass of debris, an upper limit of the extent of breakage is defined as:

$$\xi^+ = \left( \frac{M_f - M_m}{M_f} \right) \times 100\%$$  \hspace{1cm} (2)

where $M_m$ is the mass of surviving agglomerates. In practice the actual value of the extent of breakage is between these two limits and to provide reliable data the losses have to be reduced as much as possible.

3. **Experimental Results**

Figure 5 shows the extent of breakage, expressed as the fractional mass loss defined by equations 1 and 2, versus the impact velocity. The fractional mass loss is larger for the smaller agglomerates when compared to both wet and dry of the larger agglomerates. The extent of breakage of the smaller sample is about twice that of the larger dry sample. The sample that has been exposed to a humid environment appears to be far more resistant to breakage than the dry sample of the same size range, 600-710 µm.
Inspection of the debris of the dry impacted agglomerates using optical microscopy showed that the debris mainly consists of single primary particles and small clusters of only a few primary particles. Cleaver et al. (1993) have shown that the extent of breakage (Eqs 2-3) is insensitive to the criterion of the debris sieve size if the debris is much smaller than the original particles. The debris of the wet agglomerates consists of small flakes, well below the size of the mother agglomerates.

Figure 6 shows a sequence of high speed images captured from the impact of a dry agglomerate at 10 m s\(^{-1}\). The agglomerate is shown to deform strongly upon impact during the first stages of the impact. As the impact progresses, a large number of small fragments, consisting of small clusters and single primaries detach from the agglomerate and disperse into the surrounding air flow. The way the impact progresses for a wet agglomerate is very different from this. The agglomerate shows much less deformation and a few chips, flaky pieces, detach from the agglomerate rather than numerous small clusters and single primaries. This is illustrated in Figure 7, which shows an impacted wet agglomerate at 3.96 ms after the first contact with the target plate. A similar difference in behaviour between dry and wet agglomerate is also illustrated by the behaviour at free fall impact velocity. The dry agglomerate already loses a considerable amount of primaries at the lowest impact velocities, whereas the wet agglomerate was seen to bounce off the plate without incurring significant damage. The surface adhesion forces bonding the primaries of the dry agglomerate are far smaller than the strength of the liquid and solid bridges that sustain the wet agglomerate structure.
4. **Comparison with Distinct Element Analysis**

The distinct element analysis (DEA), first developed by Cundall (1971, 1988), presents a convenient way to obtain insight into the particle systems and provides fundamental information such as microscopic structure, interparticle forces, particle velocities, etc. Most importantly, this method makes it possible to relate the bulk mechanical behaviour of the assembly to individual particle properties. A detailed examination of the micromechanics, which determine the bond breaking and the internal microstructural deformation, can therefore be provided. Simulations of impact fracture of agglomerates have been reported by Thornton *et al.* (1996) for two-dimensional agglomerates (2-D) and by Kafui and Thornton (1993, 1994) for three-dimensional agglomerates (3-D). In the latter, a monodispersed spherical agglomerate consisting of 8000 primary particles in face-centred cubic arrangement was impacted against a wall. In order to analyse some of the above experimental results, Ning *et al.* (1997) have recently carried out distinct element simulations of impacts of weak dry lactose agglomerates, using the code as developed by Thornton and co-workers (Thornton 1991, Thornton and Yin 1991, Ning 1995). The model of Johnson *et al.* (1971) is used for calculation of the interparticle forces in a dry agglomerate system. For the simulations, the mechanical properties of the constituent particles were assumed to be the same as those of lactose crystals. For the latter, the material properties have been characterised by the use of nanoindentation techniques (Arteaga, 1995) and are summarised in Table 1. The agglomerate used in the simulation has an overall size of 300 μm and consists of 2000 particles of 10 μm, with a solids fraction of 0.522 and an interface energy of 0.5 J m⁻². The method of preparation of the agglomerate has been described in more detail by Ning *et al.* (1997).
Figure 8 shows the fragment size distribution upon impact. The impact damage results in several relatively small clusters of up to a few tens of particles, and one residual cluster of much larger size. An interesting point to note here is the self-similarity of the size distributions of the complement for different impact velocities, i.e. having similar slopes of the trend lines when plotted on a log-log scale. For comparison with the experimental data of impacts of dry agglomerates of 250-355 μm (Fig. 5), the total mass of the small clusters is defined as debris, and in this way the extent of breakage is calculated, as shown in Fig. 9. A good agreement between the predictions of the simulation and the experimental results is found. Figure 10 shows the simulated agglomerate at two stages during an impact at 10 m s\(^{-1}\). The agglomerate has deformed and disintegrated. The appearance is very similar to the later stages of the impact recorded in Fig. 6.

5. Discussion

5.1 Breakage Mechanism of Dry and Wet Agglomerates

Breakage of particulate solids is broadly classified in terms of brittle, semi-brittle and ductile failure, depending on the deformation mode (Shipway and Hutchings 1993, Puttick 1980). Brittle failure is caused by fracture with little or no plastic deformation. Impact under this failure mode usually produces diametrical cracks, splitting the particle into fragments, as the diametrical plane is under the greatest tensile stress (Shipway and Hutchings, 1993). If plastic flow precedes the fracture, the process is termed semi-brittle. The plastic zone produces compressive radial stresses and tensile hoop stresses. The latter type of stress propagates radial and median cracks, initiated from the plastic zone. When the load is removed, the residual tensile stresses, formed after the elastic unloading process, generate subsurface lateral cracks. The characteristics of the semi-brittle failure mode are particle
fragmentation due to the formation of median and radial cracks, and chipping due to the formation of lateral cracks (Puttick 1980, Ghadiri and Zhang 1992, Papadopoulos and Ghadiri 1996). Ductile flow is dominated by extensive plastic flow which is responsible for the rupture of the material. Ploughing and cutting are the two main mechanisms of material removal for this failure mode (Hutchings, 1992).

On the basis of a linear elastic fracture mechanics analysis of semi-brittle failure, Ghadiri and Zhang (1992) showed that the impact breakage should scale with the square of the impact velocity. Indeed, the slopes of the trend lines of the fractional mass loss versus impact velocity shown in Figure 9 is 2. The breakage mode of dry agglomerates, however, is not comparable to a semi-brittle failure mode for a solid particle. The dry agglomerates deform considerably upon impact and the debris created consists mainly of single primary particles and small clusters of a few primaries. Wet agglomerates on the other hand, appear to break in a mode which is more similar to that of a semi-brittle solid, with the formation of flaky chips from the agglomerate surface, but considerable plastic deformation is still observed.

Duo et al. (1996) have shown that even the breakage of very tough agglomerates with a porous structure (fluid cracking catalyst) exhibits a square dependence on the impact velocity. Papadopoulos and Ghadiri (1996) have shown that many different materials with widely different structures exhibit the same tendency. From this, it may be inferred that the amount of breakage scales with the kinetic energy upon impact, regardless of the material structure.

5.2 Effect of Agglomerate Size

When comparing the extent of breakage of the small and large dry agglomerates, it is evident from Figure 5 that the extent of breakage is larger for the smaller particles.
This is in contrast to the observed trends for solid particulates for which the extent of breakage increases with particle size. The model of Ghadiri and Zhang (1992) for semi-brittle particulates predicts a linear dependence of the extent of breakage on the particle size. Duo et al. (1996) have verified this experimentally for fluid cracking catalyst particles. For the brittle failure mode, Kendall (1978) relates the increased breakage propensity of larger solid particulates to an increase in the size and density of the pre-existing flaws. It is therefore extraordinary to note that the present agglomerates exhibit a higher resistance to breakage with an increase in size. It is most likely that the structure of the agglomerates is different between the two sizes. As the agglomerates are dry tumbled, the agglomerate growth is by layerwise adhesion of the primary particles. Consequently, the core can get more compacted, resulting in a stronger agglomerate with an internally non-uniform but distributed porosity. The analysis of agglomerate strength by Kendall (1988) shows that both the fracture toughness and the bending strength of agglomerated materials increase strongly with a decrease in void fraction. This aspect has therefore important implications on the manufacturing procedure.

5.3 Effect of Humidity

From Figure 5, it is evident that the agglomerates which have been kept in a humid environment have a far lower breakage propensity. The most likely reason is the transformation of amorphous lactose, produced during the milling operation, into α-lactose monohydrate which could cause the solidification of interparticle contacts. Amorphous lactose is highly hygroscopic. This could lead to surface adsorption, particularly at the contact points, resulting in the formation of solid bridges of the monohydrate form of lactose crystals. The structure of the agglomerate becomes more rigid and thus brittle, as the primaries are no longer capable of sliding along each other, but are fixed in their place by the bridges. This is confirmed by the
formation of flaky chips upon impact, instead of the detachment of small clusters and single primaries from the agglomerate. The formation of crack planes in the former is similar to those found in semi-brittle failure of non-porous particulates (Ghadiri and Zhang, 1992) and brittle spheres (Arbiter, 1969). Sebhatu (1994) found that moisture uptake causes an amorphous-crystalline transition in spray-dried (15% amorphous) lactose powder. They postulated that moisture is adsorbed preferentially in the amorphous regions, setting up conditions for crystallisation, resulting in a higher compact strength.

5.4 Self-similarity of Fragment Size Distribution

For brittle spheres, Arbiter (1969) showed that the fragment size distribution can be represented by a set of three straight lines on a logarithmic co-ordinate system, one for each of the three categories: coarse fragments (residual, or mother particle), fine fragments (complement) and dust. Figure 8 shows that the slope of the complement size distribution is the same for all the impact velocities. Arbiter et al. (1969) observed the same phenomenon with sand-cement spheres. Impact studies of PMMA particles by Papadopoulos (1998) and Distinct Element Simulations of agglomerate impacts by Thornton et al. (1997) show the same as well. This suggests that the fragment size distribution is insensitive to changes in the impact velocity.

6. Conclusions

Breakage of weak lactose agglomerates has been investigated using a single particle impact test method. The breakage mode was identified using high speed digital video recordings and was found to be very different from that of non-porous particulates, reported so far, i.e. extensive deformation takes place which resembles that of highly ductile solids. A strong effect of humidity on the extent of breakage
was found, which is attributed to the change of interparticle bonding mechanism. A comparison of impact breakage of large and small agglomerates showed that large agglomerates are less prone to breakage than smaller agglomerates. This behaviour is in contradiction with that of the solids reported so far, and is considered to have arisen from the difference in the structure of the agglomerate, evolving as a result of the preparation procedure.

A good agreement was found between the experimental results and simulations of impact of small and dry lactose agglomerates using Distinct Element Analysis. Both experiments and simulations show that the impact breakage of dry agglomerates is a function of the square of the impact velocity. This is in agreement with the behaviour of other types of particulates and with the model of Ghadiri and Zhang (1992) for semi-brittle failure. However, the present agglomerates fail in a mode which is more similar to ductile failure of non-porous particulates. The fragment size distribution obtained from the simulations show self-similar trends which are in good agreement with results reported in the literature for materials of natural grain size. Distinct Element Analysis appears to be a useful tool for providing insight into the internal mechanism of breakage of weak dry agglomerates.

Acknowledgments

The authors wish to express their gratitude to Dr G.W. Hallworth of GlaxoWellcome Research and Development for preparing the lactose agglomerates. The authors acknowledge support from the Engineering Physics and Research Council for the purchase of the Kodak High Speed Motion Analyser, used in this work. The authors are grateful to Mr Pedro Arteaga for providing data on some of the mechanical properties of lactose crystals, measured by nano-indentation and used in the DEM simulation.
### Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
</tr>
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<tbody>
<tr>
<td>$M_c$</td>
<td>mass of particles collected after the impact</td>
<td>(g)</td>
</tr>
<tr>
<td>$M_d$</td>
<td>mass of debris</td>
<td>(g)</td>
</tr>
<tr>
<td>$M_f$</td>
<td>mass of particles fed</td>
<td>(g)</td>
</tr>
<tr>
<td>$M_m$</td>
<td>mass of surviving agglomerates</td>
<td>(g)</td>
</tr>
<tr>
<td>$\xi^+$</td>
<td>Upper limit of breakage (Eq. 1)</td>
<td>(-/-)</td>
</tr>
<tr>
<td>$\xi^-$</td>
<td>Lower limit of breakage (Eq. 2)</td>
<td>(-/-)</td>
</tr>
</tbody>
</table>
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Hallworth, G.W., 1995, Personal Communication

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Table 1.
Material properties used in the simulations.

<table>
<thead>
<tr>
<th></th>
<th>Lactose crystals</th>
<th>Stainless steel</th>
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<tbody>
<tr>
<td>Young’s modulus</td>
<td>3.2 GPa</td>
<td>215 GPa</td>
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<tr>
<td>Poisson’s ratio</td>
<td>0.3</td>
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<td>Density</td>
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<td>7800 kg m(^{-3})</td>
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<td>Yield stress</td>
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<tr>
<td>Friction</td>
<td>0.35</td>
<td>0.35</td>
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<tr>
<td>Fracture toughness</td>
<td>41.9 kPa m(^{1/2})</td>
<td>NA</td>
</tr>
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</table>
Figure Captions

1. A lactose agglomerate

2. Single and small clusters of primary particles

3. Lactose agglomerate surface

4. Single particle impact test rig

5. Fractional mass loss as a function of impact velocity.

6. A sequence of images captured by high speed digital video recording at 27000 frames per second of an agglomerate impact at a velocity of 10.0 m s\(^{-1}\).

7. Impact of a wet agglomerate at 8 m s\(^{-1}\) at 3.96 ms after the impact.

8. Fragment size distribution.

9. Comparison of simulation of 300 \(\mu\)m agglomerate impact with experimental results for 250-355 \(\mu\)m agglomerate impacts.

10. Simulation of an impact of an agglomerate at 10.0 m s\(^{-1}\) at (a) \(t = 10.6\) \(\mu\)s, (b) \(t = 17.7\) \(\mu\)s after contact; colour coding: white - singlets; light grey - clusters containing 2 - 10 particles; black - clusters containing 11 - 30 particles; dark grey - residual cluster.
Figure 1. Lactose agglomerate

Figure 2. Single and small clusters of primary particles
Figure 3. Lactose agglomerate surface

Figure 4. Single particle impact test rig
Figure 5. Fractional mass loss as a function of impact velocity.

250-355 μm: ○, ξ⁺ dry; +, ξ⁻ dry.

600-710 μm: ■, ξ⁺ dry; ●, ξ⁻ dry; ×, ξ⁺ wet; ▲, ξ⁻ wet.
Figure 6. A sequence of images captured by high speed digital video recording at 27000 frames per second of an agglomerate impact at a velocity of 10.0 m s\(^{-1}\).
Figure 7. Impact of a wet agglomerate at 8 m s\(^{-1}\) at 3.96 ms after the impact.

![Figure 7](image)

Figure 8. Fragment size distribution.

![Figure 8](image)

Figure 9. Comparison of simulation of 300 µm agglomerate impact with experimental results for 250-355 µm agglomerate impacts.

![Figure 9](image)
Figure 10. Simulation of an impact of an agglomerate at 10.0 m s\(^{-1}\) at (a) \( t = 10.6 \mu s \), (b) \( t = 17.7 \mu s \) after contact; colour coding: white - singlets; light grey - clusters containing 2 - 10 particles; black - clusters containing 11 - 30 particles; dark grey - residual cluster.
Appendix G

High Speed Video Image Analysis of Flow of Fine Particles in Fluidised Bed Jets

HIGH SPEED VIDEO IMAGE ANALYSIS
OF FLOW OF FINE PARTICLES IN FLUIDISED BED JETS

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ABSTRACT

The hydrodynamics of gas/solids mixtures, e.g. in fluidisation and pneumatic conveying, has a distinct effect on the breakage and degradation of powders. Visualisation by means of high speed video imaging of these processes enables a better understanding of the mechanics of these processes and verification of the models developed for describing the process. In this paper, the development and use of high speed photography and video imaging and image processing and analysis for the measurement of particle velocities and concentrations, and divergent angles of gas jets in fluidised beds are described. The implications of the observations and measurements on design and control of these processes are indicated.

1. INTRODUCTION

Particles suspended in a fluid flow may exhibit a fluid-like behaviour. This phenomenon, termed as fluidisation, brings many advantageous features to process engineering, e.g. ease of solids handling and mixing, increased heat transfer, uniform reagent concentrations and uniform temperatures. Fluidised beds are used widely in the process industry, e.g. in the production of common salt, sugar and detergent powders, and in the oil refinery. However, they could suffer from attrition, the unintentional breakage of particles during their processing, handling and storage. This causes the formation of debris leading to degradation in quality and environmental hazards [1]. The fluid is introduced vertically upward into the bed by a distributor grid at the bottom of the container vessel. The gas velocity through the orifices in the grid is much higher than the particle terminal velocity, thus entraining the particles in the jet. Particles experience large velocity gradients in the vicinity of
the grid, where the attrition of particles commonly occurs [2]. Ghadiri and Boerefijn [3] have recently presented a comprehensive overview of correlations available for the characterisation of the attrition propensity of particles in fluidised beds. These correlations are all empirical, and do not take account of the hydrodynamics of the system, and therefore they have a limited scope of application. To obtain a fundamental understanding of the processes involved it is necessary to characterise the hydrodynamics of the system. In the following, the development and use of high speed video imaging and analysis facilities for characterisation of the hydrodynamics of the jetting region of fluidised beds will be described.

2. **MODELLING APPROACH**

A detailed description of the modelling approach is given elsewhere [3]. A summary of the modelling approach is schematically illustrated in Figure 1. A central position is taken by the attrition model itself. The attrition mechanism in a fluidised bed jet involves the entrainment of particles from the bulk surrounding the jet into a dilute jet core, followed by the acceleration of the particles, until they impact on the dense phase on top of the jet. The main assumption made in the modelling approach concerns the similarity between the breakage of a particle in a single impact on a rigid target and the breakage of a particle which becomes entrained into the dilute core, accelerated and impacted on the dense phase. The breakage of a single particle can be measured using a single particle impact testing method, developed previously by Ghadiri and co-workers (see e.g. Yüregir *et al.* [4]). The extent of attrition of particles is related to the number of particles entrained into the jet. The velocity at which the particles impact in the jet can be predicted using hydrodynamic models of the jet region. These models can also predict the entrainment rate. Thus, the combination of a single particle impact test and a model of fluidised bed jet hydrodynamics provides the basis for predicting the attrition in fluidised bed jets.

The single impact test is used to yield information on the extent and mechanism of breakage of particles at a specified impact velocity. A schematic of the experimental facility is shown in Figure 2. Particles are fed gently in a single array into the funnel, and are accelerated downward on the flow of air, until they
impact on the target plate of sapphire in the collection chamber. Sieving and
gravimetric analysis are used to quantify the breakage.

The hydrodynamic model of fluidised bed jets, developed by Massimilla and
co-workers [5] requires input of a number of hydrodynamic parameters, such as the
angle at which the jet diverges as it extends vertically into the bed, the initial solids
concentration at the jet nozzle, and the jet height. Several correlations are available
for the jet height [6], but for the jet angle and the solids concentration very few data
can be found in the literature [7, 8]. The approach for characterisation of these
parameters forms the basis of this paper.

3. EXPERIMENTAL SET-UP

Measurements of the hydrodynamic parameters of the jet have been carried out with
the experimental set-up, shown in Figure 3. The lower section of this two-di

mensional fluidised bed is made of 10 mm thick optical glass sheet, 1.5 m high and
0.25 m wide. The internal bed width is 12.5 mm. A central aluminium nozzle is set
flush in between two brass porous plates, which can be slid sideways in order to
accommodate a range of differently sized nozzles. The nozzle is set in a chamber,
made of Perspex, below the porous plates, which provide an even distribution of the
flow for background fluidisation. The chamber and the nozzle are both fed with
compressed air from separate rotameters. The nozzle has the shape of a slot across
the width of the bed, and the dead zone at each side of the slot is 0.5 mm because of
its wall thickness. The slots of the nozzles used in this work were 0.2, 0.5, 1.0, 5.0,
and 8.0 mm wide. Orifice velocities ranged up to about 60 m s\(^{-1}\). The gas jet formed
at the orifice is a flame-like void in the dense bulk surrounding it, swaying somewhat
from side to side. The upper section of the bed is made of 12.5 mm Perspex sheet,
on the inside flush with the lower section, and 0.43 m high. Entrained solids in the
particle size range 30-60 \(\mu m\) are returned to the bed by a cyclone with a horizontal
vortex, smaller particles are caught by a 0.2 \(\mu m\) absolute retention Sartopure\(^1\) pleated
paper filter. The particles used are 500 \(\mu m\) common salt and 90 \(\mu m\) silica-alumina
catalyst.
For measurement of the solids concentration and particle velocities, several different optical configurations have been used. Generally, a camera was positioned in front of the jet at a certain vertical position with reference to the nozzle exit, and images were captured. In the case of a photographic camera, the pictures were scanned and downloaded digitally to the automatic image analysis system. In the case of video images, the images were downloaded from the video recorder to the automatic image analysis system. The image analysis system is PC based and equipped with a frame grabber board. The frame grabber board is an ITI IC-PCI board with a so-called AM-CLR analogue colour acquisition module, capable of acquiring images in various formats (digital 24-bits R-G-B at 1024 x 1024 pixels, analogue S-VHS (Y/C) in colour, analogue VHS (C-Sync) in colour and monochrome). Even the most advanced professional video-recorders suffer from a certain fluctuation of the video output signal frequency. This resulted in mismatched acquisitions, the acquisition module would not lock on to the video signal and fail to display or would produce a jittered and skewed image. For this reason, trials were carried out with video-recorders equipped with an internal time base corrector, but without success. Finally, it was decided to include an external digital time base corrector in the system, a FOR-A FA-310P, which provides a perfectly stable video signal, both in jog/shuttle and normal play motion. The software used to analyse the images is Optimas versions 5 and 6. This Windows-based software has a very powerful macro-language, and provides many convenient processing tools and facilities. The images are treated on a numerical basis, i.e. they are essentially a matrix of luminance values at pixel positions.

1Sartopure is a registered trademark of Sartorius AG, Goettingen, Germany
2Imaging Technology Inc., Bedford MA, USA
3FOR-A UK Ltd, London, UK
4Optimas is a registered trademark of Optimas Corp., Seattle, USA
5Windows is a registered trademark of Microsoft Corp., Seattle, USA
3.1 Particle velocity measurements

For particle velocity measurements, a Kodak Ektapro Motion Analyzer HS 4540 was used. This camera provides frame rates up to 40,500 fps. It records digitally in black-and-white in full frame rate up to 4,500 fps. The frames are stored digitally in large on-board memory (192 MB). A full frame measures $256 \times 256$ pixels, and at 40,500 fps the frame size is reduced to $64 \times 64$ pixels. The viewport is square, even at this high frame rate, so that motion can be traced into two dimensions. The digital image sensor is highly light-sensitive, and only at frame rates beyond 4,500 fps is the light-sensitivity notably dependent on the frame rate. Two halogen spotlights of 50W were generally sufficient even at the highest frame rates. The digital frame store eliminates the need for the mechanical tape drive, which was particularly troublesome and required great care and constant attention. The digital frame store can hold 3,072 full frame images. As the frame rate goes up beyond 4,500 fps, the frame size is reduced accordingly, so that the memory can hold a larger number of images, up to 49,152 at 40,500 fps. Therefore, the minimum recording time is never reduced below 0.75 seconds, which is a very long period in terms of high-speed events. The system is equipped with a Digital Download Interface (DDI), which enables direct image transfer in Tagged Information File Format (TIFF) to a PC via an IEEE protocol GPIB-PCII(A)$^6$ interface card. Via the DDI, the camera can be completely remote controlled.

Once the images have been acquired, stored and transferred to the PC, they are analysed with Optimas software. Based on the grey scale luminance values, a particle can be readily identified in an image. Figure 4a shows a common salt particle of about 500 μm in size in a dense jet particle flow, recorded with the HS 4540 at 13,500 fps. The particle has been identified automatically by Optimas, using a grey scale threshold selection criterion. Additional selection criteria may concern the circularity of the object, to prevent selection of doublets and triplets and to aid in the identification of the same object in subsequent images. Figure 4b shows the same particle in the next image, acquired 74 μs later. Figure 4c repeats the old position marker of Fig. 4a in Fig. 4b, to indicate the distance travelled. The

$^6$National Instruments Corp., Austin TX, USA
calibrated distance travelled between the centroids of the marked areas is approximately 120 μm, yielding a velocity of 1.6 m s⁻¹. In general, Optimas will identify about 20 objects in a sequence as shown in Figure 4, so that a sequence of about 15 images yields a number of velocity values which contribute towards the statistical reliability of the method. The measurements are exported via Direct Data Exchange (DDE) into a spreadsheet program for further analysis. As with these high imaging frequencies the particle positions change only very little in subsequent frames, it is easy to recognise the same particle and determine its position in subsequent images. It should be noted, that other techniques, such as Particle Image Velocimetry would not be capable of working in these densely concentrated particle assemblies.

As an example of single particle tracking by image analysis, Figure 5 shows the trajectories of two particles leaving the dense phase and being entrained into the jet. The time between markers is 74 μs. Figure 6 shows the particle velocity as a function of height. For the sign convention, right and upward directions are taken as positive. The right-hand jet boundary is indicated roughly by the shaded area in Figure 5. Clearly, the particle marked A is moving horizontally into the jet, while the particle marked B is already well in the centre of the jet and accelerating vertically.

### 3.2 Solids concentration and jet angle measurements

Measurements of the jet angle can be combined with measurements of the solids concentration profile in the jet. In an image of the jet, the jet is more dilute and appears darker than the dense phase surrounding it. Therefore, the jet has a lower grey scale value. The relationship between voidage and grey scale was explored by [9], who showed that a linear correlation exists between the grey scale and the solids concentration in a fluidised particle assembly. Heffels et al. [10] have studied multiple backscatter from particle suspensions. They present line profiles of backscatter intensity vs backscatter angle for various solids concentrations of 100 μm glass beads in water. Figure 7 presents the backscatter intensity at 0° backscatter angle, *i.e.* direct backscatter, extracted from the data of Heffels et al. [10]. The trend line in Figure 7 results from a linear regression. A linear relationship between the
backscatter intensity and the solids concentration appears to exist. Solids concentrations in a fluidised bed jet fall in approximately the same range. Therefore, it is assumed in the following that the backscatter intensity, measured in terms of the pixel grey scale value, is directly proportional to the solids concentration in the jet.

Measurements of solids concentration have thus been carried out as follows. Using a SONY DXC 930 3CCD video camera and a JVC HZ-H713 macro lens, an image of the background, i.e. the still bed without fluidisation or jet gas flow, is taken as a reference. This is shown in Figure 8a. The grey scale luminance values in these images ranges from 0 (black) to 255 (white). The solids concentration of the bed at incipient fluidisation is known to be approximately 0.6. A second image of the bed with an active jet and background fluidisation (Figure 8b) is then subtracted from the reference image. This results in a virtually entirely black image, with the exception of the jet zone and any bubbles present, which show illuminated (Figure 8c). Some parts of the jet core will be empty, and the grey scale value in the original jet image (Figure 8b) will be minimum there. The new value of the grey scale in the pixels at this position in the image after the subtraction (Figure 8c) will be used as a maximum, to determine the range of grey scale values present. This range is then re-mapped into a solids concentration range by scaling the grey scales inversely with the solids concentration. The maximum grey scale value will be related to zero solids concentration, and the minimum, i.e. zero, to the bulk solids concentration at 0.6.

Figure 9 presents a typical three-dimensional diagram of the solids concentration of particles in the lower section of a fluidised bed jet. The divergent shape of the jet, as it extends vertically upward, can be easily discerned. The jet angle can be obtained from this profile by determining the intersection of the profile with a plane of constant solids concentration. The level of this constant solids concentration is important and cannot be chosen arbitrarily, since the slope at which the solids concentration decays into the dilute jet core varies along the jet height. This is related to the formation of a boundary layer, within which the solids concentration gradually changes from the bulk dense phase to the dilute jet core, along the height of the jet. The boundary layer is virtually absent at the orifice nozzle, where the decay along the horizontal axis is sharp. The boundary layer gradually builds up along the jet height, resulting in a moderate decay at the top of
the profile. It was found however, that the jet angle profile was fairly insensitive to the level of solids concentration for measurements in different operating conditions, if it was put at approximately 0.45.

It is well-known that moving objects may draw streaklines on photographic plates. This will contribute to the grey scale values in the image. Even though the exposure time on the video camera could be reduced down to 100 μs, it was necessary to verify the technique. This was done with an Imacon 790 image converter camera\(^7\). This camera can acquire 6-16 frames onto a single Polaroid instant photographic plate at up to 20 million fps, depending on the acquisition module chosen. The camera operates fully electronically. For a few conditions, experiments done with the SONY video camera were repeated with the Imacon 790, using the 100,000 fps acquisition module and exposure times of 2 μs. An example of an image taken with the Imacon 790 is presented in Figure 10. The results of this verificatory experiment are presented below.

4. **RESULTS AND DISCUSSION**

The measurements of solids concentrations and jet angles of fluidised bed jets of catalyst particles, obtained with the SONY video camera are shown in Figures 11 and 12. It is clearly shown in Figure 12, that for various orifice sizes, the jet angle values behave like tangents to a single hyperbolic curve. The overlap of jet angle values in certain velocity ranges reflects the effect of the nozzle size on the jet angle. For the purpose of verification, the measurements with the 1.0 mm orifice nozzle have been repeated with the Imacon 790 image converter. The results are shown in Figure 13, together with the values measured by the SONY camera. The measurement methods appear to match each other closely. The measured values of solids concentration and jet angle have been used as input for numerical simulations of jet hydrodynamics. Figure 14 shows the resulting comparison of particle velocities in a jet with a 1.0 mm nozzle. It is shown that the predictions from the model and the measured values lie closely together.

\(^7\)Manufactured by Hadlands Photonics Ltd, Bovington, UK
The measurements of particle velocities of salt particles just above the nozzle exit and at a height of 47 mm above the nozzle exit are shown in Figures 15 and 16, respectively. Comparing Figures 15 and 16, it is shown that for a large orifice-to-particle size ratio, in the low orifice gas velocity range, particles accelerate significantly, with velocities tripling along the jet height. On the other hand, for small orifice-to-particle size ratios, i.e. close to unity, in the higher orifice gas velocity range the particles do not accelerate at all. Figure 17 shows the jet core above a nozzle of 5.0 mm at an orifice gas velocity of 13.4 m s\(^{-1}\), and above a nozzle of 0.5 mm at an orifice gas velocity of 30 m s\(^{-1}\), both with 0.5 mm common salt particles. It appears that when the particle size is comparable to the orifice size, the jet core does not open up. This prevents the particles from flowing in and accelerating freely upward [11].

There are some implications resulting from these observations. In mechanical testing of powder friability, it is common practice to subject a sample of powder to fluidisation with a single jet at a fixed velocity for a standard period of time. Usually a standard geometry is chosen, such as the set-up devised by Forsythe and Hertwig [12], which involves a nozzle of 0.398 mm. This is in fact adopted as an ISO standard (ISO 5937, formerly BS 5688), often used for the assessment of the friability of catalyst and detergent powders. The nozzle size of 0.398 mm is generally close to the particle size, and the particles are thus likely to be subjected to rapid dense phase shearing flow, as shown in Figure 17b. The results of these tests are then used to predict and control powder behaviour in commercial units, which are equipped with orifices of several centimetres in diameter, much larger than the particle size. Here, the particles are likely to accelerate freely and undergo high velocity impacts. Boerefijn and Ghadiri [13] have argued on the basis of these considerations that the hydrodynamics of the two cases are by no means comparable and that the extent and mechanism of breakage in the commercial unit cannot be inferred from the test results. This casts doubts on the applicability of the test results. Also, these considerations indicate a limit on the applicability of the modelling approach, described above. A realistic model of jet hydrodynamics is required for the small ratios of orifice-to-particle size. In the case of a dilute jet core,
however, the approach has been validated, and the predictions from the hydrodynamic model verified [14, 15].

5. CONCLUSIONS

High speed digital video image analysis techniques have been developed for measurements of particle velocities and solids concentration in fluidised bed jets. The method of measurement of solids concentration has been verified using a high speed photographic camera. The measurements have verified predictions from a hydrodynamic model of fluidised bed jets, and have contributed towards the validation of the modelling approach to attrition of fluidised particles. A limit in the applicability of the model has been identified, where the similarity between single impact breakage and jet impact breakage does not appear to hold. The observations of particle flow patterns in the jet have also led to the conclusion that results of particle friability tests, performed in fluidised beds with fixed geometries, may not be generally valid for other geometries, because of a difference in the breakage mechanism. Thus, the use of high speed photography and video image analysis has proven to be essential to the development of our understanding of particle flow behaviour.

ACKNOWLEDGMENT

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REFERENCES


Footnotes:

1. Sartopure is a registered trademark of Sartorius AG, Goettingen, Germany
2. Imaging Technology Inc., Bedford MA, USA
3. FOR-A UK Ltd, London, UK
4. Optimas is a registered trademark of Optimas Corp., Seattle, USA
5. Windows is a registered trademark of Microsoft Corp., Seattle, USA
6. National Instruments Corp., Austin TX, USA
7. Manufactured by Hadlands Photonics Ltd, Bovington, UK
Figures:

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Figure 3. Experimental set-up
Figure 4. Example of single particle tracking
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   b. The same particle in a consecutive image, 74 μs later.
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Figure 17. Comparison of wide, dilute jet core (left) and narrow, dense jet core (right).
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(a) (b) (c)

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a. Common salt particle in fluidised bed jet flow.
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Figure 7. Backscatter intensity as a function of solids concentration (based on Heffels et al., 1996).
(a) Background reference image of still bed.

(b) Fluidised bed with central jet and bubbles.

(c) Result of subtraction of 8b from 8a. The jet and bubbles show illuminated.

Figure 8. Image processing for measurement of jet solids concentration profile illuminated.
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Appendix H

The Effect of Orifice Size on the Breakage of Fluid Cracking Catalyst Particles in Fluidised Bed Jets
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THE EFFECT OF ORIFICE SIZE ON THE BREAKAGE OF FLUID CRACKING CATALYST PARTICLES IN FLUIDISED BED JETS

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An experimental study of the dependence of the attrition rate of fresh and used fluid cracking catalyst on the hydrodynamics of fluidised bed jets has shown that there is a significant effect of the orifice-to-particle size ratio on the mechanism of attrition. For large ratios, a dilute jet core exists and the breakage mechanism is similar to that occurring in a normal single particle impact. For small ratios, particles are subjected to a rapid shearing flow, and surface abrasion is predominant for fresh FCC, and also occurs to a lesser extent for used FCC. A previously developed model of attrition in the jetting region is used to predict the dependence of attrition on the orifice size. For used FCC, which fails in a mode similar to that envisaged in the model, the model predictions compare well to the experimental values, but for fresh FCC, as it is prone to surface abrasion, the model does not adequately take account of the different breakage mechanism.

Keywords: attrition, fluidisation, jets, fluid catalytic cracking, orifice, abrasion, distributor

INTRODUCTION

Fluid catalytic cracking (FCC) is an important operation in the oil refining industry, where fluidised bed reactor technology is used to convert heavy oil fractions into lighter products such as transportation fuels. The catalyst used in the FCC process is produced in the form of fine powder of particle size usually below 180 \( \mu \text{m} \) by means of spray drying\(^1\). An FCC unit consists of a reactor and a regenerator, sometimes both fluidised beds. The catalyst deactivates rapidly, because of coke deposition.

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Consequently, high recirculation rates of around 50 tonnes per minute are necessary to regenerate the catalyst by burning the coke in the regenerator, which at the same time provides the heat required for the endothermic catalytic reaction. Because of the large scale of solids transport and fluidisation at high velocities (up to about 50 m s\(^{-1}\)) attrition of particles can be a serious problem, giving rise to the elutriation of fines from the unit\(^2\). Consequently, this can result in a loss of fines, a few tonnes per day for a typical FCC unit, leading to increased operating costs. Thus the mitigation of attrition of FCC powder is of great significance.

Ghadiri and Boerefijn\(^3\) present a compilation of models of attrition in fluidised beds available in the literature. There appears to be little reliable information on the dependence of the attrition propensity on the properties of individual particles, such as the particle size, shape, porosity, surface topography, and mechanical properties and on the operating parameters, such as distributor orifice size, free area and the gas superficial velocities. Only a few previous investigations have so far taken the orifice size into account\(^4-7\). All investigations express the attrition dependence on the orifice size in a power law expression with power indices ranging from 0.4 to 2, without clear indications as to what causes this variation.

In many investigations of the attrition propensity of FCC particles, the method of Forsythe and Hertwig\(^8\) - hereafter referred to as F&H - has been adopted either in its original or modified form\(^9,10\). This method employs a submerged air jet at a very high orifice gas velocity of 300 m s\(^{-1}\) as the source of attrition. The ISO standard test (ISO 5937, formerly BS 5688, part 26) of attrition propensity of sodium perborate powders has been derived from the F&H test.

Boerefijn and Ghadiri\(^11,12\) report a distinct effect of the orifice-to-particle-size ratio, \(d_{or}/d_p\), on the structure of a jet in a fluidised bed based on studies of the hydrodynamics of jets in a two-dimensional fluidised bed of FCC particles. The results of these studies suggest that for low \(d_{or}/d_p\) (below 10), the jet does not have a dilute core, causing high velocity shear in the jet region. For large \(d_{or}/d_p\) (above 10), particles can become entrained into a virtually empty jet core region, where they accelerate and finally impact on the dense phase at the top of the jet. A dimensional analysis of the parameters controlling the jet angle, recently carried out by Vaccaro\(^13\), also indicates that for \(d_{or}/d_p\) around 10 a transition in jet hydrodynamics
takes place. Breakage mechanisms involved with these two cases of entrainment are very different. In the former case, surface abrasion, and in the latter, fracture, similar to impact damage, would be predominant, depending on the mechanical properties of the particles. This suggests that attrition tests are geometry dependent and that the results may not be easily extrapolated to different geometries. The F&H test operates with a $d_{or}/d_p$ of around 4, \textit{i.e.} below 10, whereas in FCC units this ratio is larger than 1000, well clear of the transition and well into the dilute core regime. Therefore the breakage mechanisms are not necessarily comparable. Following an extensive experimental investigation of the breakage mechanism of fresh and used FCC catalyst, Boerefijn \textit{et al.}\textsuperscript{14} conclude that the orifice-to-particle size ratio interacts strongly with the propensity to surface abrasion and this may distort a comparison of two materials based on the F&H test. In the following, the effect of orifice-to-particle size on the extent and mechanism of breakage will be quantified and illustrated by means of varying the size of the orifice for a single particle size and orifice gas velocity. The ratio of orifice-to-particle size is varied around the presumed critical ratio of around $10^{11.13}$.

Ghadiri and Boerefijn\textsuperscript{3} describe a way of predicting the performance of a powder in a unit operation from measurements of the single particle attrition propensity. This method has been applied successfully to describe quantitatively the attrition of used FCC in a large-scale fluidised bed\textsuperscript{15}. This model is based on numerical simulations of the particle flow patterns in the jet combined with data from single particle impact tests. In the following, this model will be used to predict the attrition dependence on the orifice size and model predictions will be compared to experimental results in order to further elucidate the mechanism of breakage in fluidised bed jets.

**EXPERIMENTAL**

Experiments consisted of fluidisation of a batch of approximately 50 g of a commercial FCC powder with a single jet of ambient air, without background fluidisation. The duration of the test is one hour, following Forsythe and Hertwig\textsuperscript{8}. A schematic of the F&H test set-up is shown in Fig. 1. Experimental Set-up
Figure 1. It consists of a QVF glass column of 1 m height and 25.4 mm ID. The bottom is equipped with a single jet nozzle set in a brass plate. Four different nozzles were used in the present experiments, of 0.175, 0.375, 0.75 and 1.525 mm ID. The 0.175 mm nozzle was obtained by mounting a calibrated needle into a larger hole, drilled in the brass plate. The orifice gas velocity was kept constant at about 47 m s\(^{-1}\). This velocity is close to the velocities, commonly appearing in jets in FCC units and was also conveniently pitched for the wide range of flow rates necessary for the different orifices. The exact conditions are shown in Table 1. At the top of the column, the air flows out through a Sartorius 0.45 \(\mu\)m absolute retention filter to capture any fines.

The samples of fresh and used FCC particles were obtained by sieving a single BS410 sieve cut of 90-106 \(\mu\)m. The orifice-to-particle size ratios thus varied from 1.5-16. After testing, a sieve of two BS 410 standard sieve sizes below the lower limit of the original sieve cut size, \(i.e.\) 63 \(\mu\)m, was used to separate the debris from the surviving mother particles. This debris criterion is the same as that employed in the single particle impact test of these particles, as this will be used later in the model predictions. A previous study of impact attrition has shown that this criterion is insensitive to the breakage product size distribution, if the breakage product is much smaller than the original particles\(^{16}\). Material was collected both from the bed and from the filter. The extent of attrition is expressed as the fractional mass loss, \(i.e.\) the mass of debris divided by the mass of the original sample, which is approximated by the sum of mother particles and debris:

\[
R = \frac{M_d}{M_m + M_d}
\]  

(1)

Figures 2a and 2b show the fractional mass loss versus \(d_{or}/d_p\) for fresh and used FCC, respectively. The attrition propensity clearly increases monotonously with the

<table>
<thead>
<tr>
<th>Table 1. Experimental conditions</th>
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<tbody>
<tr>
<td>(d_{or})</td>
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<tr>
<td>(mm)</td>
</tr>
<tr>
<td>0.175</td>
</tr>
<tr>
<td>0.375</td>
</tr>
<tr>
<td>0.750</td>
</tr>
<tr>
<td>1.525</td>
</tr>
</tbody>
</table>
Figure 2a. Fractional mass loss of fresh FCC orifice size, and a comparison of the two materials shows that the fresh FCC attrites up to 25 times more than the used FCC. The solid lines in Figs 2a and 2b have been obtained by power law regression and have power indices of 1.63 and 0.63, respectively.

Scanning electron microscopy has been used to observe the pattern of damage and hence to diagnose the mechanism of breakage. Figures 3a and 3b show scanning electron micrographs of the original fresh and used FCC particles. Both materials have a ginger-root shape, with burs, protrusions on the surface, caused presumably by secondary agglomeration during spray drying. Clearly, the fresh FCC has a high surface roughness, and the used FCC has a very smooth surface.

Figures 4a and 4b show examples of debris of fresh FCC from a test with orifices of 0.175 and 1.525 mm, respectively. With the small orifice, a large quantity of fines is generated, and fragments are absent. With the large orifice, some large fragments are formed in the breakage process, but they are covered in small fines from an abrasion process, as is exemplified in Figure 4b.

Figures 5a and 5b show examples of debris of used FCC from a test with orifices of 0.175 and 1.525 mm respectively. With the small orifice, some small fines are generated, but with the large orifice, the debris consists mainly of a large quantity of fragments.
COMPARISON TO MODEL PREDICTION

The model approach has been described more extensively by Ghadiri and Boerefijn\textsuperscript{3}, and will only be summarised here. The mechanism of attrition is considered to be due to the entrainment of particles into a dilute jet core, followed by the acceleration of the particles, and their impact on the dense phase at the top of the jet. Intense interparticle collisions in this region cause attrition, at a rate that can be estimated
from impacts of single particles on a rigid target, and at impact velocities, corresponding to the maximum particle velocity $u_p$ in the jet. The single particle impact data are incorporated in a model of the hydrodynamics of the jet in order to describe the dependence of the fractional mass loss in the jet on the orifice size. A power law dependence of the form $R \propto d_{or}^n$ is considered, where $R$ is the fractional mass loss, $d_{or}$ the orifice size, and $n$ is the power index which is predicted by the model. We have previously reported the dependence of single particle impact attrition on the impact velocity, also in the form of power law indices\textsuperscript{17}. A hydrodynamic model of flow patterns in a jet in a fluidised bed, developed by Massimilla and co-workers\textsuperscript{18} has been employed to predict the dependence of the solids entrainment rate $W_s$ and the particle velocity $u_p$ on the orifice size $d_{or}$. The solids entrainment rate is further normalised by dividing $W_s$ by the gas mass flow rate $W_g$. We can now correlate the jet attrition rate to the orifice size:

$$R \propto \left( \frac{W_s}{W_g} \right) R_i \propto d_{or}^k u_p^m \propto d_{or}^k (d_{or})^m \propto d_{or}^n$$  \hspace{1cm} (2)

where $n = k + l m$ and $R_i$ is the fractional mass loss found in single particle impact tests, defined as in Equation 1. The power indices $k$ and $l$ are obtained from hydrodynamic modelling and $m$ from single particle impact tests. The modelling requires input of a number of parameters that describe the geometry of the jet. The values of these parameters are shown in Table 2 for the given materials and conditions. Values of the jet angle and the initial particle velocity and solids concentration have been chosen following direct measurements in a two-dimensional fluidised bed\textsuperscript{12}.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
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<tbody>
<tr>
<td>Jet angle</td>
<td>7.5°</td>
</tr>
<tr>
<td>Relative boundary layer thickness</td>
<td>0.1</td>
</tr>
<tr>
<td>Initial solids concentration</td>
<td>0.05</td>
</tr>
<tr>
<td>Fresh FCC density</td>
<td>800 kg m$^3$</td>
</tr>
<tr>
<td>Used FCC density</td>
<td>1400 kg m$^3$</td>
</tr>
</tbody>
</table>
Particle velocities, calculated from the model of Massimilla and co-workers\textsuperscript{18} range up to 10 m s\textsuperscript{-1}. Table 3 shows the resulting power indices from modelling and includes the power indices of the single impact attrition propensity reported previously\textsuperscript{17}. The power indices $n$ from the model and the experiment are very close to each other for the used FCC. For all experiments with fresh FCC, on the other hand, the experimental value is much higher than predicted by the model.

**DISCUSSION**

The significance of the F&H test for the assessment of the attrition propensity of a material cannot easily be underestimated, especially in the oil and detergent manufacturing industry. Boerefijn et al.\textsuperscript{14} have argued recently, however, that materials which are prone to surface abrasion cannot be assessed using this method, as the results cannot be extrapolated to different test geometries, such as prevail in industrial size units. The present data in many ways confirm this.

Boerefijn and Ghadiri\textsuperscript{11} observed that for small $d_o/d_p$, a clear jet core does not exist at the nozzle exit, so that particles in this area are subjected to rapid shearing flow. For large $d_o/d_p$, they observed a clear jet core, in which particles could accelerate freely right from the nozzle exit. After the first impact on the bulk at the top of the jet, the particles had lost most of their momentum and the secondary impacts took place at far lower velocities.

Figure 3a shows that the original material has a high surface roughness and this results in the abrasion of a large quantity of fine debris in the case of the small orifice (Figure 4a). In the case of the large orifice (Fig. 4b), the larger dilute area in the jet core, as observed by Boerefijn and Ghadiri\textsuperscript{11}, allows some fragmentation of the particles, similar to single particle impact. Even here, however, a large amount of fine debris is created, covering the larger fragments entirely, possibly resulting from secondary impacts. With fresh FCC surface abrasion is predominant regardless of the jet hydrodynamics. The figures presented here are intended to illustrate the point, but the observations described are based on SEM of a large number of particles.

Used FCC has gone through several cycles of reaction and regeneration in an FCC unit and has lost its surface roughness completely (Figure 3b). Some burs still exist, but not to the extent of fresh FCC. Some sintering of the particle surface at the high temperatures prevailing in the industrial unit will also have contributed to the
attrition resistance. In the case of the small orifice, the debris consists of some small fines of a few micron in size, as shown in Figure 5a. In the case of a large orifice, the debris consists of large fragments. This indicates that the rapid shearing flow in the case of small orifice-to-particle size ratios does cause some surface abrasion in the case of more resistant particles, but in the case of high orifice-to-particle size ratios the mechanism is clearly due to particle collision.

The comparison with the model prediction further confirms this. Fresh FCC abrades in a way which is quite different from that envisaged by the model, which uses the single particle impact data. It is, therefore, not surprising to see that the model produces a very poor prediction of the dependence of attrition propensity of this material on $d_{or}$. The same was the case with the dependence on $u_{or}$, reported earlier\textsuperscript{14}. Another contributing factor to this may have been that the abrasion may also have taken place in the bubbling bulk of the bed and this is not taken into account in the model. The prediction of the model for used FCC on the other hand produces a far better comparison with the experimental data. The contribution of surface to attrition in the region of small orifice-to-particle size ratios may be held accountable, however, for the existing deviation between $n_{exp}$ and $n_{mod}$.

In the case of materials which are prone to surface abrasion, the use of the single impact data may not be appropriate, as an impact at a normal angle may not adequately represent the breakage mechanism. The performance of the model may be improved by using data from tests with repeated impacts and impacts at different impact angle, where sliding impacts may cause surface abrasion.

Ghadiri et al.\textsuperscript{7} have reported values of $n = 0.6-0.76$ for another type of used FCC, measured with experiments in a different apparatus. The present values of $n$ are very close to this and this indicates further the usefulness of the present modelling approach to predict the behaviour of materials in different geometries.

CONCLUSIONS
The high propensity of fresh FCC to surface abrasion makes it highly susceptible to the aggravated flow conditions in the rapid shearing flow for small $d_{or}/d_p$, resulting in a far higher attrition rate than used FCC. For the latter material, the jet attrition model adequately describes the dependence of attrition on the orifice size.
Table 3. Attrition power indices.

<table>
<thead>
<tr>
<th>Material</th>
<th>k</th>
<th>l</th>
<th>m</th>
<th>n (mod.)</th>
<th>n (exp.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fresh FCC</td>
<td>-0.28</td>
<td>0.53</td>
<td>1.79</td>
<td>0.68</td>
<td>1.63</td>
</tr>
<tr>
<td>Used FCC</td>
<td>-0.27</td>
<td>0.55</td>
<td>2.00</td>
<td>0.83</td>
<td>0.63</td>
</tr>
</tbody>
</table>

The present results suggest strongly that results from laboratory standard tests cannot easily be extrapolated to different geometries and hydrodynamic conditions. The use of the present model, incorporating the physics of the appropriate single particle attrition test, whether single or repeated, normal or angled impact, or using a different shearing device, presents a suitable tool for predicting the mechanical behaviour of materials in different conditions.

ACKNOWLEDGMENTS

One of the authors (Miss S.-H. Zhang) is financially supported by the Han Suyin Trust Fund for Scientific Exchange, for which she is very grateful. This work is part of an on-going research programme on the mitigation of attrition in FCC units, supported by the EPSRC and the University of Surrey. The authors express their gratitude to Shell Research and Technology Centre, Amsterdam, for supplying the catalyst.

NOTATION

- $d_{or}$: Orifice Size (m)
- $d_p$: Particle Size (m)
- $R$: Fluidised Bed Jet Attrition (-/-)
- $R_i$: Single Particle Impact Attrition (-/-)
- $k$: Attrition Power Index (dependence on Solids Entrainment Rate)
- $l$: Attrition Power Index (dependence on Jet Particle Velocity)
- $m$: Attrition Power Index (dependence on Single Particle Impact Velocity)
- $M_d$: Mass of Debris (kg)
- $M_m$: Mass of Surviving Mother Particles (kg)
- $n$: Overall Attrition Power Index (equals $k + l m$)
- $u_{or}$: Orifice Gas Velocity (m s$^{-1}$)
- $u_p$: Particle Velocity (m s$^{-1}$)
\[ W_g \quad \text{Gas Mass Flow Rate (kg s}^{-1}\text{)} \]

\[ W_s \quad \text{Solids Entrainment Rate (kg s}^{-1}\text{)} \]

REFERENCES


Appendix I

Interactive Processes of Sorbent Attrition and Chemical Reaction During Fluidised Bed Sulphurisation

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INTERACTIVE PROCESSES OF SORBENT ATTRITION AND CHEMICAL REACTION DURING FLUIDIZED BED DESULPHURIZATION

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Attrition of two different limestones after calcination and sulphation in fluidized beds has been studied by a combination of experimental techniques. The aim is to shed light on the interactive processes of sorbent attrition and the change of mechanical and morphological properties associated with the progress of chemical reactions. Two experimental techniques have been used to characterize different breakage mechanisms occurring in different sections of industrial fluidized bed reactors. Abrasive attrition is characterized in situ by collection of elutriated fines as they are generated during fluidized bed calcination and sulphation of limestone. To this end a bench-scale apparatus was operated batchwise at conditions typical of atmospheric fluidized bed combustion. Fragmentation under high velocity impact conditions is studied ex situ by means of single particle impact tests on pre-conditioned samples at room temperature. Microscopic analysis of the particles has been used to reveal differences in topography, indicating different process history. The comparison of results from the two experimental techniques has indicated clearly the effects of process history on particle structure and strength. Simultaneous calcination and sulphation leads to reduced impact damage, when compared to subsequent calcination and sulphation.

Keywords: attrition, fluidized bed, limestone, calcination, sulphation
INTRODUCTION
Reduction of sulphur oxides emissions in the fluidized bed combustion is frequently accomplished by *in situ* injection of sorbents like limestone. In the fluidized bed, at typical atmospheric combustion conditions, sorbent particles undergo simultaneous chemical reactions (first calcination and then sulphation) and attrition. Sorbent attrition affects the performance of the combustor, by influencing the size distribution of the sorbent particles and the elutriation of fines in the flue gases, and the extent of conversion of the limestone by exposing unreacted limestone.

A relatively large number of studies have addressed attrition of limestone in fluidized beds. Most of the effort was directed towards the characterization of attrition of lime in an inert atmosphere. The relevance of the simultaneous occurrence of calcination and sulphation reactions to limestone attrition was first recognized by Chandran and Duqum\(^1\) and Couturier *et al.*\(^2\). Recently, Scala *et al.*\(^3\) and Di Benedetto and Salatino\(^4\) have highlighted the interactions between attrition and chemical reactions, through the changes of particle morphology and texture along with conversion. In particular, it was demonstrated that neglecting the strengthening effect of the particle surface upon sulphation may lead to a consistent overprediction of fines generation by attrition.

In the light of the different breakage mechanisms and of the typical size of generated fragments, Scala *et al.*\(^3\) provided a classification of sorbent attrition phenomena during fluidized bed calcination and sulphation. *Primary fragmentation* occurs immediately after the injection of the particles in the bed, as a consequence of thermal stresses due to rapid heating of the particles and of internal overpressure due to carbon dioxide emission. This may results into the generation of coarse as well as fine fragments. During their processing in the fluidized bed, sorbent particles are also subject to *attrition by abrasion* and *secondary fragmentation*. The extent to which these processes contribute to attrition depends on the mechanical stresses due to collisions with other particles or with the internals of the reactor. According to Blinichev *et al.*\(^5\) these phenomena can be classified on the basis of the typical size of the generated fragments: attrition by abrasion generates fines which quickly elutriate; secondary fragmentation generates coarser fragments, which cannot easily elutriate. Attrition by abrasion depends on the resistance of the bed particles to surface wear. Secondary fragmentation should be related to the resistance of the particles to
impacts against walls and internals of the bed or in the jetting region of fluidized beds.

The aim of this work is to investigate the influence of the two chemical reactions calcination and sulphation on the attrition of two limestones of different origins. In particular abrasive attrition is characterized \( \textit{in situ} \) by collection of elutriated fines as they are generated during fluidized bed calcination and sulphation. A bench-scale apparatus operated batchwise has been used to this end. Attrition under high velocity impact conditions is studied \( \textit{ex situ} \) by means of single particle impact tests at room temperature. The two techniques are complementary, since they provide a measure of the propensity to attrition in either dense phase, bubbling fluidized beds (\( \textit{in situ} \)) or lean phase, fast fluidized beds (\( \textit{ex situ} \)).

EXPERIMENTAL

\textbf{In situ experiments}

The experiments were carried out in an electrically heated stainless steel atmospheric bubbling fluidized bed reactor, 40 mm ID and 1 m high (Fig. 1). The gas distributor was a perforated plate with 55 holes 0.5 mm in diameter in a triangular pitch. The bed material consisted of mixtures of sorbent and sand. Two different sorbents were tested, namely an Italian limestone (Massicci) and a Swedish porous limestone (Ignaberga). Batches of limestone and sand were sieved in the nominal size range 0.425-0.6 mm.

Attrition was quantified during three different reaction stages: calcination (C), subsequent sulphation (S) and simultaneous calcination and sulphation (CS). The calcination experiments were carried out in air. The S and CS experiments were carried out by using a mixture of nitrogen, oxygen (8.5 %Vol.) and SO\(_2\) (1800 ppm) as fluidizing gas. The fluidizing gas superficial velocity was always kept at 0.8 m s\(^{-1}\). A limited amount (20 g) of fresh limestone (in C and CS experiments) or of precalcined limestone (in S experiments) was charged into the bed of sand (150 g) operated at 850 °C. The attrition rate of bed material was determined by measuring the amount of fines elutriated from the bed. These were collected alternately in sequences of filters for limited periods of time. The difference between the weights of the filters before and after operation, divided by the time interval of collection,
was taken as the average elutriation rate relative to that interval. Further details of the experimental apparatus and procedure are reported elsewhere.\(^3\)

Primary fragmentation tests for the two fresh limestones were performed on a heated grid\(^7\) in order to simulate the high heating rate experienced by the sorbent particles when injected into the hot bed, in conditions where abrasion of the particles is not present. The experiments were carried out by heating a sample of particles, placed on the grid by an electric current up to a temperature of 1300 K at a heating rate of about 10000 K s\(^{-1}\).

**Ex situ experiments**

*Ex situ* experiments consisted of single particle impact tests, using the air eductor device shown in Figure 2. Samples of approximately 5 g are prepared by gently sieving a BS 410 cut of 0.6-0.85 mm. The materials used are samples of Massicci and Ignaberga in the fresh (F) state, as well as from the *in situ* process stages, i.e. calcined (C), calcined and subsequently sulphated (S) and simultaneously calcined and sulphated (CS). Particles are fed in a single array into the funnel-shaped inlet section at the top of the eductor and subsequently accelerated on the air flow provided by a compressor. Upon exiting the eductor tube the particles impact on a rigid target plate, made of sapphire. The particle velocity is controlled by regulating the air flow and measured by means of a timer which is triggered by two diodes at the end of the eductor tube. The entraining air leaves the collection chamber through a brass porous plate, covered by a Whatman class 1 paper filter, retaining the impact
products down to 4 μm. The collection chamber is kept at a low vacuum (2 mbar). After impact, the material is collected and the debris is separated from the surviving mother particles using a sieve with a size of two BS 410 sieve sizes below the lower size limit of the original sieve cut, in this case 425 μm. Attrition is then quantified as the fractional mass loss, i.e. the ratio of the mass of debris to the mass of the feed material. The latter is approximated by the sum of mass of debris, \( M_d \), and mother particles, \( M_m \), collected after impact:

\[
R = \frac{M_d}{M_m + M_d}
\]  

This expression is found to be insensitive to the effects of moisture take-up. The procedure of collection of impact products and sieving will inevitably cause material losses, which may arise from the loss of the surviving mother particles as well as from fine debris. Equation 1 does not take account of losses, but it can be shown that it is between the lower and upper limit of fractional mass loss, as defined by Papadopoulos and Ghadiri.

EXPERIMENTAL RESULTS AND DISCUSSION

In situ experiments

Figure 3 shows the rate of sorbent fines elutriation \( E \) measured during the calcination and the subsequent sulphation of fresh samples of the two limestones. During calcination, \( E \) decreases with time until a steady state value \( (E_{ss}) \) is reached. Two mechanisms may be invoked to explain the observed trend: i) rounding off of the particle surface, and ii) fines generation by primary fragmentation due to thermal shock and calcination. In order to assess the extent to which the latter mechanism contributes to attrition, separate experiments have been carried out, in which fresh limestone samples were exposed to fast heating to 1300 K in a heated grid reactor. It appeared that Massicci limestone does not fragment upon heating, whereas Ignaberga undergoes primary fragmentation, with extensive generation of fragments. The mechanism through which primary fragmentation enhances attrition is twofold: on the one hand it may directly lead to the generation of elutriable fines; on the other it gives rise to relatively coarse fragments whose shape is highly angular, and therefore more prone to surface wear. The elutriation of directly generated fines, from the bed should take place over a short time scale, comparable with the
relaxation time of the thermal stresses and calcination, as the latter process could induce mechanical stresses due to generation of CO₂ gas. This time is of the order of a few minutes or less depending on the operating conditions. The generation of fines by attrition of particles or fragments, generated by primary fragmentation, should instead take place over a time scale longer than that of calcination. The combined analysis of primary fragmentation experiments from the heated grid and of *in situ* experiments suggests that rounding off is the dominant breaking mechanism active during calcination of *Massicci* (as reported earlier by Scala *et al.*). For *Ignaberga*, instead, primary fragmentation and rounding off are both active at the same time. At the steady state of the calcination stage, *E*₂₀°C is slightly larger for *Massicci* than for *Ignaberga*. This can be partly attributed to the larger mass of *Massicci* limestone remaining in the bed (and exposed to abrasive attrition) after calcination. After normalisation of *E* with respect to the actual mass of limestone, taking into account the conversion from fresh to calcined lime, the difference in the steady state attrition rate at the end of the calcination stage between the two limestones becomes negligible. It is important to note here that the process of attrition in the bed is dominated by abrasion of the lime particles by the sand particles, which are much harder than both types of lime.

Upon sulphation of precalcined lime the attrition rate of both types of limestones decreases dramatically until a new steady-state value (*E*ₚₛ) is reached (Fig. 3). As pointed out by Scala *et al.*, this trend can be related to surface strengthening of the particle as calcium oxide reacts to form calcium sulphate. The
attrition rate of Massicci is slightly larger than that of Ignaberga throughout sulphation.

Figure 4 compares the attrition rates of limestone samples during calcination of fresh limestone (C), during sulphation of precalcined lime (S) and during simultaneous calcination and sulphation of fresh limestone (CS). The feed particles of the S stage have been preconditioned in the calcination stage, hence the difference in the rate of elutriation of C and S, due to the survival of the stronger particles. As expected, for both limestones the profile corresponding to simultaneous calcination and sulphation conditions lies in between those relative to separate calcination and sulphation. The long term attrition rate $E_{oo}$ is comparable under sulphation and simultaneous calcination and sulphation conditions, and much lower than that measured in the early stage of sorbent sulphation. This is presumably because the external surface properties are the same for both samples.

Ex situ experiments

The analysis is carried out by comparing the attrition propensity of the following samples: as-received "fresh" limestone (F), samples calcined for 20 min (C), samples sulphated for about 1 hour after pre-calcination for 20 min (S) and samples simultaneously calcined and sulphated for about 1 hour (CS). Results are reported as fractional mass losses on impact versus impact velocity in Figs 5 and 6 for Massicci and Ignaberga, respectively. For the series of Massicci samples, it appears that the fresh sorbent is the one with the lowest propensity to impact damage. The C, S and CS samples have comparable fractional losses upon impact, showing a transition velocity at about 12-17 m s$^{-1}$, where the slope of the curves change. In the case of
semi-brittle failure, this velocity may be related to a change in impact breakage mechanism from the so-called *chipping* to *fragmentation*. In the case of chipping, surface chips are formed, much finer than the mother particles, whereas in the case of fragmentation, fragments of dimensions similar to the mother particles are formed. *Ignaberga* F, C and S samples, on the other hand, (Fig 6) show comparable strengths, while the simultaneously calcined and sulphated (CS) sample gives rise to a far larger fractional loss than the others. Within the tested velocity range, *Ignaberga* C and S appear to have similar transition velocities as the processed *Massicci* samples. However, the *Ignaberga* F and CS do not exhibit a clear transition velocity within the tested velocity range. With the exception of *Ignaberga* CS, the extent of impact breakage in the chipping regime is not greatly different between the various samples from different processes. On the whole, the *Massicci* limestone is less prone to fragmentation than the *Ignaberga*.

Microscopic analysis of the particles has been carried out for each sample after impact. The *Massicci* F has a pitted surface, covered with thin patches of a thin layer of fines. The debris consists of some very small fines and larger fragments. The *Massicci* C has a more rugged look, and the debris contains less fines than the fresh material. The *Massicci* S has a shiny surface, and the debris contains almost no fines whatsoever. The shine of the surface indicates that the surface porosity is very low, and may have been caused by a melting process, or sintering during the formation of CaSO₄. The CS material has an appearance similar to that of the sulphated material: the surface is smoother and has a slight shine. The debris contains some clear surface cracks. The outer surface has a different structure from the open crack faces, which have a far higher roughness. There is hardly any fine

![Figure 7. Ignaberga S mother particles.](image1)

![Figure 8. Ignaberga CS mother particles.](image2)
debris. *Ignaberga* F has a glassy look under a fine layer of fines on the surface. The debris of this material contains a large amount of fines below 100 μm. *Ignaberga* C exhibits qualitatively the same surface roughness and texture as the fresh material. The amount of fines in the debris is smaller, and the debris consists generally of larger fragments. *Ignaberga* S has a shiny surface, with a larger roughness scale than the previous two materials. The protuberances are much more rounded and much larger than in the other two materials. The debris of this material consists of highly angular fragments with only very little fine debris. *Ignaberga* CS has a comparably shiny surface and rough texture as the S sample. Some CS particles exhibit clear crack patterns on the surface, indicative of core-shell behaviour: the shell has started to crack, but the core is too tough for the crack to propagate.

On the whole, analysis of the results of impact tests for the various samples suggests that the resistance to fragmentation, expressed as fractional loss upon impact, decreases in the sequence F >> S > C > CS for Massicci, F ≈ S > C >> CS for *Ignaberga*. These sequences may be interpreted simply by considering the difference in the mechanical properties of the pure components: calcium oxide appears to be less resistant than both the fresh calcite and the sulphate. The unexpected result is that the CS sample is more prone (far more in the case of *Ignaberga*) to impact damage than the others. In particular it is noteworthy that, from the standpoint of impact resistance, simultaneous calcination and sulphation of limestone is not at all equivalent to the path consisting of calcination of limestone followed by sulphation of lime.

One tentative explanation of this observation might be that two very different structures evolve: in the CS experiments, the sulphation occurs at the same time as rounding off of the particles, whereas in the S experiments it acts on material which has survived the process of calcination. It is possible that sulphation strengthens the particle surface in the CS experiment, limiting further rounding off of the calcined particles, and therefore the elutriation rate is lower than in the C experiments. In the light of this argument, the enhanced tendency of CS samples to undergo impact fracture (but not surface wear) might be related to the presence of micro-flaws at the surface which could facilitate brittle failure, leading to progressive fragmentation. Moreover, the simultaneous occurrence of calcination and of surface sulphation in CS experiments might leave behind residual stresses after cooling to ambient
conditions that enhance structural failure upon impact loading. Micrographs in Figs. 7 and 8 compare the visual appearance of mother particles of Ignaberga S and CS after impact. In Figure 8, one CS particle clearly exhibits fissures and cracks on the surface. These cracks were observed in a number of particles in the CS sample, and none were found in the S sample. It is likely that a combination of increased surface hardness and of residual stresses comes into play in determining the much larger propensity of CS samples to impact damage at ambient conditions. On the other hand, the sulphated materials would have a much lower hardness and larger toughness at high temperature because of the proximity to the glass transition temperature of CaSO₄.

The comparison of results of single particle impact tests with those of in situ experiments suggests that different mechanisms of particle breakage prevail in the two cases. In particular, it is likely that the length scales of zones affected by mechanical stresses are different. A thin cortical region of the particle is likely to be the location of failure when surface wear dominates attrition. Surface properties should control the extent of attrition in this case. Under impact loading, depending on the velocity, mechanical stresses may extend far deeper within the particle. A combination of surface (hardness and roughness) and bulk properties should come into play in this case.

CONCLUSIONS
During in situ experiments, differences in elutriation rates from the fluidized bed reactor have indicated changes in structural and surface properties as a function of the progress of the calcination and sulphation stages. In these experiments, surface wear is the dominant breakage mechanism, whereas in the ex situ impact experiments particles are more prone to fragmentation.

After an initial period, the Ignaberga limestone exhibits a resistance to surface abrasion comparable to that of Massicci limestone in in situ experiments, while in impact tests Ignaberga shows a generally lower resistance to breakage. The influence of the internal structure on the impact resistance is emphasized by the different attrition rates of subsequently and simultaneously processed materials (S, CS), especially Ignaberga.

In in situ experiments, the attrition tendency is controlled by the competitive processes of particle rounding off and sulphation. In particular, the fully sulphated
material has a negligible propensity to surface wear. Simultaneously calcined and sulphated Ignaberga exhibit a dramatic increase of the impact attrition propensity when compared with fresh, calcined and subsequently calcined and sulphated limestones.

The two experimental techniques employed have demonstrated clearly the different breakage mechanisms occurring in industrial fluidized bed units and are complementary to each other as they focus on different areas within a fluidized bed unit. The combination of the two techniques provides a powerful tool for the assessment of particle behaviour in full-scale reactors. Even though impact testing at ambient conditions may not reflect the particle breakage behaviour at high temperature, it still provides a clear indication of structural differences between samples which have been sulphated in different ways. Further work is required to establish the relative importance of the two complementary mechanisms investigated here as a function of the geometry and the operating conditions of industrial units.

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NOTATION

\[ E \] Fines elutriation rate (kg s\(^{-1}\))
\[ E_{\text{calc}} \] Calcination stage steady state fines elutriation rate (kg s\(^{-1}\))
\[ E_{\text{sulph}} \] Sulphation stage steady state fines elutriation rate (kg s\(^{-1}\))
\[ M_d \] Mass of debris (kg)
\[ M_m \] Mass of surviving mother particles (kg)
\[ R \] Fractional mass loss on impact (-/-)

REFERENCES


Appendix J

Time-resolved Voidage Profiles at the Bed Surface and in the Freeboard of Shallow Fluidised Beds
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TIME-RESOLVED VOIDAGE PROFILES AT THE BED SURFACE AND IN THE FREEBOARD OF SHALLOW FLUIDISED BEDS

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Shallow fluidised beds are commonly used for processing of fine and light powders. A distinct feature of such systems is the lively agitation of the entire bed. Bubble structures are not very well-defined and the bed behaves in a chaotic manner. Consequently, the bed free surface cannot be clearly identified. In order to design and operate these beds effectively, characterisation of the motion of the bed surface and of the solids concentration in the freeboard is essential. In the present study, a two-dimensional fluidised bed of sodium carbonate has been used to monitor the agitation of the bed using a digital video imaging system. Voidage profiles have been measured for two bed aspect ratios and different starting particle size distributions using a digital image analysis technique. Significant effects of the initial particle size distribution and bed height on the elutriate size distribution and on the voidage profile in the freeboard have been quantified.

Keywords: fluidisation, shallow fluidised beds, elutriation, voidage profiles, freeboard, transport disengagement height
INTRODUCTION

Shallow fluidised beds of light and fine powders, with bed height-to-width aspect ratios below and around unity, have the awkward feature of lacking a clear distinction between the dense phase and the freeboard. Very little is known of the dependence of the characteristic voidage gradient between dense phase and freeboard on the operating conditions and particle properties. The aim of the present study is the characterisation of the solids concentration gradient at the bed surface and in the freeboard of a shallow fluidised bed of sodium carbonate.

The significance of the entrainment of particles into the freeboard can be manifold. For example, in fluidised combustors entrainment of carbon particles affects combustion efficiency and formation of SO$_2$, NO$_x$, and other pollutant gases$^1$. Furthermore, entrainment is of great importance for the design of bed internals, such as heat exchangers and spray nozzles$^2$. A comprehensive review of studies of entrainment has recently been published by Milioli and Foster$^3$.

The stability of fluidised beds and the origin and structure of bubbles has been the subject of continuous investigations since the beginning of fluidisation research$^4$. It is well-known that light and fine powders of Geldart classification type A may be affected by interparticle forces. The effect of interparticle forces on the fluctuations of the local voidage is still very poorly understood$^5$. In the dense phase of a bubbling bed, stabilisation of the voidage structure by interparticle forces may lead to delayed onset of bubbling with an increase in the minimum bubbling velocity. In the freeboard of the fluidised bed, above the dense phase, the phenomenon of cluster formation may result. The analysis of the entrainment of solids into the freeboard is usually based on either (i) dimensional analysis, or (ii) the mechanism of ejection of particles by either the wake or the nose of erupting bubbles$^6$. Either approach assumes the existence of a contained fluidised bed with a distinct bed surface, which separates the bulk from the freeboard.

A small number of studies have addressed the transport disengagement height (TDH). McNeil et al.$^7$ used a shallow two-dimensional fluidised bed with a perforated plate and a sparger, with a bed height-to-width ratio of 2.0. They define the TDH as the height above the bed surface at which the entrainment rate becomes constant and relate the TDH to the diameter of bubbles bursting at the surface. Bubbles are generated both at the grid holes and at the sparger jets, located just
above the grid, which makes the interpretation of the results in terms of separate
effects of the grid and sparger difficult. Baron et al.\textsuperscript{8} specifically quantified the
effect of varying bed height between 0.75 and 1.5 times the bed diameter on the rate
of entrainment of a Geldart type B powder. They found that, assuming that the
entrainment is related to the coalescence of bubbles near the bed surface, an increase
in bed height will result in larger bubbles which are less likely to coalesce and
therefore entrainment should decrease with bed height.

The effect of particle size distribution on the entrainment of particles has
been the source for some controversy in the literature. Adding fines to the sample
may result in a decrease of entrainment, as the fines may adhere to coarse particles
and cause them to stay in the bed\textsuperscript{9}. On the other hand, in turbulent fluidised beds,
there is evidence that adding fines may cause the carry-over of coarse particles with
free fall velocities larger than the fluidising gas velocity\textsuperscript{10}. In the present study,
materials from different stages of a granulation process are used and the freeboard
solids concentration profiles are presented as a function of particle size distribution
and of bed height.

The techniques employed in the past to study the entrainment of particles in
the freeboard vary from capacitance probes to laser Doppler anemometry and
particle image velocimetry. These techniques are suitable for dilute suspensions, \textit{i.e.}
the freeboard. From the work of Milioli and Foster\textsuperscript{3} it is clear, however, that there is
a need to characterise the gradient of solids concentration at the bed surface. The
present study employs a novel measurement technique to characterise the time-
dependent local voidage in the area around the bed surface of the fluidised bed.
There is some evidence in the literature that the reflected light intensity within the
solids concentration range of 0.1-0.6 can be correlated linearly against particle
concentration\textsuperscript{11,12}. Although further work is required to substantiate this
relationship, this concept is used here to measure voidage profiles also in the dense
phase of a two-dimensional bed.
EXPERIMENTAL
The experimental set-up is shown in Figure 1. It consists of a 1.2 m high two-dimensional fluidised bed with a surface area of 200 x 12 mm$^2$. The bed was fluidised with ambient air at a moderate level of about 4 times the minimum fluidisation velocity. The distributor consists of a brass porous plate, the front and rear walls are made of optical glass panes of 10 mm thickness and the side walls are made of brass strips to eliminate electrostatic charges. At the bed exit, the solids are separated initially by a horizontal cyclone for preseparation of particles down to 90 μm, then a high efficiency Stairmand$^{13}$ cyclone and finally by a Sartorius Sartopure pleated paper filter cartridge with an absolute retention down to 0.2 μm.

Measurements were carried out over a fixed time of 5 minutes. After the experiment, the bed was opened at the top and the cyclones and filters were cleared from dust. This product, the elutriate, was collected in separate vials. The bed inventory was taken out at the side of the distributor and kept in a container and finally the bed was cleaned completely.

During the experiment, video recordings were made through the optical glass walls of the movement of the bed. Usually two camera positions were covered, one for the lower part of the bed and one for the higher part of the bed, in order to obtain high resolution images of the entire expansion range of the bed.

Images of the still bed were recorded for calibration purposes, as will be explained later. Then video recordings were made at various heights of the bed in fluidised condition. The test materials and conditions are listed in table 1. The fluidisation behaviour at a bed height-to-width ratio of 0.3 was investigated for three samples of the test material obtained from different stages of the granulation process of sodium carbonate, fresh, semi- and fully granulated at a fixed value of $u_s/u_{mf}$. For the fresh material, an aspect ratio of 1.0 was also investigated. Values of $u_{mf}$ have been calculated using the Ergun equation, using a particle density based on the bulk
density at a voidage of 0.35, and a minimum fluidization bed voidage of 0.4. The calculated values of $u_{mf}$ matched closely with the point at which incipient fluidisation occurred. Before and after processing, the particle size distribution was determined using a Sympatec Helos particle sizer.

A SONY DXC930P 3CCD camera was used for video recordings in digital R-G-B, with a resolution of 768 x 572 pixels at an exposure time of 100 $\mu$s. The images were firstly recorded onto VHS tape and then grabbed from a video recorder using an in-line FOR-A FA-310P time base corrector onto an IC-PCI digital frame grabber in a PC. The images were then analysed using Optimas 6.1 software. The images were firstly converted into 8-bits grey scale format, then an image of the black background (Figure 2a) was subtracted from the fluidised bed images (Figure 2b), after which the grey scale range was mapped onto a range which had as a maximum the grey scale value of the fixed bed. The latter was obtained from the calibration image.

The grey scale value of the fixed bed corresponds to a voidage of 0.4, or a solids concentration, $\gamma$, of 0.6. The minimum value of 0 was set to the empty bed condition of voidage equal to 1.0 and solids concentration of 0.0. The grey scale

<table>
<thead>
<tr>
<th>Material</th>
<th>$\rho_b$ (kg m$^{-3}$)</th>
<th>$\rho_p$ (kg m$^{-3}$)</th>
<th>$d_{10}$ (μm)</th>
<th>$d_{50}$ (μm)</th>
<th>Bed Fill Level (mm)</th>
<th>$u_{mf}$ (m s$^{-1}$)</th>
<th>$u/u_{mf}$ (-/-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fresh</td>
<td>675</td>
<td>1929</td>
<td>10</td>
<td>96</td>
<td>60</td>
<td>0.0068</td>
<td>3.94</td>
</tr>
<tr>
<td>Fresh</td>
<td>675</td>
<td>1929</td>
<td>10</td>
<td>96</td>
<td>200</td>
<td>0.0068</td>
<td>3.94</td>
</tr>
<tr>
<td>Semi-gran.</td>
<td>542</td>
<td>1549</td>
<td>19</td>
<td>119</td>
<td>60</td>
<td>0.0084</td>
<td>3.97</td>
</tr>
<tr>
<td>Fully-gran.</td>
<td>455</td>
<td>1300</td>
<td>45</td>
<td>180</td>
<td>60</td>
<td>0.0161</td>
<td>4.06</td>
</tr>
</tbody>
</table>

Figure 2a. Still bed.  
Figure 2b. Fluidised bed.
values in the fluidised bed images were re-scaled linearly with solids concentration levels:

\[ \gamma = 0.6 \left( \frac{\text{greyscale}}{255} \right) \]  \( \text{(1)} \)

The images were spatially calibrated using the ruler which was recorded on the image. A total of ten lines were drawn on the region of interest, vertically equidistant at 10 mm interleave (in calibrated units) and the line luminance was obtained for a total of 100 images for all of these lines. This provided a measure of the grey scale along a number of levels above the bed surface, in a certain interval of time. The images were acquired consecutively at a fixed interval of 0.04 seconds and provided a temporal average. The values of these line luminances were represented by 64 samples, taken equidistantly along the line. The line had a length of 20 cm in calibrated units, spanning across the bed. For these ten lines, the 64 samples of luminance value were extracted and exported to a spreadsheet program for further data processing. The line luminance values were averaged in time and space. The spatial average refers to the average of 64 luminance values along a single line, i.e., at a single vertical position. The luminance was then translated into a solids fraction using equation 1. Figure 3 shows a typical example of the voidage at a point on a line in time, obtained by this procedure. It should be noted here that the frequency of acquisition, 25 Hz, may not be sufficient to capture all the dynamic phenomena, but in the near future the use of high-speed cameras (up to 40 kHz) is envisaged to provide measurements of time-resolved voidage characteristics. This should enable accurate cross-correlation in space and time to characterise bubbling phenomena.

RESULTS
The four experimental cases, listed in Table 1 give rise to an apparently very different fluidisation behaviour, as illustrated in Figures 4 to 7. Figure 4 shows a typical instant of a fluidised bed of 60 mm fresh sodium carbonate. It is in a vigourously bubbling state, and clear bubble structures and a bed surface cannot be
identified. Figure 5 shows the same material in a 200 mm bed at the same superficial gas velocity. In this case, bubbles develop and the bed surface is very distinct. Figure 6 shows very much the same features as Figure 4, even though the material is semi-granulated. Figure 7 shows a bed of the same height, but with fully-granulated material. In this case, the bed exhibits lively agitation, but the bed is more contained and bubbles are more clearly defined. In all cases, the emerging of bubbles at the surface was quite uniform along the width of the bed and no distinct down-flow zone at the walls was observed. These visual observations can be easily quantified using the grey scale as a measure of solids concentration, as described above. Figure 8 shows the voidage profiles of 60 mm beds of fresh, semi-granulated and fully-granulated sodium carbonate. In this figure, $h^*$ denotes the fractional bed expansion above the static bed height, $h_0$: 

$$h^* = \frac{(h - h_0)}{h_0}$$

(2)
The profiles have a number of different features. The fresh material shows a fairly monotonous rise of voidage against height, until about twice the bed height above the static height \( h^* = 1.5 \), where the slope changes slightly. The semi-granulated material starts at a higher bed voidage than the fresh material, but soon joins the fresh material data. The fully-granulated material starts off at the same bed voidage as the fresh material, but the entrainment is less, so the gradient is sharper and reaches unity already within three static bed levels distance from the bed surface.

Figure 9 compares the voidage profiles of 60 mm and 200 mm fresh material beds as a function of height above the static bed height. Clearly, the 200 mm bed expands further than the lower bed, as shown by the higher starting value of the voidage profile. Furthermore, the gradient covers a shorter vertical distance: the rise from 0.6 to 0.9 takes place within a distance of 60 mm, whereas for the lower bed, in absolute measures, this takes 1.5 times that distance. When expressed as a function of \( h^* \) this effect would be even more pronounced.

Error bars in Figures 8 and 9 show the standard deviation of values at a certain vertical position. This is the deviation along 64 equidistant sampling points along a line spanning the bed width for a sequence of images in time that has been analysed at each condition. They should be interpreted as a measure of the fluctuation amplitude of voidage in time. For example in Figure 8, the bars of semi-granulated material have a small magnitude until \( h^* = 1.5 \), which indicates that the bed has expanded to that level and only above that level does the voidage fluctuate significantly. In contrast, large voidage fluctuations are already observed within the bed of fully-granulated material in the expanded state. From this, one may infer a strong effect of voidage fluctuations on overall voidage: when small fluctuations occur, the bed can expand to far greater height. Small fluctuations may be related to a larger degree of overall expansion and to moderate bubbling, whereas large fluctuations are related to vigorous bubbling and agitation. Even when the average voidage of fresh and semi-granulated materials are similar above \( h^* = 1.5 \), the fluctuations are much larger for semi-granulated than for fresh sodium carbonate. An increased bed height causes a slight increase in voidage fluctuations, which suggests a clearer definition of the bubble structure.
Figure 8. Voidage profiles of 60 mm beds

Figure 9. Voidage profiles of 60 (□) and 200 (▲) mm

□: fresh; ▲: semi-granulated; ●: fully-granulated.

Fresh sodium carbonate

Figure 10 shows the particle size distributions of the material elutriated from the bed and recovered in the second cyclone and the pleated paper filter. Material from the first cyclone is recycled directly into the bed. The elutriate of fresh (60 mm bed) and semi-granulated have similar size distributions, the fully-granulated elutriate is much larger, and the fresh elutriate from the 200 mm deep bed is much smaller.

As shown in Figure 8, the semi-granulated material exhibits far smaller fluctuations below $h^* = 1.5$ than the other two materials. Power spectral analysis of time signals as shown in Figure 2 may elucidate more precisely whether this is caused by the formation of small bubbles or by a small degree of uniform macro-scale bed motion. A tentative explanation for the difference in expansion between fresh and semi-granulated material may be found in their particle size distributions. It can be observed clearly in Figure 10 that the elutriate of the 60 mm fresh bed contains far more fines below 10 μm than the semi-granulated material. This

Figure 10. Size distributions of elutriate

×: 60 mm Fresh; ▲: 60 mm Semi-granulated;
■: 60 mm Fully-granulated; □: 200 mm Fresh
reflects the difference in the original size distributions between fresh and semi­
granulated material, evident by comparison of the $d_{10}$ values in Table 1. It is more
than likely that the reduced fines content in the semi-granulated material causes the
increased expansion of the semi-granulated bed and the smaller fluctuations of local
voidage when compared to fresh sodium carbonate.

The original size of the fully-granulated material is of course far larger than
the fresh and semi-granulated, so there is little surprise in the fact that the elutriate is
largest of all. The most extraordinary feature here is the large difference in elutriate
between the 60 mm and the 200 mm deep beds of fresh sodium carbonate.

DISCUSSION

The present study can only be seen as a probe into the issue of the effects of particle
size and bed height on the quality of fluidisation and the voidage profile in the
freeboard, in as far as this can be distinguished from the dense phase. A number of
phenomena have been observed and reported which are all strongly related. Beds of
60 mm depth of fresh and fully-granulated sodium carbonate exhibit very similar
expansion, with similar bed surface voidage gradients, but the semi-granular, which
contains less fines, expands more homogeneously. The 200 mm deep bed of fresh
material has a better defined bubble structure and bed surface, as does the fully­
granulated material.

The main difference between the present materials is presented in the particle
size distributions. The fresh material contains more fines than the semi-granulated
material, and the fully-granulated material has a more uniform size distribution at
larger $d_{50}$. This may be used to explain the differences in the expansion behaviour,
shown in Figs 4-9. Further work is required to establish whether it is the fines
content, e.g. expressed in the $d_{10}$, or the average size, $d_{50}$, which dominates this
phenomenon. The particle size distributions shown in Figure 10 are essentially
monomodal. This feature needs to be ascertained by an alternative technique in the
future as it may be an artifact of the measurement technique rather than a true
property of the particle size distributions.

The present investigation has also indicated a strong relation between the bed
height and the elutriation mechanism. The work of Baron et al. does not provide
any basis for comparison as only the amount of elutriation was quantified, not the
size distribution. Further work is required to establish a wider basis of correlation of elutriate size and mass with starting material size distribution and bed height.

CONCLUSIONS

On the basis of an optical measurement technique, voidage profiles of materials of different size distribution have been quantified. There is some indication of an effect of fines content on the expansion behaviour of the fluidised suspension. With a decreased fines content, the suspension becomes more homogeneous, as the local voidage fluctuations were observed to decrease. The effect of a more monodisperse distribution is reduced by a shift in the average size with the progress of granulation, as this enhances voidage fluctuations. The effect of an increase in bed height on the voidage profiles appears to be two-fold: the bed expands to a higher level, but less homogeneously, as a clearer bubble structure develops. The present measurement technique appears to be a powerful non-intrusive tool for quantifying the quality of fluidisation.

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NOTATION

\(d_{10}\)  size of 10% of the cumulative sample undersize distribution (m)
\(d_{50}\)  size of 50% of the cumulative sample undersize distribution (m)
\(h\)  vertical height (m)
\(h_0\)  static bed height (m)
\(h^*\)  relative height above static bed height (-/-)
\(u_{mf}\)  superficial gas velocity at incipient fluidisation (m s\(^{-1}\))
\(u_s\)  superficial gas velocity (m s\(^{-1}\))
\(\gamma\)  solids concentration (-/-)
\(\rho_b\)  bulk density (kg m\(^{-3}\))
\(\rho_p\)  particle density (kg m\(^{-3}\))
REFERENCES


Appendix K

Analysis of ISO Fluidised Bed Jet Test for Attrition of Fluid Cracking Catalyst Particles
accepted for Fluidization IX, 1998
ANALYSIS OF ISO FLUIDISED BED JET TEST FOR ATTRITION OF FLUID CRACKING CATALYST PARTICLES

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ABSTRACT

A previously developed model of attrition in fluidised bed jets is used to analyse the standard Forsythe and Hertwig (1949) single jet fluidised bed attrition test of Fluid Cracking Catalyst particles. The model is based on the single particle impact test and hydrodynamic modelling of the flow patterns in the fluidised bed jet. Experimental results show that the attrition rate for fresh FCC particles is at least four times higher than that of used FCC particles. The attrition model successfully predicts the dependence of the attrition of used FCC on the orifice gas velocity. The model does not satisfactorily predict the dependence of attrition of fresh FCC in the conditions of the Forsythe and Hertwig test as the breakage mechanism appears to differ from that occurring in single impact. Limits to the use of the Forsythe and Hertwig (1949) test with respect to materials and test geometry are indicated.

INTRODUCTION

Fluid catalytic cracking (FCC) is an important operation in the oil refining industry, where fluidised bed reactor technology is used to convert heavy oil fractions into lighter products such as transportation fuels. The catalyst used in the FCC process is produced in the form of fine powder of particle size usually below 180 μm by means of spray drying (1). An FCC unit consists of a reactor and a regenerator, both

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fluidised beds. The catalyst deactivates rapidly, because of coke deposition. Consequently, high recirculation rates of around 50 tonnes per minute are necessary to regenerate the catalyst by burning the coke in the regenerator, which at the same time provides the heat required for the endothermic catalytic reaction. Because of the large scale of solids transport and fluidisation at high velocities (up to about 50 m s\(^{-1}\)) attrition of particles can be a serious problem, giving rise to the elutriation of fines from the unit \(2\). Consequently, this can result in a loss of fines, a few tonnes per day for a typical FCC unit, leading to increased operating costs. Thus the mitigation of attrition of FCC powder is of great significance.

Ghadiri and Boerefijn \(3\) present a compilation of models of attrition in fluidised beds available in the literature. There appears to be little reliable information on the dependence of the attrition propensity on the properties of individual particles, such as the particle size, shape, porosity, surface topography, and mechanical properties and on the operating parameters, such as distributor orifice size, free area and the gas superficial velocities.

In many investigations of the attrition propensity of FCC particles, the method of Forsythe and Hertwig \(4\) - hereafter referred to as F&H - has been adopted either in its original or modified form \(5, 6\). This method employs a submerged air jet at a very high orifice gas velocity of 300 m s\(^{-1}\) as the source of attrition. The ISO standard test (ISO 5937, formerly BS 5688, part 26) of attrition propensity of sodium perborate powders has been derived from the F&H test.

Boerefijn and Ghadiri \(7, 8\) report a distinct effect of the orifice-to-particle-size ratio, \(d_{or}/d_p\), on the structure of a jet in a fluidised bed based on studies of the hydrodynamics of jets in a two-dimensional fluidised bed of FCC particles. The results of these studies suggest that for low \(d_{or}/d_p\) (below 10), the jet does not have a dilute core, causing high velocity shear in the jet region. For large \(d_{or}/d_p\) (above 10), particles can become entrained into a virtually empty jet core region, where they accelerate and finally impact on the dense phase at the top of the jet. A dimensional analysis of the parameters controlling the jet angle, recently carried out by Vaccaro \(9\), also indicates that for \(d_{or}/d_p\) around 10 a transition in jet hydrodynamics takes place. Breakage mechanisms involved with these two cases of entrainment are very different. In the former case, surface abrasion, and in the latter, fracture, similar to
impact damage, would be predominant, depending on the mechanical properties of the particles. This suggests that attrition tests are geometry dependent and that the results may not be easily extrapolated to different geometries. The F&H test operates with a \( d_{or}/d_p \) of around 4, \textit{i.e.} below 10, whereas in FCC units this ratio is larger than 1000, well clear of the transition and well into the dilute core regime. Therefore the breakage mechanisms are not necessarily comparable. In the following, the extent and mechanism of breakage in the F&H test will be elucidated by experiments with fresh and regenerated FCC particles in three different size ranges.

Ghadiri and Boerefijn (3) describe a way of predicting the performance of a powder in a unit operation from measurements of the single particle attrition propensity. This method has been applied successfully to describe quantitatively the attrition of used FCC in a large-scale fluidised bed (10). In the following, this method will be employed to assess the usefulness of the F&H test to industrial operations. The F&H test results are compared to predictions from the model, which is based on numerical simulations of flow patterns of particles in jets, and on single particle impact tests.

**EXPERIMENTAL**

Experiments consisted of fluidisation of a batch of approximately 50g of a commercial FCC powder with a single jet of ambient air, without background fluidisation. The duration of the test is one hour, following Forsythe & Hertwig (4). A schematic of the F&H test set-up is shown in Fig. 1. It consists of a QVF glass column of 1 m height and 25.4 mm ID. The bottom is equipped with a single jet nozzle of 0.375 mm. Orifice gas velocities ranged from 50 to 300 m s\(^{-1}\), the upper limit being the normal test velocity of the F&H test. At the top of the column, the air flows through a Sartorius 0.45 \( \mu \)m absolute retention filter to capture any entrained fines.

The samples of fresh and used FCC particles were obtained by sieving single BS 410 sieve cuts, in the ranges 75-90 \( \mu \)m, 90-106 \( \mu \)m and 106-125 \( \mu \)m. The orifice-to-particle-size ratio is therefore between 3 and 4. After testing, a sieve of two BS 410
standard sieve sizes below the lower limit of the original sieve cut size was used to separate the debris from the surviving mother particles, *i.e.* 53, 63 and 75 μm respectively. This debris separation criterion is the same as that employed in the single particle impact test of these materials, as this will be used later in the model predictions. This criterion, albeit arbitrary, is based on the chipping mechanism of particulate solids, where the size distribution of the debris is much smaller than than the feed particle size. This has been described in more detail elsewhere (3, 11). Material was collected both from the bed and from the filter. The extent of attrition is expressed as the fractional mass loss, *i.e.* the mass of debris divided by the mass of the original sample, which is approximated by the sum of mother particles and debris:

\[
R = \frac{M_d}{M_m + M_d}
\]  

(1)

Figure 2 shows the fractional mass loss of all samples. It is clearly shown that the attrition increases with increasing orifice gas velocity for all samples. Also, for a given material, the extent of attrition increases with particle size. The attrition propensity of fresh FCC powder is at least 4 times higher than used FCC powder for all orifice velocities. In single particle impact tests of the same material, the fresh FCC powder also had a 4-5 times higher attrition rate than the used FCC powder (12).

Scanning electron microscopy is used to observe the pattern of damage and hence to diagnose the mechanism of breakage. Figures 3a and 3b show scanning electron micrographs of the fresh and used FCC particles, both in the size range of 75-90 μm. Both materials have a ginger-
root shape, with burs, protrusions on the surface, caused presumably by secondary agglomeration during spray drying. Clearly, the fresh FCC has a high surface roughness, and the used FCC has a very smooth surface. Figures 4a and 4b show debris from an F&H test at $u_{or} = 200 \text{ m s}^{-1}$, for fresh and used FCC particles, respectively. Figure 4a shows a large amount of very fine debris of a few micron in size. The debris of fresh FCC consisted mainly of these fines, whereas the debris of used FCC contained also some large detached burs (Fig. 4b). The presence of a large amount of fines in the debris of the fresh FCC is probably related to the high surface roughness, when compared to used FCC. This indicates that with fresh FCC, a mechanism of surface abrasion prevails, whereas for used FCC the fracture of burs, protruding from the surface, is also important. Duo et al. (12) report that the latter breakage process, also called “necking” is the dominant failure mode in single impact breakage.

**COMPARISON TO MODEL PREDICTION**

The model approach is described more extensively by Ghadiri and Boerefijn (3) and
will only be summarised here. The mechanism of attrition is considered to be due to the entrainment of particles into a dilute jet core, followed by the acceleration of the particles, and their impact on the dense phase at the top of the jet. Intense interparticle collisions in this region cause attrition, at a rate that can be estimated from impacts of single particles on a rigid target, and at an impact velocity, corresponding to the maximum particle velocity \( u_p \) in the jet. The single particle impact data are incorporated in a model of the hydrodynamics of the jet in order to describe the dependence of the fractional mass loss in the jet on the orifice velocity. A power law dependence of the form \( R \propto u_{or}^n \) is considered, where \( R \) is the fractional mass loss, \( u_{or} \) is the orifice gas velocity and \( n \) is the power index which is predicted by the model. We have previously reported the dependence of single particle impact attrition on the impact velocity, also in the form of power law indices \( (12) \). A hydrodynamic model of flow patterns in a jet in a fluidised bed, developed by Massimilla and co-workers \( (13) \) has been employed to predict the dependence of the solids entrainment rate \( W_s \) and the particle velocity \( u_p \) on the orifice gas velocity \( u_{or} \).

We can now correlate the jet attrition rate to the orifice gas velocity:

\[
R \propto W_s \; R_i \propto u_{or}^k \; R_i \propto u_{or}^k \; u_p^m \propto u_{or}^k \left( u_{or}^l \right)^m \propto u_{or}^n
\]  

(2)

where \( n = k + l + m \), and \( R_i \) is the fractional mass loss found in single impact tests, defined as in Eq. 1. The power indices \( k \) and \( l \) are obtained from modelling and \( m \) from single impact tests. The modelling requires input of a number of parameters that describe the geometry of the jet. The values of these parameters are shown in Table 1 for the given materials. Values of the jet angle and the initial particle velocity and solids concentration have been chosen following direct measurements in a two-dimensional fluidised bed \( (8) \). Following these measurements, the jet angle has been varied inversely proportional to \( u_{or} \).

Particle velocities, calculated from the model of Massimilla and co-workers \( (13) \) range from 3 to 18 m s\(^{-1} \). Table 2 shows the resulting power indices from modelling and includes the power indices of the

<table>
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<th>Property</th>
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<td>Jet angle</td>
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<td>( u_{or} )</td>
<td>50-300 m s(^{-1} )</td>
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<tr>
<td>Relative boundary</td>
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single particle impact attrition propensity reported previously (12). Invariably, the power indices $n$ from model and F&H experiment are very close to each other for used FCC. For all samples of fresh FCC, on the other hand, the experimental values are much lower than predicted by the model: the deviation is never less than 25%.

**DISCUSSION**

The weaker particles in fresh FCC break down during the FCC operation at a much higher rate than the stronger particles, and therefore the used FCC particles will have a higher resistance to impact damage than fresh FCC particles. This is clear from the results shown in Fig. 2. Furthermore, the higher surface roughness and the existence of a large number of surface burs in fresh FCC, as compared to used FCC, would give rise to a far greater extent of impact attrition of the fresh FCC particles. Additionally, the sintering of the used FCC particles in the high temperature FCC process may reduce the attrition propensity. Irrespective of these differences, both fresh and used FCC appeared to comply very well with the model of Zhang and Ghadiri (14) for impact breakage, which predicts that $m$ should equal 2 on the basis of linear elastic fracture mechanics. Also, scanning electron microscopy of debris from impact tests for both fresh and used FCC showed no distinct difference in the features of the debris: it consisted of detached, or "necked" burs. Therefore, in single particle impact tests, the fresh and used samples exhibit the same type of failure, but to a different extent.

The experimental value of $n$ for used FCC is close to the values reported in the literature for used FCC. Zenz and Kelleher (15) report a value of 2.5, Werther and Xi (16) a value of 3. For mullite aluminosilicates, Kono (17) reports a value of 2, but this was obtained in a bed with a conical distributor section, which promotes
the entrainment of solids into the jet.

All previous investigations base the quantitative analysis of attrition on the amount of fines elutriated from the bed and found in the exit collector (3). In their study of the hydrodynamic influences of particle breakage in fluidized beds, Ayazi Shamlo et al. (18) report that at times no less than 28% of fines remained in the bed. In the present set-up, the amount of fines recovered from the bed varied between 30 and 90 percent of the total amount of fines recovered, depending on the operating conditions.

Measurement of hydrodynamic parameters, such as particle velocities, jet angles, solids concentration, and jet boundary layer thickness, in two-dimensional fluidised beds have shown that the model predictions of particle velocity and solids entrainment rate compare well with experimental values (8). It can therefore be assumed that the model predictions for the present hydrodynamic cases are valid. The model does not predict well the attrition dependence of the fresh FCC on the orifice velocity. This can be clearly observed from a comparison of the values of the power index $n$ for the model and experiments with fresh FCC powder in Table 2. There are two coupled processes which are considered responsible for this discrepancy. For small orifice-to-particle-size ratios, a rapid shearing flow occurs in the jet, as has been visualised using high-speed video recordings in two-dimensional fluidised beds (7). This process particularly affects particles with high surface roughness, like fresh FCC. In this case, surface abrasion may predominate, as confirmed by the presence of a large quantity of fine debris of a few micron in size (Fig. 4a). Consequently, single particle impact at a normal angle may not adequately represent the damage process for this type of material in the jet region. It may be more appropriate to use data from tests with repeated impacts and impacts at a different impact angle, where sliding impacts can cause surface abrasion. For particles with friable surfaces, the contribution to attrition from the bubbling dense phase of the fluidised bed may also be significant. This aspect has not been accounted for in the present analysis.

When the predominant mechanism of failure is comparable to single impact attrition, the attrition model is very well capable of predicting the dependence of the attrition propensity on the orifice gas velocity. This has also been shown by Ghadiri
et al. (10) for a fluidised bed of used FCC with a large orifice-to-particle-size ratio. In conclusion, the present analysis shows that the F&H test is not universal for all types of powder, as its performance is affected by a combination of particle properties and the ratio of orifice to particle size.

CONCLUSIONS

Tests following the F&H method have shown that the attrition propensity of fresh FCC powder is at least 4 times higher than the equivalent used FCC powder. Fresh FCC powder appears to suffer mainly surface abrasion in the jet. This is related to the small orifice-to-particle-size ratio. Used FCC powder is less prone to surface abrasion and exhibits more fracture of protrusions, similar to single impact breakage.

The attrition model, based on hydrodynamic modelling of the jet and on data from single particle impact tests, is capable of predicting the dependence of the attrition rate on important operating parameters, such as the orifice gas velocity, when the breakage mechanism is comparable to the single impact breakage.

Laboratory scale tests like the F&H test are unsuitable for prediction of material behaviour in different geometries and large scale operations, as a different breakage mechanism may prevail. The concept of the present attrition model is useful for predicting the material behaviour in different geometries. For prediction of the behaviour of material which is prone to a different breakage mechanism from that occurring in single impact tests, the breakage of single particles should be quantified using an alternative test method, focusing on shear and sliding wear. The development of such a test is on-going (19).

Further work on the effect of the orifice-to-particle-size ratio is on-going, investigating different orifice sizes.

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**NOTATION**

- $k$ : Attrition Power Index (dependence on Solids Entrainment Rate)
- $l$ : Attrition Power Index (Dependence on Particle Velocity)
- $m$ : Attrition Power Index (Dependence on Particle Velocity)
- $M_d$ : Mass of Debris (kg)
- $M_m$ : Mass of Surviving Mother Particles (kg)
- $n$ : Overall Attrition Power Index (Equals $k + lm$)
- $R$ : Fluidised Bed Jet Attrition (-/-)
- $R_i$ : Single Particle Impact Attrition (-/-)
- $u_{or}$ : Orifice Gas Velocity (m s$^{-1}$)
- $u_p$ : Particle Velocity (m s$^{-1}$)
- $W_s$ : Solids Entrainment Rate (kg s$^{-1}$)

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