PROPAGATION OF ULTRASOUND IN INHOMOGENEOUS VISCOELASTIC MEDIA

by

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To those who were deprived of their right to education, particularly those of Latin America.
In this thesis an investigation was centered on models of inhomogeneous viscoelastic media, which consisted of glass beads (lead and soda glass) embedded in a silicone rubber matrix (KTV-602). The specimens were classified into groups according to the nominal size of the glass beads content (mean diameter ranging from 63.8 μm to 582 μm) in a range of concentrations (up to 5% volume). Ultrasonic velocity and attenuation as a function of frequency were measured by transmission techniques in the range of 0 to 10 MHz.

An extended investigation was centered on the effects of diffraction on velocity and attenuation measurements. The effects of diffraction in distilled water were found to be in good agreement with modified mathematical transducer models.

The measured velocity and attenuation in the specimens were compared with theoretical predictions based on the multiple scattering of waves formulation of Waterman & Truell (1961) in combination with the Kramers-Kronig relationship between absorption coefficient and phase-velocity dispersion in the constituent materials of the specimens. Good agreement was obtained between the experimental and the theoretical approaches used, which suggest their applicability for inhomogeneous viscoelastic materials. From the attenuation coefficient and velocity, the composition of materials can be determined. The attenuation coefficient was shown to be a more reliable acoustic parameter than velocity, although both are necessary for a complete characterization of composite materials.
I wish to express my gratitude to ... 

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CHAPTER 1

INTRODUCTION
CHAPTER 1

INTRODUCTION

Ultrasonic techniques are amongst the most promising and most rapidly advancing methods of diagnosis in industry, medicine and basic sciences. A number of different applications of ultrasonic techniques that have proved very valuable in basic research and characterisation of media can be found in application to gases, liquids and solids. Examples of these applications can be generally listed as: mixing ratio in composites and solutions, porosity, plastics manufacturing, mixtures of gases, preferred grain orientation, technological properties of metals, evaluation of concrete, determination of elastic constant of materials, diagnosis in medicine, discrimination between normal and pathological biological tissues, marine engineering, fishing technology, food technology, and many others. Ultrasound is also found in nature, for guidance and communication, used by bats, insects, birds, rodents and marine mammals.


Although a few of the widely used textbooks have been published over a decade ago, in recent years remarkable improvements have been
made in automation of the instrumentation, but the physical principles have remained the same.

Not surprisingly, the improvements in performance of equipment have been dedicated more to commercial imaging devices, where marketing competition is higher than to basic research in laboratories. These improvements of instrumentation will certainly continue to be made as physicists investigate and understand better the propagation of ultrasound and engineers apply more fully the principles of communication theory made possible by advances in integrated circuit technology.

The physical parameters which can give potential information for the characterization of a medium upon the incidence of ultrasonic energy are: wave propagation velocity, attenuation, absorption, scattering, and specific acoustic impedance.

When a wave travels through a real medium, its intensity is reduced as a function of distance. In practice, when one is measuring the attenuation of a beam of ultrasound in a medium, one is measuring the total attenuation arising from all the different mechanisms involved. These mechanisms may be: absorption, reflection, refraction, diffraction and scattering. The absorption process involves the conversion of the ultrasonic energy into some other form of energy (thermal), and is dependent on the nature of the medium. The process of reflection, refraction, diffraction, and scattering are simply that part of the wave that travels in other than the original direction of propagation. These losses are dependent on the physical and geometrical properties of the medium (or media) of propagation. Reflection and refraction occur at interfaces between regions with different acoustic impedances. Diffraction occurs at finite barriers that are interposed in the path of the beam. Scattering losses are
characteristic of the structure of materials, related to acoustic impedance mismatch and to the size of the scatterer compared to the wavelength. It may not be possible to devise an experiment that will isolate the attenuation due to just one of the above mechanisms. However, theoretical calculations based on some experimental observations may be considered to correct the experimental results to obtain the parameter aimed at.

In an attenuating medium with frequency dependent attenuation coefficient, the phenomenon known as dispersion of waves occurs, i.e., the phase velocity is frequency dependent. A particular medium may have a negligible frequency-dependent velocity if its attenuation (or absorption) is small. An example of such a medium is water at low megahertz frequencies. However, the density and the elastic constants of materials usually vary with temperature. Consequently, the velocity is also dependent on temperature. A group of waves may be formed by superimposing a number of waves of different frequencies. If the wave velocity is frequency dependent, the group velocity may also depend on frequency. An apparent change in the velocity of waves in a medium may also occur as a result of diffraction, as in the case of attenuation, and appropriate corrections are necessary.

Absolute measurements of velocity and attenuation are very difficult because of a number of influencing factors. Relative measurements are easier, for example: changes of the acoustic parameter during a given test, or a comparison between two specimens under the same testing conditions.

Ultrasonic diagnosis of materials is more valuable if proven theories of wave propagation are used in connection with the analysis of the results. It appears that the majority of the work published in the literature has been concerned with elastic and non-elastic
materials, and as far as the author is aware, the case of inhomogeneous viscoelastic media is still lacking knowledge. Examples of inhomogeneous viscoelastic media, either manufactured or found in nature, at normal conditions of temperature and pressure, are: soft solids, polymers, rubber-like composites, various geological materials, various biological materials, and biological tissue models. For instance, in medical applications of ultrasound, it is necessary to know how ultrasound is propagated through tissues, so that one can make better use of ultrasonic techniques for diagnosis. To achieve that knowledge, it is necessary first of all to acquire the knowledge of how ultrasound is propagated through inhomogeneous viscoelastic materials. For this, one needs to consider particular controlled systems or well defined models of inhomogeneous viscoelastic materials. The study of propagation of elastic waves through inhomogeneous viscoelastic materials is of particular importance to determine the effect that the inhomogeneities introduce to the material properties, or vice-versa. Most inhomogeneous materials can be classified as: laminate, fibrous and particulate.

The investigation presented in this thesis is concentrated on materials consisting of elastic spherical inclusions (the scatterers) embedded in a viscoelastic matrix. There are theories of ultrasonic wave propagation in inhomogeneous media in the literature, but the test of such theories in inhomogeneous viscoelastic media is lacking.

A variety of publications and investigations concerning wave propagation in inhomogeneous materials can be found in the literature. Recently, a major review (of about 300 scientific publications) on the subject was carried out by Anson & Chivers (1984a, 1984b). Among the scientific publications reviewed there were: reviews; single and multiple scattering theories; corrections; comparison with
experiments; long wavelength approximations; effective media approaches; thermal effects; and static and dynamic approaches. However, most of the recent theoretical investigations appear to concentrate on multiple scattering theories of waves.

For plane wave propagation in an infinite inhomogeneous system, the excess attenuation caused by the inhomogeneities can be written as
\[ \alpha_{\text{exc}} = \alpha_{\text{tot}} - \alpha_{\text{mat}} \]
where \( \alpha_{\text{tot}} \) denotes the total attenuation in the composite and \( \alpha_{\text{mat}} \) the absorption in the matrix material. This type of system is usually approximated mathematically by considering either single scattering or multiple scattering approaches. Simple single scattering approaches give information on attenuation but not on velocity. The only ways to obtain velocity from scattering properties are by considering either multiple scattering approaches (Epstein & Carhart, 1953; Waterman & Truell, 1961; Allegra & Hawley, 1972); effective medium approaches (Berryman, 1974; Kuster & Toksoz, 1974); or using the Kramers-Kronig relationship (O'Donnel et al., 1981, Beltzer et al., 1983). At present, it is not yet known how high the concentration has to be for the various scatterers to be correlated and a single scattering approximation to be unphysical, i.e., to disagree with experimental observations. Multiple scattering corrections are certainly found to be dominant when the scatterers are densely packed. As far as the author is aware, the main theoretical problem is that the exact form of the multiple scattering correction is not yet known. Although situations involving inhomogeneous viscoelastic media have already received some theoretical attention, experimental verification of these theories has not to date been forthcoming. This verification will be the main object of this thesis. An experimental approach based on particular models of inhomogeneous viscoelastic materials was used, of which results were compared with a numerical solution based on an
analytical approach. The analytical approach used (multiple scattering of waves) is discussed in chapter 2 and its numerical solution is discussed in chapter 8.

This study has been centered on specimens of silicone rubber blocks with suspensions of randomly distributed glass beads of different sizes and concentrations, which form simple and well defined experimental models of inhomogeneous viscoelastic media. At normal conditions of temperature and pressure, silicone rubbers are viscoelastic materials, and glasses are elastic materials. Silicone rubbers exhibit high absorption coefficients (longitudinal and traverse), while glasses exhibit very low absorption coefficients. Glass beads are easily obtainable in ranges of sizes. Because of the acoustic impedance mismatch between silicone rubber and glass, a good scattering coefficient results. Specimens of silicone rubber matrix (RIV-602) with suspension of glass beads (soda and lead glass) randomly distributed provided the laboratory models with a range of concentration of glass beads, classified into four groups according to the mean diameter of the beads. A fifth group of specimens of pure silicone rubber was used as reference specimens. These experimental models are described in detail in chapter 3.

Velocity and attenuation of ultrasound in the above models of inhomogeneous media were measured by using transmission techniques. A detailed description of the techniques and methods used for the measurements is given in chapter 4.

As pointed out earlier, diffraction is one of the influencing factors that must be considered for reliable results of velocity and attenuation. An extensive study concerning this phenomenon, for a pair of transducers (transmitter and receiver) in distilled water, formed the basis of chapter 5.
The preliminary and final measurements of velocity and attenuation in the models of inhomogeneous viscoelastic media are presented in chapter 6 and 7, respectively. The predictions given by the multiple scattering theory calculations are presented together with the experimental results. The discussion of these results is consolidated in chapter 8.

This thesis is concluded in chapter 9 where the achievements and applications of the study are given. Further investigations that could possibly clarify certain aspects left without explanations are suggested for future work.
CHAPTER 2

MULTIPLE SCATTERING OF WAVES FORMULATION
As mentioned in the previous chapter, Anson & Chivers (1984a and 1984b) recently reviewed and compared a large number of publications concerning wave propagation in inhomogeneous media. Therefore, a literature review will not be presented here and the reader is referred to those references. Among the theoretical approaches reviewed and analysed by Anson & Chivers was the formulation of multiple scattering of waves by Waterman & Truell (1961). This formulation had been utilised by various authors and shown to be valid for different scattering systems at low volume concentration of scatterers, for example: glass-filled polymer solids (Datta & Pethrick, 1980); glass marbles in araldite resin (Lefebvre et al., 1980); epoxy matrix containing lead inclusions (Sayers & Smith, 1983); polystyrene spheres in castor oil (Adach et al., 1986) among others. Therefore, it was decided to use this formulation for the materials of interest in the present investigation of glass beads in silicone rubber.

The formulation of Waterman & Truell (1961) is in fact an extension to the work of Foldy (1945) and of Lax (1952). Foldy (1945) had introduced the concept of configurational averaging by utilizing the joint probability distribution for occurrence of a given configuration of independent isotropic point scatterers to average the resulting wave over all configurations. Lax (1952) then generalised this procedure to include point scatterers with quite general scattering properties, using a quantum mechanical formulation.
The problem that Foldy had addressed was the relation that exists between the exciting field acting on a scatterer at a point, and the total field which would exist at that point if the scatterer were not there. Without the solution to that problem one cannot obtain a governing equation for the desired average field quantity. Foldy made the assumption that the two fields were equal, whereas Lax related them by a constant of proportionality which was assumed to be close to unity. Waterman & Truell derived an equation governing wave motion in a medium containing an array of finite anisotropic scattering regions randomly distributed. The anisotropy gave rise to an extra term in Foldy's equation, which was a second order term in number density of the scatterers, i.e., the last term in equation (2.8).

An incident plane longitudinal wave is assumed entering normally on an infinite half-space matrix containing a statistically independent distribution of non-overlapping anisotropic scatterers.

When an incident plane wave, represented by the wave potential $\Psi$, impinges upon an elastic spherical scatterer of radius $a$, four other waves are generated. All these four waves may also be represented by wave potentials: the scattered longitudinal and shear wave potentials in the matrix material, $\Psi$ and $\Pi$, respectively; and the longitudinal and shear waves potentials in the scatterer, $\Psi'$ and $\Pi'$, respectively. Thermal waves in either matrix or in scatterer materials are not being considered here. The complex wave potentials under consideration may be written in spherical coordinates as:
\begin{align}
\Psi_l &= \sum_{n=0}^{\infty} i^n (2n + 1) j_n(kr) P_n(\cos \theta) \tag{2.1} \\
\Psi_r &= \sum_{n=0}^{\infty} i^n (2n + 1) A_n h_n(kr) P_n(\cos \theta) \tag{2.2} \\
\Pi_l &= \sum_{n=0}^{\infty} i^n (2n + 1) B_n h_n(kr) P_n(\cos \theta) \tag{2.3} \\
\Psi' &= \sum_{n=0}^{\infty} i^n (2n + 1) C_n j_n(k'r) P_n(\cos \theta) \tag{2.4} \\
\Pi' &= \sum_{n=0}^{\infty} i^n (2n + 1) D_n j_n(k'r) P_n(\cos \theta) \tag{2.5}
\end{align}

where \( r \) is the radial coordinate, \( k = \omega/c_l + i \alpha_l \) and \( K = \omega/c_s + i \alpha_s \) are respectively the complex longitudinal and shear wave numbers in the matrix material, and \( k' = \omega/c'_l + i \alpha'_l \) and \( K' = \omega/c'_s + i \alpha'_s \) are those in the scatterer material. The letter \( c \) symbolizes velocity and \( \alpha \) symbolizes absorption, the prime refers to the scatterer material, \( \omega \) is the angular frequency, and \( i = \sqrt{-1} \). \( P_n \) is the \( n \)th order Legendre functions, \( j_n \) is the \( n \)th order spherical Bessel functions, and \( h_n \) is the \( n \)th order spherical Hankel functions of the first kind. The constants \( A_n, B_n, C_n \) and \( D_n \) are scattering coefficients. Upon applying boundary conditions at the surface of the spherical scatterer \((r = a)\), i.e., continuity of normal and tangential displacements and normal and shear stresses, a set of four equations in four unknowns \((A_n, B_n, C_n \) and \( D_n)\) is subsequently obtained. These equations are readily available in Gaunaurd & Uberall (1982) and therefore will not be reproduced here. However, a printing mistake is found in that reference, i.e., \( A_1^* = x_{q1} j_{1n}(x_{q1}) \) should be \( A_1^* = -x_{q1} j_{1n}(x_{q1}) \). By determining the scattering coefficient \( A_n \), the forward scattering and backscattering amplitudes, \( f(0) \) and \( f(\pi) \), are obtained from the
following equations:

\[ f(0) = \frac{1}{ik} \sum_{n=0}^{\infty} (2n + 1)A_n \]  

\[ \text{(2.6)} \]

and

\[ f(\tau) = \frac{1}{ik} \sum_{n=0}^{\infty} (-1)^n (2n + 1)A_n \]  

\[ \text{(2.7)} \]

An expression that yields the complex wave number (or complex propagation constant), \( \beta \), describing the scattering medium (the composite) is given in the form

\[ \left( \frac{\beta}{k} \right)^2 = 1 + \frac{4\pi n_\omega f(O)}{k^2} + \frac{4\pi^2 n_\omega^2 [f(O)^2 - f(\tau)^2]}{k^4} \]  

\[ \text{(2.8)} \]

where \( \beta = \omega/c^*_l + i \alpha^*_l \), \( \omega \) is the angular frequency, \( c^*_l \) is the longitudinal phase velocity (the asterisk symbolizes the composite), \( \alpha^*_l \) is the longitudinal attenuation, and \( n_\omega \) is the number of scatterers per unit volume. An equivalent expression to equation (2.8) was previously derived by Urick & Ament (1949) for the case of particle suspension in liquids. If the second order terms of equation (2.8) are ignored, or if isotropic scatterers are considered \([f(0) = f(\tau) = f]\), Waterman & Truell's formulation becomes exactly the same as the one derived by Foldy.

By averaging over different types and/or sizes of scatterers \( \langle f(O) \rangle \) is determined, which is the average forward scattering amplitude, and \( \langle f(\tau) \rangle \) the average backscattering amplitude, by including an average value of \( A_n \) in equations (2.6) and (2.7). The parameters needed in the computation of \( \langle f(\theta) \rangle \) are: size (radius) distribution of scatterers; density; and frequency dependent shear and longitudinal velocities and absorptions in both scatterer and matrix material.

In summary, the behaviour of the scattering medium, characterized
by the complex propagation constant $\beta$, may be specified explicitly in terms of the number of scatterers per unit volume and the far field scattering amplitude $f(\theta)$ obtained for a single scatterer. However, when the scatterers become sufficiently densely concentrated and closely spaced, the scatterers may not be treated as independent due to non-negligible interference between the reflected wave functions. Nevertheless, as pointed out earlier in this chapter, it is not yet known when the concentration starts to become considerably high to start causing failure of this theoretical formulation. However, it has been found suitable for concentrations below 15% by volume for lead spheres in epoxy matrices (Sayers & Smith, 1983).

The results of the theoretical approach are discussed in chapter 8 together with the experimental observations in the materials concerned, i.e., glass beads in silicone rubber matrix.
CHAPTER 3

THE PHYSICAL MODELS
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3.1 - INTRODUCTION

The samples used for this study of ultrasonic propagation consist of glass beads (glass ballotini, Jencons Scientific Ltd, England) suspended in matrices of silicone rubber compound. These samples were built by Dr. R.A. Bacon in 1979.

The advantages that these samples have for investigations of the fundamental ultrasonic interaction mechanisms are: a very long term stability, and relatively wide ranges of bead size and concentration. Another advantage of using silicone rubber is that it is transparent, thus permitting a visual assessment of the uniformity of the suspension. Chivers and Bacon (1983) examined some of these samples on diagnostic ultrasonic scanners, presenting images with scattering characteristics similar to biological tissue. These models are not ideal for scanner assessment because of their low ultrasonic velocity when compared with biological tissues. In this particular type of room temperature vulcanizing silicone rubber (RTV-602) the approximate value of the velocity of ultrasonic waves is about 1020 m s⁻¹ at the temperature of 20°C instead of about 1500 m s⁻¹ for soft human tissues. However, an attenuation of ultrasound at a given frequency can be achieved similar to that of human tissues if the right combination of concentration and size of scatterers (the beads) in the matrix is chosen.
The matrix consisted of a silicone rubber compound (RTV-602) mixed with 0.5% of catalyst (SCR-05). As reported by Bacon et al. (1980), the silicone rubber liquid was degassed at low pressure after the hardening agent and the glass beads had been mixed in. The mixture was then poured into moulds of nominal dimension 3 cm x 4 cm x 5 cm, and rotated in an oven at a temperature of about 35 °C for approximately an hour, before being removed from the mould, in a gel state.

The samples were constructed with four different groups of ballotini glass beads having a nominal diameter in the range 53-63 μm, 90-106 μm, 250-300 μm and 500-600 μm and for various concentrations by weight of rubber. They were classified into five groups: one group of two samples which did not contain any ballotini and four other groups according to the bead size. The total number of samples was 32 and their properties are tabulated in tables 3.2.a, 3.2.b and 3.2.c.

To check the characteristics concerning each sample, given by Bacon et al. (1980), a series of measurements were carried out. These measurements were:

- dimensions of the samples;
- size of beads;
- density of bead material (glass);
- density of matrix material (silicone rubber RTV-602);
- effective density of each sample.

With the above set of measurements, it was then possible to estimate the concentration of beads in suspension in the samples. This was necessary for comparison of the experimental results of ultrasonic velocity and attenuation with the theoretical prediction of propagation of ultrasound in the samples. The results of velocity and attenuation are presented in chapter 6 and 7 and discussed in chapter 8.
3.2 - DIMENSIONS OF SAMPLES

The dimensions of the samples, given in table 3.2.a, are the average of 2 measurements obtained with the use of a dial gauge at the centre of each side. The typical error figure of thickness measurement is given in table 3.2.a. A pair of glass plates of 15 mm x 15 mm x 1 mm dimension was used, with the plates on the centre of either side of the sample, in order to spread the load caused by the dial gauge, which was mounted on a specially built instrument as shown in figure 3.1. The dimensions of the samples were taken from comparison with blocks of aluminium having the nominal dimensions (measured with vernier callipers accurate to 0.001 mm) of the samples. The standardization and accuracy of the dimensions of each sample is a very important aspect of the aim of the present work, which is to seek the variation of the ultrasound propagation with the inclusion of scatterers, over the whole group of samples. An error in thickness of 1% would cause an error of 1% in attenuation measurements and an error of 1% in velocity measurements. The magnitude of these errors is not necessarily insignificant when the physical mechanisms of interaction of the ultrasonic waves with the medium are sought.
FIGURE 3.1: Instrument for measurement of dimensions of the samples. A - aluminium cylinders of height equal to the nominal dimensions of the samples, B - stand, D - dial gauge, G - glass plates, M - micro-manipulator, S - sample.
3.3 - SIZE OF BEADS

Histograms of distribution of size of the glass beads were obtained with the use of the Cambridge Instruments Quantimet-920 system available at the Department of Materials Science and Engineering of the University of Surrey.

The Quantimet-920 is a multipurpose quantitative image analyser. Images for analysis can be obtained from a microscope (optical or scanning) or from a photographic image (negative or positive) or from a video-recorder. "Live" images from the video-type scanners (television-style input camera) are converted into an array of 704 lines each of 896 picture elements (pixels). The grey level of each pixel (64 grey levels) is converted to an eight bit binary number after a threshold level setting was detected. Its image analysis processor analyses the digitised images, producing measurements and effecting image manipulation.

Amongst the typical applications of the Quantimet-920 are particle shape and sizing for quality control. The histograms of the size (diameter) distribution of the beads given by the Quantimet-920 are presented in figure 3.2. The nominal diameter, total number of beads counted, the mean diameter and respective standard deviation are also presented. A macroviewer and an optical microscope were used for the analysis of the beads. The former was considered adequate for the two larger bead sizes and the latter for the two smaller ones. The minimum magnified image for analysis occupied an area of at least 25 pixels, which is five times greater than required for minimum detectable levels.
GROUP II
nominal diameter: 500-600 μm.
total number of beads counted: 527
mean diameter: 582 μm.
st. dev.: 85.6 μm.

GROUP III
nominal diameter: 250-300 μm.
total number of beads counted: 350
mean diameter: 317 μm.
st. dev.: 87.3 μm.

GROUP IV
nominal diameter: 90-106 μm.
total number of beads counted: 173
mean diameter: 101 μm.
st. dev.: 21.4 μm.

GROUP V
nominal diameter: 53-63 μm.
total number of beads counted: 629
mean diameter: 63.8 μm.
st. dev.: 21.7 μm.

FIGURE 3.2: Size distribution of the glass beads.
3.4 - DENSITY OF COMPONENTS OF SAMPLES

The densities of the glass beads and of the silicone rubber matrix were measured with the use of a density bottle. To measure these densities, the following procedure was adopted:

A density bottle of 50 cm³ was used. A mass \( m_i \) of the material to be measured (glass beads or pieces of silicone rubber) was placed little by little in the bottle half filled with distilled water, followed by extraction of air bubbles. This extraction of air bubbles in the bottle was achieved by placing the bottle in a ultrasonic bath cleaner and in a vacuum chamber, in turn, until a virtual elimination of air content (assessed by eye) was obtained. The bottle was then filled up with water followed by the same extraction of air content procedure described above. The total mass, \( m'_t \), of bottle, water and the material is given by:

\[
m'_t = m_0 + m_i + (V_o - V_i) \cdot \rho_w
\]  

where \( m_0 \) is the mass of the density bottle; \( m_i \) is the mass of the material being measured (glass beads or bits of silicone rubber); \( V_o \) is the volume capacity of the bottle; \( V_i \) is the unknown volume of material and \( \rho_w \) is the density of water.

The density of water, \( \rho_w \), was also measured with the same bottle and using the same degassing procedure as above. Two measurements were taken, giving an average value and variation of 0.9827 ± 0.0001 g cm⁻³ at the temperature of (18.6 ± 0.1) °C. The value tabulated by Kaye & Laby (1973) is 0.99859 g cm⁻³ at 18°C.

Adding an extra measured mass \( m'_1 \) (for example, \( m'_1 \approx m_i \)) and consequently an extra volume \( V'_1 \) of material in the bottle, making a total mass and volume of material \( m_2 (= m_i + m'_1) \) and \( V_2 (= V_i + V'_1) \) respectively, the overall mass \( m''_t \) is:

\[
m''_t = m_0 + m_2 + (V_o - V_2) \cdot \rho_w
\]  

(3.2)
Subtracting eq.(3.1) from eq.(3.2) one can write
\[ m''_t - m'_t = (m_2 - m_1) - (V_2 - V_1) \cdot \rho_w \]  
(3.3)

The ratio by which the mass of material was increased can be determined by:
\[ r = \frac{m_2 + m'_2}{m_1} = \frac{m_2}{m_1} \]  
(3.4)

Providing that the density of the material was not changed during the process, the same ratio \( r \) is valid for the volume of material,
\[ r = \frac{V_2 + V'_2}{V_1} = \frac{V_2}{V_1} \]  
(3.5)

Thus, eq.(3.3) can be written as
\[ m''_t - m'_t = (r - 1) \cdot m_1 - (r - 1) \cdot V \cdot \rho_w \]  
(3.6)

By rearranging this equation, the volume \( V_1 \) of material can be determined as
\[ V_1 = \frac{m'_1 - m''_1 + (r - 1) \cdot m_1}{(r - 1) \cdot \rho_w} \]  
(3.7)

The density of the material can subsequently be determined from
\[ \rho \_1 = \frac{m_1}{V_1} \]  
(3.8)

The value of the ratio \( r \) was approximately 2, i.e., \( m_1 \approx m'_1 \) and \( V_1 \) ranged from 5.8% to 13.8% (depending on the size of the glass beads or the pieces of silicone rubber) of the volume capacity of the bottle, \( V_0 \). The densities of the glass beads and of the pieces of silicone rubber were measured twice. The average value of the density of the material over the two sets of measurements are tabulated in Table 3.1 at the temperature indicated. The typical variation between any two measurements on the same material was approximately 0.001 g cm\(^{-3}\). The manufacturers of the glass ballotini have quoted their densities to be in the range of 2.48 g cm\(^{-3}\) to 2.95 g cm\(^{-3}\).
### TABLE 3.1

<table>
<thead>
<tr>
<th>material</th>
<th>group</th>
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#### 3.5 - DENSITY OF SAMPLES

To determine the concentration of beads present in the samples, the density of each individual sample had to be measured. For this, the mass and the volume of each sample were measured. A digital balance (accuracy of ±0.5 mg) was used for the measurement of the mass of each sample. The device developed for volume measurement (accurate to ±0.1 cm³) of each sample is shown schematically in figure 3.3. That device was based on the principle of volume of water displacement. The measured density of each individual sample at the temperature of 20±2°C is tabulated in table 3.2.b. The average value of the measured densities of samples 1 and 20 (the two clear silicone rubber blocks) was used for the computation of the concentration of beads in the other samples, as discussed in section 3.6. The error figure shown on the top of column 3 of table 3.2.b is the variation of the measured densities of samples 1 and 20 with respect to their average value.
FIGURE 3.3: Volume of sample measurement device.
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* Estimated effective value due to settling of beads onto one side of sample along the nominal thickness of 3 cm.

** The calculated figures were approximated after calculations.
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</tbody>
</table>

* estimated effective value due to settling of beads onto one side of sample along the nominal thickness of 3 cm.

** The calculated figures were approximated after calculations.
3.6 - CONCENTRATION OF BEADS IN SUSPENSION IN MATRIX

The volume concentration of beads suspended in the matrix was determined from the measured density of each of the components comprising the composite and the measured density of the composite itself. The relationship between the densities and the percentage of volume concentration of beads is given by:

\[ \% \text{vol} = 100 \frac{(\rho^* - \rho)}{(\rho' - \rho)} \]  

(3.9)

where \( \rho^* \) is the density of the composite; \( \rho \) is the density of the matrix (average value between samples 1 and 20), and \( \rho' \) is the density of the bead.

The original data concerning these samples, given by Bacon et al. (1980), were presented in terms of percentage by weight of beads, since the weight of glass beads and of silicone compound plus catalyst were the quantities they measured for the construction of the samples.

However, they noticed that at the degassing process stage of manufacturing the samples, the glass beads (particularly the two larger sizes, group II and III) tended to sink to the bottom of the containing vessel. This may have caused a loss of concentration of beads when the mixture was poured into the moulds.

To compare the measured concentration against the figures given by Bacon et al. (1980), the following equation was used:

\[ \% \text{wt} = \% \text{vol} \frac{\rho'}{\rho^*} \]  

(3.10)

The above parameters are presented in table 3.2.b for ease of comparison. The accuracy of mass density measurements were limited by the accuracy with which the volume could be measured. However, the apparently significant differences in concentration between the present measurements and those of Bacon et al. (1980) for samples 2, 3, 4 and 8
are not thought to be related to errors of measurements by the overall density method described in section 3.5. Those large differences are thought to have been originated from the settling of beads at the degassing process stage of manufacturing the samples, which might be expected to be more prominent for the larger beads. Bacon et al. (1980) also assessed the finished models, by means of an optical microscope, counting the number densities. They found that samples 2 and 3 had significantly lower density number than initially intended, and that the others were generally of the same order as predicted by the weighings. The figures they reported are transcribed in tables 3.2.b and 3.2.c for comparison.

In addition, sample 4 has effectively a higher concentration of beads than that determined from its measured density. The reason for this is that the beads settled onto one side (3 cm thickness) of the sample during the process of manufacturing, making a gradient of increasing concentration of beads in suspension, leaving a slab of clear silicone rubber of approximately 4 mm thick. The estimated concentration within the remainder of the block containing the beads in sample 4 (assuming uniform distribution) is that marked with an asterisk in tables 3.2.b and 3.2.c.

3.6.1 - ERRORS IN MEASURED CONCENTRATION

The error figures of the measured concentration of beads in the samples (tabulated in table 3.2.b) were determined from the formulation of the most probable error (Topping, 1979). This approach of determination the magnitude of errors of measurements of concentration involved each physical quantity of equations (3.9) and (3.10).
Consider the fractional volume concentration
\[
\text{vol} = \frac{\rho^* - \rho}{\rho^* - \rho} \tag{3.11}
\]

The most probable error in the volume fraction is given by
\[
\epsilon_{vol}^2 = \left(\frac{\partial \text{vol}}{\partial \rho}\right)^2 \epsilon^2 + \left(\frac{\partial \text{vol}}{\partial \rho^*}\right)^2 \epsilon_{\rho^*}^2 + \left(\frac{\partial \text{vol}}{\partial p^*}\right)^2 \epsilon_{\rho^*}^2 \tag{3.12}
\]
where \(\epsilon_r\), \(\epsilon_{\rho^*}\), and \(\epsilon_{\rho^*}\) are respectively the errors of measurements in the densities of the matrix, the glass beads and the composite.

By solving the derivatives of eq.(3.12) one can write:
\[
\epsilon_{vol} = \text{vol.} \left[\left(\frac{\text{vol} - 1}{\rho^* - \rho}\right)^2 \epsilon_r^2 + \left(\frac{1}{\rho^* - \rho}\right)^2 \epsilon_{\rho^*}^2 + \left(\frac{1}{\rho^* - \rho}\right)^2 \epsilon_{\rho^*}^2\right]^{1/2} \tag{3.13}
\]

Similarly, the errors of measurements in the fractional weight concentration of beads were calculated from
\[
\text{wt} = \text{vol.} \frac{\rho^*}{\rho^*} \tag{3.14}
\]
given in the form of
\[
\epsilon_{wt}^2 = \left(\frac{\partial \text{wt}}{\partial \text{vol}}\right)^2 \epsilon_{\text{vol}}^2 + \left(\frac{\partial \text{wt}}{\partial \rho^*}\right)^2 \epsilon_{\rho^*}^2 + \left(\frac{\partial \text{wt}}{\partial \rho^*}\right)^2 \epsilon_{\rho^*}^2 \tag{3.15}
\]
which yields
\[
\epsilon_{wt} = \text{wt.} \left[\frac{\epsilon_{wt}^2}{\text{vol}^2} + \frac{\epsilon_{\rho^*}^2}{\rho^*} + \frac{\epsilon_{\rho^*}^2}{\rho^*}\right]^{1/2} \tag{3.16}
\]
The error figures tabulated in table 3.2.b are given in terms of percentage, which are the results given by eq.(3.13) and (3.16) multiplied by the factor of 100, for each sample in question.

From comparison with the weight concentration given by Bacon et al. (1980) it was concluded that they did not invalidate the measurements of concentration made by the present author. Therefore, the values of volume concentration set out in column 4 of table 3.2.b were the ones used in the theoretical calculations described in chapter 8.
3.7 - NUMBER DENSITY OF BEADS AND THEIR MEAN SEPARATION

To estimate the number density of beads and their mean separation, the beads were assumed to be uniformly distributed. Let $V$ be the total volume of matrix plus beads and $n$ the total number of beads. Thus, the volume of the composite associated with one of the beads is:

$$V' = \frac{V}{n} \tag{3.17}$$

The volume occupied by the matrix alone is $(V - v)/n$, where $v$ is the total volume of the beads. Considering the beads being of spherical shape, the volume of each one is given by $v/n = (4/3)\pi r^3$, where $r$ is its mean radius.

The number density of beads in the matrix, i.e., the number of beads per unit volume of the specimen, $n/V$, can be calculated from their relative volume concentration as

$$n = \frac{3 \cdot \%vol}{V \cdot 4\pi r^3 \cdot 100} \tag{3.18}$$

Assuming $V'$ to be a cube of side $s$, as shown in figure 3.4, with the bead at the centre of the cube, one can rewrite eq. (3.17) in the form

$$s^3 = \frac{V}{n} \tag{3.19}$$

Therefore, the mean separation between the centre of two neighbouring beads is given by

$$s = \left(\frac{V}{n}\right)^{1/3} \tag{3.20}$$

The estimated mean number density of beads in suspension in each sample, as well as their mean separation, are tabulated in table 3.2.c.
FIGURE 3.4: Theoretical estimation of the separation between two neighbouring beads in matrix. There is one element of matrix, a cube of side $s$, associated with each bead localised at the centre of the cube.
4.1 - INTRODUCTION

The conventional techniques of measurement of velocity and attenuation of ultrasonic waves in solid materials can be classified into two groups: [a] those using continuous waves, and [b] those using pulsed waves. Each of these two groups presents a variety of techniques tending to aim for: specific applications; overcoming of limitations; and greater precision.

Many of the basic text books on sound and ultrasound discuss various techniques of measurements to some extent. As this is not the main object of the present work, only a brief account will be included here, and the reader is referred to the following bibliography for further details: Babikov (1960), McSkimin (1961a), Blitz (1963), Beyer & Letcher (1969), Krautkramer & Krautkramer (1977), Wells (1977), Pollard (1977) and Szilard (1982).

Continuous waves, or tone bursts of sufficient duration, can be used in resonance techniques for measurement of velocity and attenuation as a function of frequency, as long as standing waves are established. This is possible in materials of very low attenuation, or in specimens of high attenuation but short path length.

Travelling wave techniques employ continuous waves in the low frequency range (below 100 kHz). They have been used for measurements on solids in the form of a long, thin rod or wire. Standing waves must
be avoided. They require the use of a transmitting transducer coupled at one end of the specimen and a travelling receiving transducer coupled along the rod. Since the frequency is known, the velocity can be calculated from the measured wavelength. The attenuation is determined from the displacement decay curve. The single pulse transit technique can also be used in specimens in the form of a long rod or wire, as above. For a short pulse, the group velocity is measured, whereas for a long tone burst the phase velocity is measured, by measuring the relative time of transit of the pulse at the position of the receiving transducer. The attenuation is measured from the decay in pulse amplitude.

Optical techniques make use of the interaction between a beam of travelling ultrasonic waves of high frequency and a beam of monochromatic light. One beam is set perpendicular to the other. Successive compressions and rarefractions of the mechanical waves give rise to an optical diffraction pattern. These techniques apply only to optically transparent materials. The intensity of the light is proportional to the intensity of the mechanical waves, hence allowing the determination of the attenuation. The velocity is obtained from the change in the refractive index of the material.

Reflection coefficient techniques relate the characteristic acoustic impedances of the materials involved, when an acoustic interface is encountered.

Through-transmission techniques employ two transducers, one as transmitter and the other as receiver. The transducers are placed coaxially, facing one another on opposite sides of the specimen. The velocity is determined by measuring the time of flight (or transit time) that an ultrasonic pulse takes to travel through a specimen of known thickness. The attenuation is obtained by measuring the change
in amplitude of the ultrasonic signal relative to a reference level.

The pulse-echo technique is widely used in various ways in the non-destructive testing of materials. The pulse can be either of narrow-band or of wide-band in the frequency domain. This technique can be arranged to use either a single transducer acting as both transmitter and receiver (sometimes called transceiver) or a probe incorporating separate transmitting and receiving transducers. Taking a specimen of known thickness, the time between multiple echoes (from a reference reflective acoustic interface) of a transit pulse is proportional to the velocity. The decay rate of the amplitude of the echoes determines the attenuation.

Regarding the frequency range of interest (1 to 10 MHz) and the type of specimens described in chapter 3, some of the above techniques were considered impractical for one reason or another. The impracticalities concerning each technique (for the purposes of this investigation) are given below:

Resonance technique: the relatively high attenuation in the specimens and their dimensions would impede the establishment of standing waves.

Travelling wave technique: the specimens did not meet the geometry required, i.e., that of a thin rod.

Optical Techniques: the specimens were transparent but diffusive of light, and highly attenuating at high frequencies.

Reflection techniques: not always applicable to inhomogeneous medium because the interfaces are not always representative of the medium as a whole.

Pulse-echo technique: the relatively high attenuation in the specimens and the four reflection losses of signal at the interfaces would lead to a low signal-to-noise ratio.
The only technique left to investigate the velocity and attenuation in the specimens in the frequency range of interest was the through-transmission one, and it was consequently adopted for this work.

There are two simple and reliable methods of measuring velocity and attenuation with the through-transmission technique: [a] comparison method, and [b] substitution method. By the comparison method is meant the comparison of measurements of time of flight and of amplitude of the signal in two or more specimens of the same constitution but of different length. The substitution method consists of the replacement of the specimen by a reference medium of known velocity and attenuation, to which comparisons are then made. In laboratory experiments, the reference medium normally used is distilled water, for its very low absorption at low megahertz frequency, known velocity, known dependence of its parameters with environmental conditions, and the fact that it is easily obtainable. The substitution method was adopted for practical reasons. It is discussed in detail in section 4.2.

The limitations associated with the through-transmission techniques are, in a way, similar to some associated with other techniques, i.e.: the characteristics of the transducers; the coupling of the specimen with the transducers or with any delay-line (if used); the alignment of the transducers and of the specimen with respect to the acoustic beam; the degree of transmission at the interfaces; the geometry of the specimen; the portion of the signal being observed; diffraction effects; the environmental conditions; and the electronic and mechanical equipment. Pulsed waves (of wide or of narrow frequency band) or continuous waves can be applied. Standing waves (where resonance can occur) must be avoided. A suitable choice of
experimental arrangements permit variants of the through-transmission technique to accommodate the requirements of the investigator. These may include the geometry and the type of specimen, automation of the measurements, frequency spectrum analysis, the parameters to be measured, operating range, system calibrations, the degree of precision of measurements, versatility and portability.

Having regard to the equipment and facilities available in the laboratory, six variants of the through-transmission technique were used, all employing the substitution method. The reference medium used was distilled water, in which the specimen was immersed.

The six measurement techniques used are listed below and described separately in subsequent sections of the present chapter. The frequency ranges utilised are shown in brackets.

i - digitised signal

[a] - wide-band excitation - (1 and 2 MHz)

[b] - tone-burst excitation - (1.5 to 8 MHz)

ii - time interval averaging - (1.5 to 8 MHz)

iii - time delay spectrometry (0 to 9 MHz)

[a] - pair of transducers

[b] - transducer—hydrophone

iv - swept frequency technique (0 to 11 MHz)

Based on the author's own experience while carrying out this work, when the velocity and attenuation are measured by the substitution method, the reference medium must also be acoustically characterized. For reliable results on the specimen under investigation, a regular check on the characteristics of the reference medium should be made. The main parameters are the temperature and velocity. For this, the time of flight of the waves is an easy and quick parameter to be
checked, especially if a separate and dedicated ultrasonic source and receiver are set up in that same medium of reference.

Because of diffraction, the measured velocity and attenuation will depend on the transmitter-receiver (Tx-Rx) separation. A detailed discussion and analysis on this matter is presented in chapter 5. Ideally, one should have plane waves entering the specimen under characterization, which would permit a theoretical model to be compared with the experimental results in an easier manner. Practically this may mean a limitation on the experimental set up. Nevertheless, an approximation to plane waves can be encountered in the far field of the acoustic beam, permitting the diffraction correction to be of a minor magnitude. If the experiments are to be carried out in the near field of the acoustic beam, then the diffraction corrections will play a more important role.

4.2 - SUBSTITUTION METHOD

Ultrasonic velocity and attenuation in a specimen can be determined relative to a reference medium of known characteristics. This way of quantifying the ultrasonic velocity and attenuation in a particular specimen of interest is known as the substitution method.

The following discussion on the principles of measurements of velocity and attenuation are based on the assumption that plane waves are entering the specimen. Therefore, no diffraction effects are being considered.

Consider the transmission method as schematically represented by figure 4.1. The ultrasonic transmitting transducer, Tx, and the ultrasonic receiving transducer, Rx, are positioned coaxially at a separation Tx-Rx = z. The transducers and the sample S of thickness l are immersed in a water bath (the reference medium).
FIGURE 4.1: Substitution method for evaluation of ultrasonic velocity and attenuation in a sample $S$ (not to scale). (a) transducers immersed in water, and (b) transducers immersed in water with a sample in between.
4.2.1 - ULTRASONIC VELOCITY IN THE SAMPLE

The time of flight of the ultrasonic plane waves (measured at the receiver, Rx), when the sample S is not present in the acoustic field (fig. 4.1.a) is \( t = z/c \), where \( c \) is the ultrasonic velocity in water. The time that the ultrasonic waves take to travel through the sample alone is \( t_s = \frac{L}{c_s} \), where \( c_s \) is the velocity in the sample. When the sample is in the ultrasonic path in water (figure 4.1.b) the measured time of flight at Rx is:

\[
\begin{align*}
\frac{z}{c} + \frac{L}{c_s} = \frac{z}{c} + \frac{L}{c} + \frac{L}{c} \\
\frac{t_s}{c} = \frac{t}{c} + \frac{L}{c} \\
\text{Rearranging this equation, one can write} \\
c_s = \frac{L}{t_s - t + \frac{L}{c}} \quad (4.2)
\end{align*}
\]

Therefore, provided that the ultrasonic velocity in water is known, the velocity in the sample can be obtained by measuring the times of flight with and without the sample present.

4.2.2 - ULTRASONIC ATTENUATION IN THE SAMPLE

The ultrasonic attenuation, as in the case of velocity, can be measured by using the substitution method. Suppose that the amplitude of the received signal at Rx of figure 4.1.a is \( A_w \). When the sample is immersed in the ultrasonic field, figure 4.1.b, the received signal amplitude is \( A_s \).

If no reflection losses at the interfaces water/sample/water take place, and the absorption in the water is negligible, the amplitude attenuation \( \mu \) of the ultrasonic waves at normal incidence over the sample can be simply expressed as

\[
A_s = A_w \exp(-\mu L) \quad (4.3)
\]
Equation (4.3) can be re-written in logarithmic form as
\[ \mu = \frac{1}{l} \ln \left( \frac{A_s}{A_w} \right) \quad \text{[Np/length]} \] (4.4)
which gives the result in units of Nepers per unit length of the sample. Some authors prefer to use the decibel scale, which is a factor of 8.686 greater. Therefore the attenuation \( \alpha \) can be expressed as:
\[ \alpha = 8.686 \cdot \mu \quad \text{[dB/length]} \] (4.5)

When the reflection loss at the interfaces water/sample/water, and the absorption in water become of significant magnitude, the attenuation in the sample must be expressed accordingly. This situation is discussed in chapter 7.

4.3 - PROCEDURES BEFORE MEASUREMENTS

4.3.1 - ALIGNMENT OF TRANSDUCERS

Good alignment of the transducers is a difficult task and an important step towards obtaining accurate measurements.

The alignment procedure used in this work for a pair of 7.5 MHz transducers (Panametrics V-320, Acuscan immersion transducers, 12.7 mm diameter) was in the following sequential manner:

i: The transmitting transducer was mounted on a holder assembled on one side wall of the acoustic tank. That holder (Bristow et al., 1980) facilitated the manipulation of the orientation of the transducer to maximize the echo signals (8 MHz tone burst) reflected from the opposite wall of the tank, thus aligning the transmitter acoustical axis with the longitudinal scanning mechanism which was fixed to the tank, and which was to hold the receiving transducer.
ii: The receiving transducer was suspended from the tracking mechanism system which allowed movement along x, y and z axes, i.e., the horizontal, vertical and longitudinal axes respectively. In addition, angular movements about the x and y axes were possible for the adjustment of the orientation of the receiver facing the transmitter. This orientation was firstly achieved by eye, with the transducers separated by about 1 mm.

iii: With the receiver positioned at about 300 mm from the transmitter, the transmitter:receiver pair was acoustically aligned by the maximization of amplitude of the received signal in the region of the resonant frequency, by looking at the spectrum of the received signal of the pair Tx-Rx. A sinusoidal tracking generator from a 110 MHz Marconi spectrum analyser was connected to the transmitting transducer. After the waves were propagated through the water in the acoustic tank, the received signal was fed into the spectrum analyser receiver and displayed on its screen with the vertical axis display switched to a linear scale. The frequency was swept continuously in the range of 0 to 12 MHz. A -3 dB filter bandwidth of 50 kHz was used and synchronously swept at a speed automatically selected to be the fastest possible for negligible loss of amplitude of the spectral lines.

iv: The final step was to use a tone burst signal to check the alignment at a particular frequency. An 8 MHz tone burst was used for driving the transmitter. With the receiver positioned at about 300 mm, the second reverberating signal was observed on an oscilloscope screen at its maximum amplification (without causing distortion or losing part of the signal on the display). The 8 MHz frequency was chosen for two reasons: the proximity to the fundamental resonance of the transducers, giving consequently high sensitivity, and because there
was less diffraction than at lower frequencies. Once again, the orientation of the receiver was checked against its signal amplitude. This step is a refinement of the alignment.

v: By bringing the receiver close together with the transmitter, it was observed by eye that, from their geometry, they were coaxially positioned and facing parallel to each other, although this was not a necessary condition for the acoustic alignment of the transducers.

Two particular cases differ slightly from the procedure described above:

[a] - when the 1 MHz and the 2 MHz pair of transducers of 15 mm diameter (T.M.S., presently Systems & Instrumentations) were used, step [iii] had a frequency range of 0 to 6 MHz. In step [iv], instead of tone burst excitation, the transmitter was excited with a short pulse of high voltage (approximately 300 volts) causing the transducers to respond at their natural characteristics. The transducer separation was 150 mm.

[b] - step [iii] did not take place for the alignment of the transducers for the experiments involving the time delay spectrometry technique (described in section 4.4.3)
4.3.2 - ALIGNMENT OF THE SAMPLE

For a parallel sided specimen, the transit time reaches a minimum, although not sharp, at normal incidence. Consequently the sample was aligned in the acoustic field (for all techniques used, with exception of the time delay spectrometry - see section 4.4.3) by adjusting its orientation to give the minimum transit time of an ultrasonic pulse.

However, it was observed that all the samples exhibited less attenuation of signal when positioned at oblique incidence (about 10°). Unfortunately, due to lack of time, no experiment was carried out to measure the dependence of attenuation and velocity on the angle of incidence of the waves.

4.3.3 - SAMPLE HANDLING

To achieve temperature equilibrium, the samples were kept in polyethylene bags submerged in the water tank for a few hours before the investigations were started.

4.3.4 - RELATIVE POSITION OF THE SAMPLE

Ideally, one should have plane waves interrogating the samples. However, practically this may not be possible. Nevertheless, approximately plane waves may be encountered in the far field of the acoustic beam (see chapter 5). If measurements are to be carried out in the near field, diffraction corrections must enter into the interpretation of the results (see chapters 6 and 7). The samples were therefore placed in the far field of the transmitter.
4.3.5 - TEMPERATURE DEPENDENCE

The rate of change of velocity with temperature (in the vicinity of 20°C) in the two relevant media concerned in this investigation are: water: $3.07 \text{ m s}^{-1} \text{C}^{-1}$ (Greenspan & Tschiegg, 1959; Del Grosso & Mader, 1972); RTV-602 silicone rubber: $-2.8 \text{ m s}^{-1} \text{C}^{-1}$ at 0.6 MHz (Foldes, 1974). The absorption of ultrasound in water at 20°C and 1 MHz is approximately $2.2 \times 10^4 \text{ dB mm}^{-1}$ (Nozdrev, 1965) – see section 5.6. The rate of change of attenuation with temperature in RTV-602 estimated by Chivers & Bacon (1983) was about $-1.25 \text{ dB cm}^{-1} \text{C}^{-1}$, although they suggested further investigation (see section 7.5).

Temperature fluctuations may well disturb a series of measurements, depending upon the precision being needed. This may originate from differences in temperature between the water bath and the external environment. Although no formal experiment was performed, it was observed that enclosing the acoustic tank in expanded polystyrene had a significant effect in reducing the temperature fluctuation in the tank, which was intended to be maintained at 20°C.

4.3.6 - ULTRASONIC THERMOMETER

It is known that the velocity of sound is dependent upon the temperature of the medium through which the waves are being propagated. Therefore, one can use the velocity of the waves (which is proportional to the time of flight for a given distance) to determine the temperature of the medium of propagation, or simply as a sensor for a temperature controller.

The sensitivity of the device will depend on how well one can measure the velocity in the first instance. Subsequently, the sensitivity of the device to temperature variation will depend on the temperature coefficient of the speed of sound in the medium. The
effectiveness of the device in controlling the temperature in a given environment will depend on heat capacity, thermal conductance, etc. Lynnworth et al. (1971) used a metal-alloy rod for their device which was designed for operating at very high temperatures in nuclear reactions applications.

An ultrasonic thermometer was used to monitor the temperature of the water bath, in which the specimens were to be studied, in all the experiments performed (with exception of the time delay spectrometry technique). The temperature of the water bath was kept at 20.0 ± 0.1°C. The temperature was firstly read from an ordinary mercury thermometer (precision ±0.1°C). Secondly, a pair of 2 MHz ultrasonic transducers (15 mm diameter) immersed in the water and spaced 250 mm apart, was dedicated as the ultrasonic thermometer. The transmitting transducer, \( \Theta_t \), was excited by a short pulse of high amplitude (Metrotek MP-203). After the ultrasonic waves travelled through the water, the received signal at the receiving transducer, \( \Theta_r \), was monitored on a C.R.O. with its time base expanded to have a precision of ±20 ns. The highest peak of the received signal was used as reference. The water was circulated and stirred by means of a water circulator and a stirrer, after which at least two minutes interval was given, allowing the turbulence in water to die away, before any reading of time of flight was taken. Assuming the velocity of ultrasound in water as 1480 m s\(^{-1}\) (at 20°C) and assuming its temperature coefficient to be linear at the vicinity of that temperature, i.e., 3.07 m s\(^{-1}\)°C\(^{-1}\), (Greenspan & Tschiegg, 1959; Del Grosso & Mader, 1972) the precision achieved was approximately ±0.06°C.

When a wide band pulse was used for the investigation of the acoustic properties of the samples (at 1 and 2 MHz), the transducers used for the investigation were also used as thermometers with the
sample absent from the field. In this particular case, the precision achieved was ±1°C, because the author, at the time, was not aware of the magnitude of the temperature coefficient of the acoustic parameters being measured.

Despite the existence of conventional thermometers, the ultrasonic thermometers can provide improved precision and reliability in experiments involving either ultrasound or other physical phenomena.

4.4 - THE TECHNIQUES

Table 4.1 gives an outline of the techniques used for this investigation of propagation of ultrasound in the physical models described in chapter 3. A detailed discussion of each technique of measurement used is presented in the following sections, which employed the procedures before measurements discussed in section 4.3.
<table>
<thead>
<tr>
<th>MEASUREMENT TECHNIQUE</th>
<th>Tx Rx</th>
<th>f (MHz)</th>
<th>EXCITATION</th>
<th>Tx-Rx</th>
<th>Tx-Sz</th>
<th>TEMP. CONTROL PARAMETERS</th>
<th>MEASURED PARAMETERS</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>water sample</td>
<td>sample vel.</td>
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<td>digitised signal</td>
<td>T1 T1</td>
<td>1</td>
<td>wide-band</td>
<td>150</td>
<td>80</td>
<td>yes</td>
<td>no</td>
</tr>
<tr>
<td></td>
<td>T2 T2</td>
<td>2</td>
<td>wide-band</td>
<td>150</td>
<td>80</td>
<td>yes</td>
<td>no</td>
</tr>
<tr>
<td>time int. averaging</td>
<td>P P</td>
<td>1.5-8</td>
<td>tone-burst</td>
<td>200</td>
<td>100</td>
<td>yes</td>
<td>no</td>
</tr>
<tr>
<td>time delay</td>
<td>P P</td>
<td>0 - 9</td>
<td>freq. sweep</td>
<td>160</td>
<td>100</td>
<td>no</td>
<td>no</td>
</tr>
<tr>
<td>spectrometry</td>
<td>P H</td>
<td>0 - 9</td>
<td>freq. sweep</td>
<td>160</td>
<td>100</td>
<td>no</td>
<td>no</td>
</tr>
<tr>
<td>swept frequency</td>
<td>P P</td>
<td>0 - 11</td>
<td>freq. sweep</td>
<td>200</td>
<td>100</td>
<td>yes</td>
<td>no</td>
</tr>
<tr>
<td></td>
<td>P P</td>
<td>0 - 11</td>
<td>freq. sweep</td>
<td>500</td>
<td>250</td>
<td>yes</td>
<td>no</td>
</tr>
</tbody>
</table>

**TRANSDUCER TYPE:**

T1, T2: T.M.S. (presently Systems & Instrumentation); mediumly damped.

T1: 1 MHz nominal frequency; unfocussed; 15 mm diameter.

T2: 2 MHz nominal frequency; unfocussed; 15 mm diameter.

P: Panametrics Acuscan Immersion Transducers;
   ref.No. V-320; 7.5 MHz nominal frequency; unfocussed;
   12.7 mm diameter.

H: Hydrophone; H-405; 1 mm diameter.

Tx-Rx: Transmitter-Receiver separation.

Tx-Sz: Transmitter-Sample separation.
4.4.1 - DIGITISED SIGNAL

At the very early stage of this study, a computer programme was available for the existing micro-computer system in the laboratory, which was used to digitise the received signal for the quantification of the velocity and the attenuation of ultrasound in the medium of interest. That computer programme, called "kitvel", was written in Pascal by Dr.R.A.Bacon (Chivers & Bacon, 1983). A slightly improved version of the programme "kitvel" was named "edkit-2" and used with the system schematically illustrated in figure 4.2. The new version allowed electronic compensation of the attenuation of signal, resulting in maintaining the signal to noise ratio at an approximately constant level for the measurement of the whole batch of samples. The zero-crossing velocity and the signal velocity (detected start of signal) were simultaneously computed.

A more detailed block diagram of the system shown in figure 4.2 can be seen in figures 4.3 and 4.4. The specific differences and purposes of the systems of figures 4.3 and 4.4 are discussed in detail in sections 4.4.1.1 and 4.4.1.2, where the wide-band and the tone-burst excitation are respectively discussed.

The transmitting transducer can be driven either by a short pulse of wide-band frequency or by a sine-wave tone-burst. The first type of excitation makes the transducer respond at its natural characteristic frequencies. The latter forces the transducer to operate at a particular frequency, although some spurious frequency component signals, particularly at the beginning and at the end of the burst, can appear from either the electronic system, or from the transducer characteristic itself, or from both.
FIGURE 4.2: Ultrasonic velocity and attenuation measurement system from digitised signal.
The length of the transmitted ultrasonic pulse was determined by the duration of the excitation pulse and the damping characteristics of the transducers.

After the transmitted waves have travelled through the medium of interest, the received signal was fed via variable delays through a box-car system, which allowed the repetitive pulse to be digitised and stored at the micro-computer system.

Figure 4.2 illustrates the main block diagram of the system used. In the following two sections of this chapter the description of the electronics associated with the two types of excitation pulse used are given. Nevertheless, for convenience, their common blocks are described in this section.

The master oscillator was used to trigger two delay timers in the delay generator, the outputs of which were used to trigger the transmitter driver, the time base of the display, and the scan delay generator of the box-car system.

The box-car system comprised a linear gate and a scan delay generator. The computer programme ("redkit-2") controlled the box-car linear gate by means of the DAC-0 output connected to the external scan of the box-car scan delay generator. The output of the linear gate was taken, via the ADC-0 input to the microprocessor. The programme searched for positive and negative maxima and zero-crossings. The programme itself has several different modes of operation, and may be varied according to the type of the waveform emerging from the medium under investigation. The start of the received pulse is assumed to be at a threshold level (just above the noise level) set up by the user, and detected by the box-car/ADC system. The pre-set number of sequential peaks and zero-crossings to be digitised were counted from the assumed start of the pulse. A calibration mode was used before any
measurement was attempted. This was used to calibrate the time scale of the external scan control of the box-car via the DAC, against the trigger delay available on the velocity measurement apparatus.

The ultrasonic velocity in water, the thickness of sample, and any electronic compensation, such as time delay or variation in the amplifier gain or attenuator resetting, were entered for the computation of the results.

A pre-set number of positive and negative peaks and zero-crossings in the received pulse (with and without the specimen present) were searched, permitting the immediate evaluation of the attenuation and the velocity, respectively. The velocity was evaluated from the difference in time of flight of the pulse, while the attenuation was evaluated from the change in the received pulse amplitude.

4.4.1.1 - WIDEBAND EXCITATION

A block diagram of this experiment is shown in figure 4.3. Two pairs of transducers (T.M.S., presently Systems & Instrumentations, England) were used in this experiment. They had PZT ceramic elements of 15 mm diameter plane discs. The transducers were mediumly damped and of nominal frequencies of 1.0 and 2.0 MHz. (The frequency spectra of these pairs of transducers, at 220 mm separation in water are presented in appendix A). The transmitter was excited with a short pulse of high voltage (approximately 300 volts), at a repetition rate of about 0.1 kHz. The samples were placed at about 80 mm from the transmitter and the transducers were spaced 150 mm apart, immersed in a water tank.
FIGURE 4.3: Ultrasonic velocity and attenuation measurement system with wide-band excitation.
At the time this particular experiment was performed, the author was not aware of the magnitude of the variation of the velocity in silicone rubber with the temperature (see section 4.3.5). Nevertheless, the temperature was kept at \((20 \pm 1) ^\circ C\).

The first two peaks and two zero-crossings of the ultrasonic pulse were ignored (see section 4.4.1) and the following four peaks and four zero-crossings were digitised and used for the computation of the velocity and attenuation of the ultrasonic signal, respectively.

The velocity results obtained with this technique were very inconsistent. One of the possible causes for the variations in the velocities obtained could have been the fluctuations in temperature of \(\pm 1 ^\circ C\). In the light of later experience it was decided to discard these results.

The attenuation results obtained with the method in discussion were considered to be consistent and are presented in chapter 7. The temperature effect on attenuation is discussed in sections 4.3.5 and 7.5, and was not thought to be significant within the fluctuation of \(\pm 1 ^\circ C\) as in the case of the velocity.

4.4.1.2 - TONE BURST EXCITATION

The basic difference in the experimental set up for this technique compared with the one described in the previous section, was the block involving a 20 dB attenuator, a pulse generator and a function generator monitored by a frequency meter, as shown in figure 4.4.

The transmitter driver with its output attenuated by 20 dB was used as the external trigger for the pulse generator whose output gated the function generator. The resulting tone-burst wave was used to excite the transmitting transducer Tx.
FIGURE 4.4: Ultrasonic velocity and attenuation measurement system with tone-burst excitation.
A sine wave tone-burst of approximately 3 μs in duration was used at a repetition rate of approximately 0.5 kHz. The frequency range used was from 1.5 MHz to 8 MHz, in steps of 0.5 MHz. Two sets of measurements were taken (see table 4.1): [a] with the transducers separated by 200 mm, the frequency range was 1.5 to 3 MHz; and [b] with the transducers separated by 500 mm, the frequency range was 3 to 8 MHz. In both cases, the samples were positioned half way between the transmitter and the receiver. The transducers used were unfocussed, having 0.5" (12.7 mm) element diameter and a nominal fundamental resonance frequency of 7.5 MHz (Panametrics, Acuscan Immersion Transducers, ref. number V-320). The characteristics of the pair Tx-Rx in the frequency range of interest is presented in chapter 5.

The received signal at Rx was fed into a Metrotek wide band RF receiver amplifier (MR-101) via its calibrated attenuator and 0.5 MHz high pass filter. The remainder of the diagram of figure 4.4 is described in section 4.4.1.

The first four peaks and four zero-crossings were ignored (see section 4.4.1) and the following two peaks and two zero-crossings of the ultrasonic pulse were digitised and used for the required computation of attenuation and velocity, respectively. Two velocities were simultaneously computed: one from the start of pulse (detected pre-set threshold level at the beginning of the pulse, see section 4.4.1); and the other from the two digitised zero-crossings. The former was named signal velocity and the latter the zero-crossing velocity. The velocity results obtained on some of the samples are presented in chapter 5, whereas the attenuation results are presented in chapter 7.
4.4.2 - TIME INTERVAL AVERAGING

This method is concerned with ultrasonic velocity only. It was used for measuring both the velocity in the medium in which the sample was immersed, and the velocity in the sample itself.

The ultrasonic velocity in the samples and in water were measured in the range of 1.5 - 8.0 MHz, in steps of 0.5 MHz.

The block diagram given in figure 4.5 shows the system used. A function generator (30 MHz Wavetek, model 164) was operated on gated mode. The pulse repetition rate (about 1.3 kHz) and the gate length (approximately 3 μs) were set up at the pulse generator (5 Hz-5 MHz Thandar, model TG-105). The pair of transducers used (Panametrics V-320) were unfocussed and had a nominal frequency of 7.5 MHz and 12.7 mm diameter plane diiss. The tone-burst generator was used to drive the transmitting transducer, Tx, and at the same time to start the counter timer (100 MHz universal counter timer, Marconi Instruments, model 2437). After the received signal at the receiving transducer, Rx, had been electronically attenuated (at certain frequencies, to avoid overloading the RF amplifier) and amplified, it was fed into the "stop-channel" of the counter timer, displaying the time interval averaging between the start and the stop signals. A trigger level was set on both "start" and "stop" channels of the counter timer. The start and stop channels were set to trigger at 0 V and 40 mV, respectively. The amplitude of the signal used to excite the transmitter was ±6.8 V. The amplitude of the first positive peak of the received signal was kept below 40 mV, by means of the variable attenuators, so that it would always be ignored by the timer. The second positive peak of the received signal was kept at 60 mV. This was found to be suitable to avoid the initial transient of the pulse generated at the gating.
FIGURE 4.5: Time interval averaging system for velocity measurements.
The push button controls of the timer were selected in a way that 1000 intervals would be averaged, giving a resolution of approximately 3.3 ns, the average being displayed at every 10 seconds.

The temperature of the water bath was kept at \((20.0 \pm 0.1) ^\circ C\) by means of a water circulator and stirrer. The temperature was firstly read from an ordinary mercury thermometer and secondly monitored ultrasonically by the time of flight of an independent pair of 2 MHz transducers (see section 4.3.6).

In this experimental set up, the Tx-Rx separation used was 500 mm, and the sample was placed at approximately 300 mm from the transmitter source. The velocity in the reference medium (distilled water) was measured according to the piecewise method described in section 5.3.1, in three intervals of 100 mm each. The origin of measurements was at \(Tx-Rx = 200 \text{ mm}\), and the frequency used was 8 MHz. These measurements were all carried out in a water tank with the receiver mounted on a tracking mechanism under micro-computer control by means of a stepping motor. The reproducibility of these measurements of velocity of ultrasound in water was within 0.1%.

A computer programme ("sampvel", written in pascal language) was used to investigate the diffraction effects on velocity in water for the pair of 7.5 MHz transducers (see chapter 5) and to evaluate the velocity in the samples, accordingly.

On measuring the velocity of ultrasound in the specimen, the programme requested the time of flight with and without the specimen in the acoustic beam. These times were displayed directly on the counter timer, at each driving frequency. The velocity in the sample was immediately computed. The relevant data were stored in a disk file for subsequent analysis.

Some preliminary measurements were taken in order to see if the
relative position of the sample with respect to the transducers would affect the results. These are presented in chapter 6. They had shown very little variation, i.e., within the calculated error bars of measurements. The error analysis for these measurements was similar to that discussed extensively in chapter 5. However, as ideally one should have plane waves entering the sample, it was decided that, from a practical point of view, the samples would be positioned at about 300 mm from the transmitter.

4.4.3 - TIME DELAY SPECTROMETRY

The Time Delay Spectrometry (T.D.S.) technique for acoustics applications, appears to have been devised by Heyser in 1967, to make anechoic acoustical measurements on audio transducers. Its adaptation to medical ultrasound systems has been published elsewhere (Heyser & Le Croissette, 1974). The technique itself, and the time and frequency relationships involved for attenuation and velocity measurement in biological tissue, is well covered in a later publication (Le Croissette & Heyser, 1976). Further studies were carried out by the same team (Le Croissette et al., 1979; Gammell et al., 1979) and by others (Crosby & Mackay, 1978; Chivers et al., 1985). The T.D.S. technique was used in this investigation for measurements of ultrasonic attenuation in the samples described in chapter 3. Measurements of velocity were not possible due to practical constraints related to technical limitations of the instrument used.

Time delay spectrometry utilizes an ultrasonic transmission signal repetitively swept in a linear manner over a pre-selected frequency range. There is a time delay associated with the transmission time through the propagation medium. If no velocity dispersion occurs, the
transmission time is constant throughout the sweep. The signals arriving at the receiver by various paths can be distinguished by their time delay, which, for a linear sweep, is proportional to the difference between the received frequency and the frequency of the transmitter. The frequency pass-band of the receiver is, therefore, tracked in a coherent manner to follow the source with the time delay appropriate to accept only those signals delayed by the pre-set time delay, i.e., the signals coming from a selected path. The first signal received is normally that via the direct path from transmitter to receiver. Therefore, by using this technique, one can discriminate against signals with different arrival times, especially those due to reverberation and multipath propagation. Upon emergence from the specimen, the frequency components with a given time delay (frequency offset) are reassembled to yield the frequency spectrum. This is the time-delayed spectrum after which the process is named (Heyser & Le Croissette, 1974). This can be achieved by a narrow bandwidth signal which may, in turn, be passed through a narrow pass-band filter in the receiver to remove those signal components that did not arrive with the appropriate time delay, within the limits of the experimental equipment.

For the case of a single medium (water) between the transducers, the frequency offset is given by

$$f_r - f_r = \frac{z}{c} \cdot \frac{df}{dt} \quad (4.6)$$

where $f_r$ is the frequency of the transmitter, $f_r$ is the central frequency of the receiving filter, $z$ is the transducer separation, $c$ is the velocity of ultrasound in water, and $df/dt$ is the linear frequency sweep rate.

The introduction of a specimen of properties different from those
of water between the transmitter and the receiver has potential implications in terms of the appropriate band-width and frequency offset that are applicable. Suppose that a specimen of thickness $l$ and ultrasonic velocity $c_s$ is introduced into the path. The frequency offset is then

$$f_r - f_s = \left( \frac{z - l}{c} + \frac{l}{c_s} \right) \frac{df}{dt}$$

(4.7)

The frequency shift introduced is then

$$\delta f = \left( \frac{z - l}{c} + \frac{l}{c_s} - \frac{z}{c} \right) \frac{df}{dt}$$

$$= \frac{l}{c_s} \left( 1 - \frac{c_s}{c} \right) \frac{df}{dt}$$

(4.8)

Thus if $c_s/c < 1$, the shift will be positive and therefore a greater frequency difference due to the extra delay will be observed. The requirement for an unchanged filter band-width and frequency offset during measurements is that the frequency shift, $\delta f$, is less than the band-width of the filter.

A Hewlett Packard 3585-A spectrum analyzer, which provides facilities for the T.D.S. technique, was used for this experiment. Figure 4.6 shows a transmitting transducer excited from the swept frequency tracking generator of the HP-3585-A spectrum analyzer, which is controlled by a local micro-processor unit. The output from the receiver (either a transducer nominally identical to the transmitter, or a miniature hydrophone) was fed back to the input of the spectrum analyzer.

This experiment, i.e., measurements of attenuation in the set of samples described in chapter 3 with the T.D.S. technique, was part of a sequential study reported by Chivers et al. (1985). That report presents a detailed analysis on the technique and therefore it will not be reproduced here.
FIGURE 4.6: Block diagram of Time Delay Spectrometry ultrasound. (a) with pair of transducers and (b) with a hydrophone as receiver.
The frequency range was set at 0 to 9 MHz and was swept in 0.8 s (sweep rate $\frac{df}{dt} = 11.25$ MHz s$^{-1}$) with a 3 dB intermediate frequency filter band-width of 300 Hz. The frequency offset was $f_r - f_s = 1.225$ kHz. The above parameters were found to be adequate to eliminate signals which were not travelling along the direct path of the acoustic beam.

The frequency shift introduced by the insertion of a specimen of silicone rubber RTV-602 of thickness 30 mm and velocity 1020 m s$^{-1}$ in water ($c = 1480$ m s$^{-1}$) was $\delta f = 102.8$ Hz, which was well within the 300 Hz filter band-width, and therefore it was not considered to affect the attenuation measurements. The variation of this frequency shift caused by the ultrasonic velocity dispersion (from approximately 1019 m s$^{-1}$ to approximately 1023 m s$^{-1}$) over the frequency range of 0 to 9 MHz was calculated to be approximately 1.3 Hz, which again was well covered by the filter band-width used.

The HP-3585A spectrum analyzer is also provided with a facility for storing two spectra in memory, which can be displayed separately or together on the C.R.T. It also permits the difference between the two spectra, with and without a specimen present, to be displayed. This gives as a result, the continuous frequency dependence of the apparent ultrasonic attenuation, i.e., uncorrected for diffraction effects and reflections at the interfaces, of the medium being studied, provided that a logarithmic display was used. Furthermore, any spurious signals due to secondary modes of vibration of the transducers are eliminated.

Two sets of measurements of attenuation were taken on each sample across the nominal thickness of 30 mm (see table 4.1). One of them utilized a pair of heavily damped transducers, nominally 7.5 MHz, unfocussed and with circular disk elements of 0.5" (12.7 mm) diameter (Panametrics, Acuscan Immersion Transducers, ref.No.V-320). The
other set of measurements had a wide-band miniature hydrophone (H-405) of 1 mm diameter as receiver. For both sets of measurements, the separation between the transmitter and the receiver, immersed in water, was 108 µs ± 0.1 µs (about 160 mm) at a temperature of 16.8°C. This distance was used since the equipment had been set up previously for measurements on hydrophones (Chivers et al., 1985) as being the optimal obtainable within the instrumental constraints involved.

Figure 4.6.a and 4.6.b illustrate the experimental set up for the pair of 7.5 MHz transducers (Tx: transmitter and Rx: receiver) and for the hydrophone (Hx) as receiver, respectively. The data were recorded on a X-Y recorder (Bryans, model 29000-A4) connected directly to the spectrum analyzer, both with compatible scale calibrations and later digitised onto a computer for easier comparison with the other techniques used. These results are presented in chapter 7 of this thesis.

The samples were aligned in the acoustic field so as to obtain the maximum amplitude of the received signal as displayed on the C.R.T. The samples were positioned at approximately 30 mm from the receiving transducer. It was observed that by maximising the received signal amplitude at a frequency of about 5 MHz (the alignment procedure used) the samples faces were not always parallel to the faces of the transducers elements. This occurred for both transducer arrangements of figure 4.6. The causes and effects of this deviation from normal incidence (5° to 15°, assessed by eye) of the sample in the acoustic field are discussed in chapter 7.
4.4.4 - **SWEPT FREQUENCY TECHNIQUE**

The swept frequency technique, like the T.D.S. technique, sweeps a sinusoidal signal in a linear manner over a selected range of frequencies. However, the swept frequency technique differs fundamentally from the time delay spectrometry in terms of the frequency offset and the filter band-width.

This technique was used to measure attenuation in the set of specimens described in chapter 3, in an experiment performed in parallel with the measurements of velocity and attenuation obtained by the use of the digitised tone-burst technique described in section 4.4.1.2. Therefore, these two techniques had the same alignment procedure for the transducers and the sample, as well as the sample treatment (see section 4.3). The temperature of the water bath was set at 20.0±0.1°C, monitored and controlled by the method discussed in section 4.3.6.

A spectrum analyzer (Marconi Instruments, TF-2370) was used for the purposes of this technique. Figure 4.7 shows a block diagram of the experimental set up involved in this investigation of ultrasonic attenuation as a continuous function of frequency.

The spectrum analyzer itself was provided with a tracking generator which outputs a sinusoidal signal (at a level of -10 dBm when terminated in 50 ohms) varying in frequency according to the user's selected range. In this experiment, the range of 0 to 11 MHz was used, and the swept signal was applied to the transmitting transducer. The swept signals sensed by the receiving transducer were fed into the receiver of the spectrum analyzer. The display then showed the apparent characteristics of the specimen under test in terms of a frequency response (uncorrected for diffraction effects and reflections at the specimen interfaces).
FIGURE 4.7: Swept Frequency Technique for ultrasonic attenuation measurements.
Unlike the HP-3585A spectrum analyser used for the T.D.S. technique experiment, the TF-2370 had no frequency offset facility. A built in receiving filter was swept in synchronism with the tracking generator, and in order to achieve sensible resolution it was then necessary to employ as low a sweep rate as possible. A -3 dB filter of band-width 5 kHz (the narrowest available for the selected frequency range) was used and swept at the sweep speed of 0.24 MHz s^{-1} (the slowest available for that span). An X-Y chart recorder facility (Bryans, model 29000-A4) was linked with the spectrum analyser to record the frequency spectra.

Two series of measurements were taken for the complete set of samples, firstly with the pair of transducers spaced by about 200 mm and secondly by about 500 mm (see table 4.1). In both arrangements, the intervening sample was placed half way between the transducers.

The vertical scale of the display was set to be logarithmic, so that the apparent attenuation (uncorrected for diffraction) at any particular frequency was found from the difference between the two spectra, with and without the intervening sample. The two series of frequency-dependent attenuation are presented in chapter 7, where the digitised points are plotted together with the other techniques results and the theoretical model prediction.

Some preliminary studies, related to diffraction effects on attenuation measurements, involved the use of this technique and are presented in chapter 5 (section 5.6) and in chapter 7. This technique was also employed in the measurement of the frequency spectra of the Panametrics and the T.M.S. transducers used in the investigation reported in this thesis. These frequency spectra of the transducers are presented and discussed in chapter 5 and in appendix A.
CHAPTER 5

DIFFRACTION EFFECTS OF A PAIR OF TRANSUCERS ON ULTRASONIC VELOCITY AND ATTENUATION MEASUREMENTS
5.1 - INTRODUCTION

A series of four experiments was designed in order to investigate a discontinuity in the ultrasonic velocity as function of frequency, obtained for the samples of silicone rubber with glass beads in suspension. That discontinuity was named the velocity 'hop'. The magnitude of the velocity hop varied from sample to sample and was less than ±0.3%, i.e., less than ±3 m s⁻¹. It was observed when the distance between the pair of transducers (Tx-Rx) was changed from approximately 200 mm to 500 mm. This change in Tx-Rx separation was an attempt to operate in the far field region of the acoustic beam, and at the same time to avoid reflections of the ultrasonic waves at the lateral surfaces of the specimen and its original holder, while investigating the ultrasonic properties of the whole set of samples over the range of 1.5 MHz to 3 MHz (for Tx-Rx = 200 mm) and 3 MHz to 8 MHz (for Tx-Rx = 500 mm). The measurements of velocity of ultrasound were taken with the technique described in section 4.4.1.2 (digitised tone burst).

To illustrate the velocity hop, four samples with typical results were selected for presentation here: the two pure silicone rubber, sample 1 and 20 (group I), sample 2 (group II) and sample 27 (group V). Samples 1 and 20 did not show a clear velocity hop, but will be used in this investigation as reference samples. Sample 2 had a negative
velocity hop and sample 27 had a positive one (see figure 5.1). These four samples were used for the investigation of the velocity hop according to the sequential study outlined in table 5.1.

In figure 5.1, the asterisk represents the zero-crossing velocity and the open circle represents the signal velocity. The difference between these two measured velocity is discussed in section 4.4.1.

The first of the four experiments of table 5.1 was concerned with diffraction effects on the ultrasonic velocity. The aim of this work was to investigate experimentally the change in the measured velocity of the ultrasonic beam between a pair of transducers (Panametrics, V-320, 7.5 MHz, 0.5" diameter, unfocussed) due to diffraction and to compare the results with the theory. The remainder of the experiments of table 5.1 are presented in chapter 6.

Some initial theoretical calculations intended to determine whether or not the velocity hop was due to diffraction, produced ambiguous answers. Therefore, it was necessary to proceed with the experiment in order to find out if diffraction phenomena could have contributed to the "hop" in velocity in the samples.

Similarly, some studies on diffraction effects on attenuation measurements were performed. These are discussed in section 5.6 and in chapter 7, which also followed the outline of table 5.1.
FIGURE 5.1.a: Hop in ultrasonic velocity in samples as a function of frequency. * - zero-crossing velocity, o - signal velocity. (see also figure 5.1.b).
FIGURE 5.1.b: Hop in ultrasonic velocity in samples as a function of frequency. * - zero-crossing velocity, o - signal velocity. (see also figure 5.1.a).
**TABLE 5.1**
Approach to understand the velocity-hop
(at a constant temperature)

<table>
<thead>
<tr>
<th>ORDER</th>
<th>FIX</th>
<th>VARY</th>
<th>MEASURE</th>
<th>OBJECTIVE</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>f</td>
<td>Tx−Rx</td>
<td>Cw</td>
<td>For a given frequency, to find out how the measured velocity depends on the transducer separation. (Compare with ideal diffraction corrections). [In chapter 5].</td>
</tr>
<tr>
<td></td>
<td></td>
<td>water</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td></td>
<td>Tx−Rx</td>
<td>f</td>
<td>Cs</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sz</td>
<td></td>
<td>To measure the velocity in a sample covering the whole range of frequency (1-8 MHz) without changing Tx-Rx separation. [In chapter 6].</td>
</tr>
<tr>
<td>3</td>
<td></td>
<td>Sz</td>
<td>f</td>
<td>Cs</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>To find out whether the measured velocity in the sample depends on Tx-Rx separation. [In chapter 6].</td>
</tr>
<tr>
<td>4</td>
<td></td>
<td></td>
<td>f</td>
<td>Cs</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Sz</td>
<td>Cs</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>To find out whether the measured velocity in the sample depends on its relative position in the acoustic field (longitudinal axis dependence). [In chapter 6].</td>
</tr>
</tbody>
</table>

**keywords to table 5.1**

- f: frequency
- Tx-Rx: transducer separation (on longitudinal axis)
- Sz: relative position of Sample S along the longitudinal axis z
- Cw: ultrasonic velocity in water
- Cs: ultrasonic velocity in sample
5.2 - LITERATURE REVIEW

It seems that studies available in the literature on effects of diffraction phenomena of ultrasonic waves propagating in a medium, are mostly concerned with relative loss of signal necessary for correction of attenuation measurements rather than with the phase-shift, necessary for correction of velocity measurements. However, studies on the variation of velocity due to phase-shift caused by diffraction alone are even more scarce and some of the existing ones are not very clear. For this reason, the review presented here will be concentrated mainly on the diffraction effects on velocity measurements.

Gitis and Khimunin (1969) reviewed the effects of diffraction in ultrasonic measurements for the various geometrical and ultrasonic parameters involved. Lord (1966) calculated that the measured phase velocity of a monochromatic continuous wave would be in excess of that for plane wave propagation, where the average phase caused by geometrical diffraction is positive and is in addition to the true plane-wave value, yielding, therefore, an advance in phase as a result of interference effects. However, in the case of the elastic disturbance being in the form of a pulse of large frequency spectrum, Lord also calculated that the group velocity would be lower than the plane-wave velocity.

Ilgunas et al. (1964) measured the velocity in water, at different frequencies of low megahertz by an interferometric method. They were particularly concerned with the ratio of the diameter of the crystal (transceiver) to the wavelength (d/λ) as well as size of the reflector (D/λ) to be used. They showed that the measured velocity tends to a plane wave value for d/λ > 50, and is exponentially in excess of that value for smaller ratios. They also concluded from their experiment, that only a planar field ensures that the reflector will not influence
the result, even for diameters much less than that of the crystal. Barshauskas et al. (1964) continued the work of Ilgunas et al. (1964) on measurement of dispersion in liquids. They have also pointed out some reports which claimed to have measured a negative dispersion in distilled water and other liquids. That observed effect was in fact associated with diffraction phenomena. They related the velocity of an ideal plane wave \( (c_\infty \text{ at } d/\lambda = \infty) \) to be determined by

\[
c_\infty = c - c_\infty \Delta c_\omega/c_\omega,
\]

where \( c_\infty \) and \( c_\omega \) are the measured values of the speed of ultrasound in the liquid investigated and in water, respectively, and \( \Delta c \) is the diffraction increment of the velocity in water, all at the same \( d/\lambda \) ratio. If \( c = c_\omega \), that equation becomes \( c_\infty = c_\omega - \Delta c_\omega \), meaning that the measured velocity, \( c_\omega \), is greater than the plane-wave value by the diffraction increment, because \( \Delta c_\omega \) was shown to be positive.

McSkimin (1960 and 1961b) used a pulse overlap method to determine the effect of diffraction on the phase velocity of propagation of high frequency ultrasonic waves. He investigated the dependence of the velocity as a function of \( d/\lambda \), of frequency and of distance traversed by a short ultrasonic train of waves. He also experimentally determined, by the phase comparison of reflected longitudinal waves in fused silica blocks, that the effect of diffraction is an increase in the measured velocity over that of the plane wave.

Papadakis (1966 and 1975) presented an analysis of diffraction loss and phase change in isotropic and anisotropic materials, from single apertures (of several wavelengths across) with particular emphasis on the case of pulse-echo systems. Papadakis (1967) used that analysis of diffraction phase corrections to determine the ultrasonic phase velocity by a pulse-echo-overlap method. The phase change increases monotonically, but nonlinearly, to a limit of \( \pi/2 \) as the distance
increases from zero to infinity. The diffraction loss fluctuates with the increase of the distance traversed before becoming monotonic increasing logarithmically. As a function of distance, however, the phase presents plateaus where the loss presents peaks. A variation in the measured time of flight, Δt, in measurements of phase-velocity may be caused by the phase increment as given by the following equation:

\[ Δt = \frac{\Delta φ}{2\pi f} \]  

(5.1)

where Δφ is the phase increment in radians at a given interval of distance and f is the frequency. The time of flight corrected from diffraction effect, i.e., the plane wave travel time, \( t_{m}^{(p)} \), is the result of the measured time of flight, \( t_{m} \), added to Δt. Thus,

\[ t_{m}^{(p)} = t_{m} + Δt \]  

(5.2)

Equation (5.1) shows quite clearly that the time correction, at a given distance, decreases with increasing frequency.

For a given distance z (or Tx-Rx) between the transmitter and the receiver, the plane wave velocity (the corrected velocity) is given by

\[ c_{m}^{(p)} = \frac{z}{t_{m}^{(p)}} \]

\[ = \frac{z}{(t + Δt)} \]

or

\[ c_{m}^{(p)} = \frac{z}{t} \cdot \frac{1}{[1 + (Δt/t_{m})]} \]  

(5.3)
By expanding eq. (5.3) in terms of binominal series, it becomes:

$$c_{m}^{(p)} = \frac{z}{t_{m}} \left[ 1 - \frac{\Delta t}{t_{m}} + \frac{(\Delta t)^{2}}{(t_{m})^{2}} - \cdots \right] -1 < (\Delta t/t_{m}) < 1 \quad (5.4)$$

Numerically, the terms of order greater than 1 can be neglected. Therefore, eq. (5.4) can now be written in an approximate form, i.e.,

$$c_{m}^{(p)} = c_{m} \cdot [1 - (\Delta t/t_{m})] \quad (5.5)$$

The term $c$ of eq. (5.5) is the measured phase velocity and $c_{m} \cdot \Delta t/t_{m}$ is its correction to obtain the plane wave value, $c_{m}^{(p)}$. Substituting $c_{m} \cdot \Delta t/t_{m}$ by the symbol $c$, eq. (5.5) can then be written in a simpler manner,

$$c_{m}^{(p)} = c_{m} - \Delta c \quad (5.6)$$

Therefore the true phase velocity is less than the measured value by an amount $\Delta c$. Papadakis (1966 and 1975) also explained that because the slope in the function of phase versus distance traversed fluctuates between zero and fairly large numbers, the group velocity can be considerably lower than the phase velocity in the Fresnel region and that this dispersion is caused by diffraction alone, as previously calculated by Lord (1966) and experimentally observed by Barshauskas (1964).

Khimunin (1975) calculated and tabulated the exact diffraction corrections to the results of ultrasound phase velocity measurements for ideal (piston-like) circular transducers of equal radius. He also discussed the diffraction corrections within the approximations of plane and spherical waves. Incorporated in his discussion is the relationship between the absorption in the medium and the size of the wave number, $k_{0} = 2\pi/\lambda$. He pointed out that the phase-shift hardly changes with variation of the absorption, $\alpha$, in the medium as long as the condition $\alpha \ll k_{0}$ is fulfilled. Also, the phase shift at average pressure for $z = 0$, in general does not equal zero, especially in the
case of small values of the parameter \( ka \) (\( ka < 10 \)), where \( k = k_0 - i\alpha \) is the complex wave number, and \( a \) is the radius of the transducer. He completed that statement by saying that this deviation from zero phase-shift depends on the absorption in the medium under investigation. From his analysis, which agreed with that of Papadakis (1966), he concluded that the nearest approximation of the measured ultrasound velocity to the true value may be obtained under the conditions of formation of either ideal plane waves or ideal spherical waves.

A computer program which calculates the diffraction loss and the phase-shift due to diffraction phenomena, using a numerical model approach, was already available at the time this investigation was initiated (program written by Dr. R.A. Bacon). The model utilises the algorithm given by Rhyne (1977), but permits assessment of effects such as the intrinsic attenuation and velocity in the medium, as well as the size of the discs and their lateral alignment simultaneously, for piston and non-piston like sources. For this investigation, only piston-like transducers will be considered. The model is fully discussed by Bacon & Chivers (1981), and will, therefore, not be repeated here. However, they defined phase as \( \pi/2 \) radians at the surface of the transmitter (zero separation) and phase as zero radians at infinity, whereas Lord (1966), Papadakis (1966, 1967 and 1975), Gitis and Khimunin (1969), Khimunin (1975) and Rhyne (1977) defined phase zero radians at zero distance and \( \pi/2 \) at infinity. The computation of the change in velocity was to follow the analysis of Papadakis (1966 and 1975) discussed previously in this section. The numerical model of Bacon & Chivers (1981) was then modified to have the phase-shift computation compatible with the aforementioned publications. Calculation of the expected phase velocity due to the
phase-shift was also implemented on the programme. Figure 5.2 shows the variation of diffraction loss and phase as a function of the normalised distance, \( S = z \lambda / a^2 \), for a pair of piston-like transducers of equal diameter and coaxially aligned in water. The position of the minimum \( \Delta \) shown in figure 5.2 is given by \( S_\Delta = 0.8/(0.5 - b) \), where \( b \) is the anisotropy parameter for longitudinal wave propagation (Papadakis, 1975). In the case of water, \( b = 0 \) (isotropic medium), which gives \( S_\Delta = 1.6 \). The theoretical results of the "apparent" phase velocity and respective phase-shift at four different frequencies (1 MHz, 1.5 MHz, 3 MHz and 8 MHz) are presented in figure 5.3, for a pair of piston-like transducers, of radius 6.35 mm, immersed in water. The theoretical diffraction correction of the phase velocity was calculated in respect to the value of planar wave velocity of 1480.8 m s\(^{-1}\), measured at 20.0 ± 0.1°C in the far field at 8 MHz.
FIGURE 5.2: Theoretical relationship between ultrasonic phase-shift and loss of signal amplitude due to diffraction for a pair of piston like transducers.
FIGURE 5.3.i: Theoretical variation of phase and 'apparent' phase velocity as a function of distance between the transmitter and the receiver. Radius of transducers $a = 6.35 \text{ mm}$, plane wave velocity $c = 1480.8 \text{ m s}^{-1}$, (a) at 1 MHz, and (b) at 1.5 MHz. (see also figure 5.3.ii).
FIGURE 5.3.ii: See figure 5.3.i. (c) at 2 MHz, and (d) at 8 MHz.
5.3 - VELOCITY MEASUREMENTS

This experiment was performed in a water tank, with the receiving transducer positioned with its face parallel to and coaxial with the transmitting transducer. The successive change of position of the receiver was achieved with the use of a stepping motor mounted on a tracking mechanism. The tracking mechanism was micro-computer controlled. The same computer programme ("sampvel") used for operating the stepping motor also acquired the experimental data and stored it on a disk file. The data was processed afterwards with the use of another programme ("backvel"). This investigation employed the time interval averaging technique discussed in section 4.4.2 and illustrated in figure 4.5.

The time of flight of the first zero-crossing (end of the first half-cycle) of the ultrasonic tone burst pulse (of length approximately 3 𝜇s) was read on a digital counter timer for each longitudinal separation of the transmitter and receiver pair (Tx-Rx).

The alignment of the transducers was made from the procedure outlined in section 4.3.1. The temperature of the water bath was monitored ultrasonically (section 4.3.6) and kept at 20.0 ± 0.1°C.

The measurements of velocity in water were performed for the transducer separation in the range of 0 mm to 580 mm. The measured velocity at every separation was computed by two different methods: piecewise and non-piecewise method (Rider, 1984). Both methods used the same experimental set up and data, but with the data processed accordingly. A discussion on these methods is given below.

This approach, illustrated in figure 5.4, removes any systematic error, which includes the travel time through the protective layers of the transducers, the extra (unknown) distance introduced by those layers, and any extra delay generated by the electronic circuitry.
However, error in distance and time measurement may not be always systematic and, therefore, would not be always suppressed by this approach. This will be later discussed together with the error analysis of the measurements.

To check the dependence of the ultrasonic velocity as a function of transducer separation, Tx-Rx, and direction of data processing, the experimental and theoretical data were processed as symbolised by the arrows in figures 5.5 to 5.8: $\rightarrow$ from the near field to the far field, and $\leftarrow$ from the far field to the near field. From the diffraction theory of phase shift, the values of piecewise phase velocity and non-piecewise phase velocity were calculated for the transmitter and the receiver being separated by steps of 1 mm.
FIGURE 5.4: Piecewise and non-piecewise method of measurement of ultrasonic velocity in water. $T_x$: transmitter, $R_x$: receiver, $z_i$ and $t_i$ are distance and time of flight, $a$ is the radius of the transducers, and the arrows indicate the direction of data processing, i.e., from near-field to far-field, or from far-field to near-field.
5.3.1 - PIECEWISE METHOD

The piecewise method considers the time difference between two consecutive measured times of flight for the receiver in any two particular positions of the acoustic field. In other words, the piecewise method does not have a fixed origin of time of flight and position. It relates the Tx-Rx separation and time of flight with the previous separation and its time of flight.

In figure 5.4, Tx is the transmitter and Rx is the receiver at a position $z_{n-1}$ and $z_n$ with time of flight $t_{n-1}$ and $t_n$, respectively, and so on.

Thus, the measured piecewise velocity in the interval $z_{n-1} - z_n$ is:

$$c_{n-1\rightarrow n} = \frac{Z_n - Z_{n-1}}{t_n - t_{n-1}} \quad (5.7.a)$$

Similarly, the velocity in the interval $z_n - z_{n+1}$ is:

$$c_{n\rightarrow n+1} = \frac{Z_{n+1} - Z_n}{t_{n+1} - t_n} \quad (5.7.b)$$

Figure 5.5 illustrates the theoretical relationship between the phase velocity and the piecewise velocity in water at 1 MHz (1 mm step separation). With the use of the approach of direction of data processing, as indicated by the arrows, the piecewise velocity is shifted by one step along the Tx-Rx separation axis, because its value is plotted at the position $z_n$ for the interval $z_{n-1} - z_n$, and at $z_{n+1}$ for the interval $z_n - z_{n+1}$.
FIGURE 5.5: Theoretical relationship between phase angle, phase velocity and piecewise velocity in water as a function of transducer separation. (a) Normalised distance to $a^2/\lambda$ (b) Non-normalised distance. Step separation = 1 mm, frequency = 1 MHz, radius of transducers $a = 6.35$ mm, plane wave velocity $c = 1480.8$ m s$^{-1}$, absorption $\alpha = 2.4 \times 10^{-4}$ dB mm d/λ = 8.6.
With the piecewise method of measurements of velocity, the magnitude of the error bars may be relatively large due to the high sensitivity of the method, because of the use of a non-fixed origin, and particularly when measurements are taken within small intervals of distance. A detailed discussion on the errors involved is given in section 5.4.

Typical variations of the measured piecewise velocity in water at the frequencies of 1.5 MHz, 3 MHz and 8 MHz are presented in figures 5.6 to 5.8. The respective values of \( \frac{d}{\lambda} \) were approximately 12.9, 25.8 and 68.7, where \( d \) is the geometrical diameter of the transducers and \( \lambda \) is the wavelength of the plane wave. The error bars indicated in those figures represent the reproducibility of measurements obtained in three consecutive days, for the same water in the tank.

5.3.2 - NON-PIECEWISE METHOD

Similarly to the piecewise method, the non-piecewise method takes time interval and distance difference to compute the ultrasonic velocity. Nevertheless, the non-piecewise method considers a fixed origin of distance and time of flight.

Let the origin be at the position \( z_{n-1} \) in figure 5.4. Thus, the non-piecewise velocity measured in the interval \( z_{n} - z_{n-1} \) is:

\[
C_n = \frac{z_n - z_{n-1}}{t_n - t_{n-1}}
\]  

(5.8.a)

Similarly, in the interval \( z_{n+1} - z_{n-1} \), the velocity is

\[
C_{n+1} = \frac{z_{n+1} - z_{n-1}}{t_{n+1} - t_{n-1}}
\]  

(5.8.b)

Figures 5.6 to 5.8 show typical results of the variation of the measured non-piecewise ultrasonic velocity as a function of transducer separation, at the frequencies of 1.5 MHz, 3 MHz and 8 MHz. The error
bars indicated in those figures represent the reproducibility of measurements obtained on three consecutive days, for the same water in the tank. It can be noticed quite clearly that the effects of diffraction are masked, although not totally, by processing the data in the $\leftarrow$ direction. The non-piecewise velocity is the average velocity over the transducer separation. When the direction of the data processing is from the far field to the near field, the value of the plane-wave velocity weighs against the diffraction effects. Consequently, these effects may be masked. On the other hand, by processing the data in the $\rightarrow$ direction, the diffraction effects can be detected, although they were not of the same magnitude as that calculated for the ideal piston-like model.
FIGURE 5.6.i: Predicted phase velocity and piecewise velocity compared with measured piecewise velocity in water at 20°C and 1.5 MHz (d/λ = 12.9). The error bars represent the reproducibility of measurements achieved. The arrows indicate the direction of data processing.
FIGURE 5.6.ii: Predicted phase velocity and non-piecewise velocity compared with the measured non-piecewise velocity in water at 20 °C and 1.5 MHz (d/λ = 12.9). The arrows indicate the direction of data processing and the error bars represent the reproducibility of measurements achieved.
FIGURE 5.7.i: See figure 5.6.i. Frequency = 3 MHz (\(d/\lambda = 25.8\)).
FIGURE 5.7.ii: See figure 5.6.ii. Frequency = 3 MHz (d/λ = 25.8).
FIGURE 5.8.1: See figure 5.6.1. Frequency = 8 MHz. (d/λ = 68.7).
FIGURE 5.8.i: See figure 5.6.ii. Frequency = 8 MHz. (d/λ = 68.7).
5.4 - ERROR ANALYSIS

The errors of measurement involved in this kind of experiment are introduced by distance, time, temperature, age of water, turbulence in water, alignment of transducers, and electronic instability.

An equation that gives the error in velocity can be written from the basic relation \( c = \frac{d}{t} \), where \( c \) is the ultrasonic velocity, \( d \) is the distance and \( t \) is the time of flight.

Let \( \xi_d \) and \( \xi_t \) be the errors in distance and time of flight. Thus, "the most probable error" in \( c \), i.e., the most probable value of \( \xi_c \) (Topping, 1979) is given by

\[
\xi_c^2 = \left( \frac{\partial c}{\partial d} \right)^2 \xi_d^2 + \left( \frac{\partial c}{\partial t} \right)^2 \xi_t^2
\]  \hspace{1cm} (5.9.a)

or

\[
\frac{\xi_c^2}{c^2} = \frac{\xi_d^2}{d^2} + \frac{\xi_t^2}{t^2}
\]  \hspace{1cm} (5.9.b)

Nevertheless, this study employed differential methods such as the piecewise and the non-piecewise methods described previously and expressed by the equations (5.7) and (5.8). Suppose two distinct transducer separations being taken at \( d_1 \) and \( d_2 \) with respective times of flight \( t_1 \) and \( t_2 \). The velocity in the interval \( (d_2 - d_1) \) is then calculated as:

\[
c = \frac{d_2 - d_1}{t_2 - t_1}
\]  \hspace{1cm} (5.10)

However, supposing that the errors in the above quantities are \( \pm \xi_{d_1}, \pm \xi_{d_2}, \pm \xi_{t_1} \) and \( \pm \xi_{t_2} \), the most probable error in \( c \) is then given by

\[
\xi_c^2 = \left( \frac{\partial c}{\partial d} \right)^2 \xi_{d_2}^2 + \left( \frac{\partial c}{\partial d} \right)^2 \xi_{d_1}^2 + \left( \frac{\partial c}{\partial t} \right)^2 \xi_{t_2}^2 + \left( \frac{\partial c}{\partial t} \right)^2 \xi_{t_1}^2
\]  \hspace{1cm} (5.11)

This expression can be finally written in a simpler form, i.e.,

\[
\xi_c = \pm c \left[ \frac{\xi_{d_2}^2 + \xi_{d_1}^2}{(d_2 - d_1)^2} + \frac{\xi_{t_2}^2 + \xi_{t_1}^2}{(t_2 - t_1)^2} \right]^{1/2}
\]  \hspace{1cm} (5.12)
5.4.1 - Error in Distance Measurement

The distance travelled by the receiving transducer held by the tracking mechanism is dependent on the pitch of its driving screw thread, which automatically sets the transducer separation. An experiment was performed in order to check the pitch of the screw thread against its nominal value of 1 mm.

A dial gauge mounted on the tracking mechanism was used for this assessment and two sets of readings were taken. The accuracy of this distance measurement system was better than 0.5 μm. The mean value and its standard deviation (82 observations) obtained in the range of 0-410 mm were 0.99925 mm and 0.00082 mm, respectively. Similarly, the mean value and its standard deviation (34 observations) obtained in the range of 410-580 mm were 0.99850 mm and 0.00085 mm, respectively. The error in distance measurement was the major contributor to errors in velocity measurement, as it will be demonstrated below. It must be noted that if the distance is under-estimated, the velocity will be over-estimated.

Supposing there was no error in time of flight measurement, i.e., \( \varepsilon_1 = \varepsilon_2 = 0 \) s, and a plane wave velocity of 1480 m s\(^{-1}\) in eq.(5.12), the typical error in the piecewise velocity due to an error in distance of 0.8 μm/mm would then be ±1.18 m s\(^{-1}\). The same order of magnitude of errors was obtained for the non-piecewise velocity.

Because of the high sensitivity of the piecewise method, the results are subjected to scatter more than for the non-piecewise method. The piecewise method does not have a fixed origin and this contributes largely to errors of measurements. Ill-positioning of the moving transducer can occur randomly, and is difficult to assess.

The back-lash of the tracking mechanism was also measured (0.43 mm) by using the same technique described above, and avoided as a source of
error in distance because the direction of measurement was not changed during the experiment.

In order to reduce the errors in distance, another type of tracking mechanism would be required. This could be achieved, possibly, by replacing the present driving screw thread by a pair of driving pulleys linked by an inelastic cord.

5.4.2 - ERROR IN TIME MEASUREMENT

The error in time of flight measurement may result from temperature fluctuations, misalignment of transducers, instability of the electronic circuitry, age of the water, and turbulence in the water.

The fluctuations in time of flight during one typical set of measurements, $\varepsilon_{t_1}$, were as large as those shown in table 5.2. The reproducibility obtained in time of flight measurements, $\varepsilon_{t_3}$, at the same frequency and transducer separation, comparing 2 to 4 sets of measurements taken on 3 consecutive days is also shown in table 5.2. The transducers were realigned between each set of measurements to check the reproducibility of the measurements.

<table>
<thead>
<tr>
<th>$f$ (MHz)</th>
<th>$\varepsilon_{t_1}$ (ns)</th>
<th>$\varepsilon_{t_3}$ (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5</td>
<td>0.62</td>
<td>2.8 (4)</td>
</tr>
<tr>
<td>3.0</td>
<td>0.54</td>
<td>2.2 (3)</td>
</tr>
<tr>
<td>8.0</td>
<td>0.52</td>
<td>1.6 (2)</td>
</tr>
</tbody>
</table>

Key words:

- $f$: frequency.
- $\varepsilon_{t_1}$: fluctuation in time of flight in one typical set of measurements.
- $\varepsilon_{t_3}$: reproducibility in measurement of time of flight within 3 days for the same water.

The figures indicated in the parenthesis are the number of sets of measurements taken at that frequency.
To illustrate the order of magnitude of the error in the measured velocity, taking into account the fluctuation in time of flight measurement, only the worst case will be considered here. This was the case of the measurements at 1.5 MHz, as shown in table 5.2, i.e., \( \mathcal{E}_t = 0.62 \text{ ns} \). Let the errors in distance \( \mathcal{E}_d = \mathcal{E}_{\pm d} = 0 \) in eq.(5.12), and consider a plane wave velocity of \( c = 1480 \text{ m s}^{-1} \). The typical error in the piecewise velocity measured in a 1 mm interval was then \( \pm 1.36 \text{ m s}^{-1} \), becoming smaller as the interval was increased. The error in the non-piecewise velocity in the first interval was of the same magnitude as that of the piecewise velocity. When the interval of distance was as large as 500 mm, the error due to fluctuation in timing measurement alone was as small as \( \pm 0.003 \text{ m s}^{-1} \).

The error bars in figures 5.6 to 5.8 are the combinations of the errors in distance, \( \mathcal{E}_d \), and the reproducibility in time of flight, \( \mathcal{E}_t \), at the given frequency, as given by eq.(5.12).

5.4.3 - **Temperature Dependence**

Although the temperature of the water tank was ultrasonically monitored, its magnitude was read from an ordinary mercury thermometer. All ultrasonic measurements were made at \( 20.0 \pm 0.1^\circ \text{C} \).

The temperature dependence of the sound velocity in water is approximately \( 3 \text{ m s}^{-1}^\circ \text{C}^{-1} \) (Greenspan & Tchiegg, 1959 and Del Grosso & Mader, 1972). Consider a variation in temperature by \( \pm 0.1^\circ \text{C} \) and plane-wave propagation. Taking the value of velocity in water as \( 1480 \text{ m s}^{-1} \), the variation in time of flight would then be approximately \( \pm 1.36 \text{ ns} \) in 10 mm interval which is about the order of magnitude of the reproducibility of time measurement within 3 consecutive days of experiments, and about twice as large as that within one typical set of measurements (see table 5.2). Therefore, temperature fluctuations were
thought to have affected partially the reproducibility of measurements, although very little compared with the contribution given by the errors in distance.

5.4.4 - **AGE OF WATER**

In the present work, i.e., the investigation of the diffraction effects of the ultrasonic propagation, the same water in the tank was used for 3 consecutive days. However, the ageing of the water in the acoustic tank was thought, in principle, to cause variation in the velocity of propagation of ultrasound, since the condition and composition of the water may change with the time. The order of magnitude of variation was not known.

To investigate how the ultrasonic velocity in water varies with its age, a dedicated experiment was performed. Extreme care was devoted to keep the electronic and mechanical equipment parameters constant during the entire course of the experiment period (46 days). Figure 5.9 illustrates the experimental arrangement for this particular objective. It is essentially the time interval averaging technique (section 4.4.2). A tone burst pulse signal, of frequency 8 MHz and duration approximately 1.7 μs, was used for this investigation.
FIGURE 5.9: Block diagram for measurement of ultrasonic velocity as a function of age of water.
The start and the stop channels of the 100 MHz counter timer (Marconi Instruments, model 2437) were set to be triggered at a threshold level on the positive slope of the first half cycle of the R.F. pulse, allowing the signal velocity to be measured. The amplitude of the leading peak of the transmitted signal and of the received signals were maintained at 6.8 V and 72 mV, respectively. The room temperature (read at the vicinity of the acoustic tank) was kept at \((20 \pm 2)°C\) to diminish possible expansion (or contraction) of the acoustic tank and transducer holders, keeping the transducers at a stationary position. The transducers were firmly secured at a separation of \(300 \pm 2\) mm. The water surface was covered with expanded polystyrene spheres, and the tank was surrounded with expanded polystyrene boards (about 50 mm thick), in order to reduce heat exchange with the laboratory during measurements, thus, keeping the water at an approximately constant temperature.

The water temperature was maintained at \((20.00 \pm 0.05)°C\) by means of a closed circuit water circulator and a stirrer. Before any measurement of time of flight was taken, the circulator and stirrer were switched off and a minimum of 2 minutes was given to allow turbulence to die away. The temperature was read (just before and after the measurements of time of flight) from a mercury thermometer, with an optical lens system (4x) attached to its gradation.

To accelerate the age (or contamination) of the water (initially distilled water) in the tank (of approximately 50 litres capacity) the author and some of his colleagues regularly immersed their hands in it, even when the hands were relatively dirty. This was considered to be a realistic way of producing contamination in the water since it was of practical means. Many experimental situations, manipulation of transducers, specimens, etc., require the direct insertion of hands in
the acoustic tank when filled with water.

At one occasion (on the 43rd day) an accidental knock (with the hands) occurred against the receiver holder and caused an increase in time of flight by approximately 3 ns. On its merit, this accidental knock served as a test for the mechanic stability of the system.

An experimental recess was made between the 17th and the 35th day, i.e., no man-induced contamination of water was made and no measurement was taken. It was observed that effectively no change in velocity in water had occurred during that period of time.

From the first day of measurement of time of flight of the ultrasonic waves to the last, the colour of the water gradually changed from limpid to cloudy straw (assessed by eye). No change in colour was noticed during the experimental recess (17th to 35th day). The total variation of time of flight over the entire period of 46 days was approximately 100 ns as shown in figure 5.10. In conclusion, the largest variation of velocity of ultrasound was only about 0.74 m s⁻¹, i.e., it increased from 1480.34 m s⁻¹ to 1481.08 m s⁻¹.
FIGURE 5.10: Variation of velocity of ultrasound in water as a function of its age.
5.4.5 - TURBULENCE IN WATER

The time of flight of the ultrasonic waves in water can be upset by turbulence. Turbulence in water in this work was observed when: [a] the water circulator and stirrer, used for temperature control, had just been switched on/off. Measurements were only carried out when the water circulator was switched off, and the water had reached a relatively steady state (assessed visually); [b] the receiving transducer had just been moved from one position to another, in order to change the Tx-Rx separation; and [c] by the vibrations caused by the cooling fans of some of the equipments used, placed on the same bench of the water tank. According to table 5.2, the variation in time of flight due to this sort of disturbance was less than 0.5 ns. However, the magnitude of errors caused by turbulence in water (random error) depends on the relative time interval taken for measurements. Relatively small Tx-Rx separation, or relatively small time intervals are likely to be more sensitive to errors originated by turbulence.

5.4.6 - THRESHOLD LEVEL...VS. TIME OF FLIGHT

The start and the stop of the counter timer of the diagram of figure 5.4 is determined by a pre-set threshold level. This threshold level, $y_0$, is related to the amplitude, $A$, of the wave signal (Chivers, 1984). The ratio $y_0/A$ should be kept constant in order to achieve greater accuracy in time of flight measurement.

The minimum uncertainty arises if the ratio $y_0/A$ is as low as possible. For a given $y_0$ and change in $A$, greater effects are expected at lower frequencies, as also observed experimentally (see section 5.4.2). Assuming $y_0/A \ll 1$, the estimation of error in time measurement, $\Delta t$, at a given frequency, $\omega = 2\pi/\tau$, where $\tau$ is the period, and a variation in amplitude, $\Delta A$, can be obtained from
\[ \Delta t = \frac{-\tau \cdot \frac{y}{A}}{2\pi \cdot \frac{A}{A}} \]  \hspace{1cm} (5.13)

For \( \Delta A/A \approx 1\% \) and \( \frac{y}{A} = 1/10 \), the error in time would be
\[ \Delta t \approx \frac{1}{62.83} \cdot \frac{1}{100} \cdot \tau \]  \hspace{1cm} (5.14.a)

or
\[ \Delta t \approx 1.59 \times 10^{-4} \cdot \tau \]  \hspace{1cm} (5.14.b)

For the case of 1.5 MHz (the lowest frequency used), the period is \( \tau \approx 666.7 \text{ ns} \). With this period, the result of eq. (5.14) would be \( \Delta t \approx 0.11 \text{ ns} \), which is at least five times less than the errors obtained experimentally. Consequently, this source of error was considered negligible.

5.5 - THE REMAINDER OF TABLE 5.1

From the experiments reported in the previous sections it was learned that when a replacement method is to be used, the velocity in the reference medium (in this instance, water) must be measured immediately before any measurement in the specimen is carried out, and at the same Tx-Rx separation as when the specimen is present.

From eq. (4.2), the relation between the velocity in the sample and the velocity in the water, i.e., \( c_s = \frac{L}{(t_w - t + L/c)} \), it can be said that if a wrong value of velocity in the water is used in replacement methods for the computation of the velocity in a specimen, the result will also be wrong and in the same direction. This can be easily demonstrated with the use of a specimen having exactly the same value of velocity as the water (numerical examples of the order of magnitude of the error in velocity in silicone rubber block due to error in velocity in water are given in section 6.2). Therefore, the water must also be characterized before any attempt is made to characterize a
specimen immersed in it under the same conditions. In addition to the characterization of the water itself, an evaluation of the diffraction effects must be carried out, identifying where the far field starts. One should then work in the far field. In order to ensure that the characteristics of the water, for instance its ultrasonic velocity, does not vary during the time of the measurements on the specimen, the ultrasonic velocity in the water should be regularly checked.

The largest velocity hop observed was of the order of magnitude of 3 m s\(^{-1}\) (see section 5.1). For those measurements, the transducers were separated by approximately 200 mm and 500 mm. The difference in the measured velocity in water due to diffraction at those two particular positions was approximately 0.5 m s\(^{-1}\) at 3 MHz (see section 5.3). Whether diffraction effects could have contributed to the "velocity hop", observed in the samples characterization, is considered further in chapter 6, where the results related to the remainder of the experiments outlined in table 5.1 are presented and discussed.

5.6 - ATTENUATION MEASUREMENTS

The transfer characteristics of the pair of 7.5 MHz transducers (Panametrics, V-320) was obtained experimentally and compared with the numerical model of Bacon & Chivers (1981) and of Papadakis (1975). Two basic experiments were performed: [i] the frequency response of the pair Tx-Rx at different separations; and [ii] the transfer of amplitude of signal as a function of the continuous variation of Tx-Rx separation at different frequencies. These experiments are discussed below.

The swept frequency technique (section 4.4.4) was used for the first of the above experiments, with the exception that no specimen was interposed between the transducers, but just distilled water.
Frequency spectra were obtained for Tx-Rx separation from 0 mm to 600 mm, at approximately 19°C. Only the frequency spectra at Tx-Rx separation of 0 mm, 200 mm and 500 mm were digitised onto a computer for presentation in figure 5.11. The transducers were taken out of the acoustic tank and measurements were taken for Tx-Rx = 0 mm, with the transducers coupled by a film of water and by a film of ultrasonic coupling gel. No difference in spectrum was noticed between any of these measurements at 0 mm. Another set of measurements for the same separation was taken by rotating one of the transducers (gel coupled) by 45° and 90° about the longitudinal axis. Very little difference was observed at high frequencies, i.e., approximately 0.5 dB between 0° and 90°. The frequency spectrum for the 45° position lay between the other two. The plateau in the frequency response for Tx-Rx = 0 mm is not fully understood. However, it suggests: [a] the minimum transmission coefficient of the protective/matching layers (of unknown material and thickness) at the face of each piezoelectric element of the transducers; [b] interference phenomena within the protective/matching layers; [c] the complex structure of the near field of the acoustic beam; [d] the loading of the receiver over the transmitter, thus changing its electrical impedance; [e] any combination of the above possibilities.

Frequency spectra for the same pair of transducers were also obtained with the time delay spectrometry technique (section 4.4.3) at the transducer separations of about 160 mm and 320 mm (Chivers et al. 1985). Although these are not reproduced here, they were compared with the results acquired with the above ordinary swept frequency technique, at Tx-Rx = 150 mm and Tx-Rx = 300 mm. Both techniques exhibited almost identical spectral shape, and the largest observed difference in amplitude was about 1 dB.
FIGURE 5.11: Frequency spectra of the pair of 7.5 MHz transducers in water. (Panametrics Acuscan Immersion Transducers, unfocussed, 7.5 MHz nominal frequency, 12.7 mm diameter elements, ref. No. V-320, serial numbers Tx: 39651 and Rx: 39652). The spacing between the transmitter and the receiver are indicated on the curves. The temperature of water was approximately 19°C.
FIGURE 5.12: Block diagram for measurement of diffraction loss of ultrasonic waves in water. Stepping motor speed 1 mm/s, plotter speed 20 mm/cm.
A slight modification of the system in figure 4.7 was implemented to obtain the transfer of signal amplitude as a direct function of the continuous variation of the transducer separation, at discrete frequencies. This technique is represented in figure 5.12.

The transmitting transducer (at a fixed position in the tank) was excited by a continuous sine wave signal, generated at the signal generator of the spectrum analyser (Marconi Instruments, TF-2370).

The receiving transducer was attached to the tracking mechanism of the acoustic tank. The tracking mechanism was driven by a stepping motor controlled by a micro-computer system. The speed of the stepping motor was set to 1 mm s\(^{-1}\), operated by a computer programme named "steploss". The time base of the X-Y chart recorder (Bryans Plotter, 29000-A4) was calibrated against the stepping motor speed, to register the position of the receiving transducer, Rx, in respect to the position of the transmitting transducer, Tx. The receiving transducer was moved only along the longitudinal axis. Both the stepping motor and the X-Y chart recorder were operated simultaneously. The vertical axis of the chart recorder was, in turn, calibrated against the spectrum analyser signal amplifier.

Three sets of measurements of variation of amplitude of received signal as a function of the transducer separation were taken. The investigation was made at different frequencies (indicated in figure 5.13). For each frequency investigated, the starting separation of the transducers was 0 mm, i.e., with the transducers physically in contact. In each of the three sets of measurements, a new alignment of the transducers was made, and the reproducibility of the results obtained was within ±0.2 dB. One typical set of experimental results (at the temperature of 17.6±0.2°C) was digitised into a computer for subsequent comparison with the numerical model prediction of an
idealised pair of piston-like transducers, the solid line in figure 5.13, which shows the diffraction loss effect alone.

The original measurements of signal amplitude were reprocessed with respect to a reference amplitude of signal, at an arbitrary position, so as to give 0 dB amplitude loss at zero transducer separation. By doing so, the unknown frequency dependent absorption and transmission coefficient of the protective/matching layers (of unknown thickness and material) in front of each piezoelectric element of the transducers were compensated for. To facilitate the comparison of the results at all frequencies, the transducer separations were normalised to \( \frac{a^2}{\lambda} \), where \( a \) is the nominal radius of the transducer and \( \lambda \) is the wavelength. The wavelength was calculated from the measured piecewise velocity (section 5.3) in intervals of 100 mm in the far field of the acoustic beam. For guidance in figure 5.13, the value of \( \frac{a^2}{\lambda} \) at 1 MHz is approximately 27.25 mm.

The experimental results obtained are presented in figure 5.13.a. For a better view of these results, they were reproduced in figure 5.13.b, but with the vertical scale shifted by -0.2 dB for each frequency. Notice that, as the frequency is increased, the measured amplitude loss is also increased. One of the plausible factors for this increase in loss of amplitude with the frequency was thought to be the frequency dependent absorption of ultrasound in water. The absorption of ultrasound in water given in the literature is \( \alpha = 2.4 \times 10^{-4} \text{ dB mm}^{-1} \text{ MHz}^{-2} \) at 17.6 °C (Nosdrev, 1965). The original experimental data (figure 5.13.a) was then reprocessed to account for the absorption of ultrasound in water. The results obtained with this process are presented in figures 5.13.c and 5.13.d. The experimental results in figure 5.13.d are the reproduction of the ones in figure 5.13.c, but shifted by -0.5 dB for each frequency to give a
better view of the near field.

As the frequency is increased, the peaks and troughs in figure 5.13.c do not coincide and become less pronounced. In particular, the position and magnitude of the minimum, indicated by the arrows, on the experimental curves were different at different frequencies. It can also be seen clearly that the data do not coincide in the far field of the acoustic beam. The above observations suggest that: [a] the absorption in the water is slightly greater than the value given in the literature; and [b] the normal derivative of the velocity potential across the transducers had changed with the frequency applied.

To verify the first of the above hypotheses, the original data was again reprocessed, but this time assuming a slightly greater absorption coefficient in water, i.e., \( \alpha = 2.8 \times 10^{-6} \text{ dB mm}^{-1} \text{ MHz}^{-2} \). These reprocessed data are presented in figures 5.13.e and 5.13.f. As before, the experimental results in figure 5.13.f are the reproduction of the ones in figure 5.13.e, but shifted by \(-0.5 \text{ dB}\) per set of data to give a better view of the results at each frequency. Notice that a better fit of the experimental results with the theoretical model of an idealised piston-like transducer was achieved at almost all frequencies. A distinct discrepancy is observed when comparing the experimental and theoretical results at frequencies below 4 MHz. The results above and including 4 MHz appear to be in very good agreement with the theory. From this experiment and approach of re-processing the experimental data, measurement of the absorption coefficient in the water used was made at the temperature at which the experiment was carried out. The order of magnitude of the value of the obtained absorption coefficient is in accordance with those found in the literature, and therefore realistic. Thus, the method outlined above may possibly be used for
FIGURE 5.13.i: Amplitude loss of signal as a function of normalised transmitter-receiver separation in water. The solid line represents the predicted loss due to diffraction for an ideal piston-like pair of transducers. (a) measured amplitude loss at the frequency indicated, (b) experimental results shifted by 0.2 dB per frequency for a better view of the near field.
FIGURE 5.13.ii: See figure 5.13.i. (c) experimental results reprocessed to account for absorption in water $\alpha = 2.4 \times 10^{-4}$ dB mm$^{-1}$ MHz$^{-2}$ (Nozdrev, 1965), (d) experimental results shifted by 0.5 dB per frequency for clarity.
FIGURE 5.13.iii: See figure 5.13.i. (e) experimental results reprocessed to account for absorption in water, $\alpha = 2.8 \times 10^{-4}$ dB mm$^{-1}$ MHz$^{-2}$ (empirical value), (f) results shifted by 0.5 dB per frequency for clarity.
measuring absorption coefficient in continuous media. A possible explanation for the discrepancy between the experiment and the theory below 4 MHz is thought to be related to the second of the above hypotheses, of which discussion follows.

Roderick & Truell (1952), Papadakis (1963), and Martin & Breazeale (1971) have shown some evidences of changes in diffraction when the transducer radiates as other than a piston source. Papadakis (1975) investigated various plausible functions to simulate transducers. All his functions had the normal derivative of the velocity potential taken to be circularly symmetrical and with the maximum at the centre of the source and the minimum at the rim. Nevertheless, he pointed out that the actual response of a transducer may not correspond to any of the functions he had chosen.

The functions for velocity potential derivative that Papadakis had considered are tabulated in table 5.3. The profiles of the normal derivatives of the velocity potential across the transmitting transducers are illustrated in figure 5.14.a. The gradient in the velocity potential are to be expected, since in practice the transducers are plated only over a fraction of their area. The value of the radius of the plated area, \( a_p \), was taken to be 0.8a, where a is the radius of the transducer disk. The reception was limited to the plated area of the transducer and the weighting function over this area was made unity. The radius of the plated area, in both transmission and reception, was assumed to be the effective area of the transducers. The effects that these functions have on the amplitude loss and the phase-shift are shown in figures 5.14.b and 5.14.c, respectively. The abscissa in each of these figures is the distance normalised to \( a_p/\lambda \). The calculated curves for the all-plated piston source are also included for comparison. The principal effect of reducing the
amplitude of motion of the outer part of the radiating transducer is to lower the diffraction loss and its fluctuation with distance. This seems to confirm the hypothesis [b] stated above. In addition, the position of the loss peaks and troughs along the normalised distance depends upon the amount of radiation generated outside of the plated area. This is to be expected because the receiver of radius $a_p$ is sensing the field of a larger transmitter with an effective radius $a_t$ in the range $a_p < a_t < a$. The phase is affected in such a way as to smooth the curves. The plateaus found for the piston case are changed into regions of low slope.

**TABLE 5.3**

Functions for velocity potential derivative

<table>
<thead>
<tr>
<th>type</th>
<th>function</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>Sinusoid  $\cos(\pi f/4a)$</td>
</tr>
<tr>
<td>B</td>
<td>Sinusoid  $\cos(\pi f/2a)$</td>
</tr>
<tr>
<td>C</td>
<td>Gaussian $\exp(-f^2/1.28a^2)$</td>
</tr>
<tr>
<td>D</td>
<td>Gaussian $\exp(-f^2/2.56a^2)$</td>
</tr>
<tr>
<td>E</td>
<td>Fermi  $1 [1 + \exp(5f/a - 4)]$</td>
</tr>
<tr>
<td>F</td>
<td>Fermi  $1 [1 + \exp(25f/a - 20)]$</td>
</tr>
</tbody>
</table>

* after Papadakis (1975).

When comparing the results of figure 5.13.e and 5.13.f with the studies of Papadakis (1975), it appears that the transmitting transducer radiated differently at different frequencies. For frequencies close to its resonant frequency (i.e., 4-8 MHz) it seems that the transducer had radiated somewhat like a piston-source or like the function of type F (in table 5.3 and figure 5.14). At the lower frequencies (i.e., 1-3 MHz) it seems that the transducer had operated somewhat like the functions A, C, or D.

In conclusion, for precise diffraction corrections for attenuation measurements in a continuum medium, it is necessary to investigate
FIGURE 5.14: Effects of velocity potential derivative on diffraction loss and phase. (a) profiles of the normal derivative of the velocity potential across the transmitting transducer. The functions are given in table 5.3. (b) and (c) are the relative diffraction loss and phase for the input functions of (a), respectively. The abscissa in (b) and (c) is the normalised distance to \( a_p^2/\lambda \), where \( a_p \) is the radius of the plated area of the transducers, and \( \lambda \) is the wavelength. The curves for piston source are included for comparison. (after Papadakis, 1975).
further the hypothesis [b] above. Due to lack of time, it was not possible to pursue this investigation and, therefore, it is left as a suggestion for a future research project.

5.7 - CONCLUSION

In this chapter, it was demonstrated that diffraction effects can be measured for necessary corrections in both phase velocity and attenuation measurements, although it was found to be a difficult task.

The piecewise and the non-piecewise velocity measurements have shown that diffraction effects can be detected. Nevertheless, for the particular case of the non-piecewise method, such effects may be masked if the data is processed with the origin in the far field and directed towards the near field. Measured diffraction effects may not always appear to be of the same order of magnitude as an ideal piston-like model. However, the study reported here showed good agreement with simple ideal models used within the limitations of the measurement devices and the experimental procedures adopted.

From the analysis of error in velocity measurements, it was concluded that errors in the time measurement technique, age of water, turbulence in water, as well as temperature fluctuations and re-alignment of the transducers contributed very little to the errors in velocity. The major contribution to the inaccuracy of measurement of the ultrasonic velocity relied on the mechanics of the tracking mechanism system, i.e., on the estimation of the distance between the transducers for the non-piecewise method and on the distance travelled by either the receiver or the transmitter, for the piecewise method.

Evidence was shown that the velocity potential derivative describing the mode of vibration of the transmitting transducer was frequency-dependent and varied between a plate-like and a piston-like
source. To evaluate the precise magnitude of diffraction corrections for attenuation measurements, a non-absorbing medium (or a medium with precisely known absorption coefficient) must be used. From the experiment related to diffraction corrections for attenuation measurements, the absorption of ultrasound in water was measured, giving the same order of magnitude of the values found in the literature.

The effects of diffraction on phase velocity and attenuation measurements are further discussed in chapters 6, 7 and 8 where the phenomenon is related to the measurements on the specimens described in chapter 3.
CHAPTER 6

VELOCITY RESULTS
CHAPTER 6

VELOCITY RESULTS

6.1 - INTRODUCTION

In chapter 5, table 5.1 outlined the approach to understand a velocity hop observed in the measurements of velocity in some of the samples. The first of a series of four experiments is discussed in detail in that chapter. In the present chapter, the remainder of the experiments of table 5.1 is discussed and followed by a new set of measurements giving the final results of velocity for the whole set of samples described in chapter 3. For convenience, the discussion of the final results of velocity is presented in chapter 8.

6.2 - PRELIMINARY RESULTS

In the early stage of this research work, a velocity hop at 3 MHz had been observed when the transducer separation was changed from 200 mm to 500 mm. The frequency range covered in the former separation was 1.5-3 MHz, and in the latter it was 3-8 MHz. The magnitude of the velocity hop varied from sample to sample. A sequential study was designed to investigate the origins of such hops in measured velocity. This sequential study is outlined in table 5.1. The first of the experiments was related with diffraction effects, and it is presented and discussed in chapter 5. The remainder of the study related with table 5.1 are presented in this section. Samples 1, 20, 2 and 27 were chosen for experiments 2 and 3. Originally, samples 1 and 20 did not
show a clear velocity hop. Sample 2 had shown a negative velocity hop of about 2 m s\(^{-1}\) and sample 27 had shown a positive velocity hop of about 1 m s\(^{-1}\). When the velocity hop was originally observed, the technique of measurement used was the digitised tone-burst described in section 4.4.1.2.

To investigate the occurrence of the velocity hop, the sample was positioned half-way between the transducers (as in the earlier experiments) and measurements of velocity were then obtained with the time interval averaging technique (see section 4.4.2). A threshold level was set on the counter timer to ignore the leading peak (kept below 80 mV throughout the entire frequency range used) of the received tone burst. The threshold level was then detected at the second cycle of the received tone burst (stop-channel of the counter timer) which had its peak amplitude kept constant at approximately 160 mV by means of the variable attenuators. Measurements were taken over the frequency range of 1.5 MHz to 8 MHz for both transducer separations of 200 mm and 500 mm. The results are presented in figure 6.1.

In the measurements reported in figure 5.1 (chapter 5), the velocity in water had been extracted from the data table of Del Grosso & Mader (1972). In the experiments described in this chapter, the velocity in water was measured according to the piecewise method, described in chapter 5, at the frequency of 8 MHz. The starting transducer separation was 100 mm and measurements were taken in four intervals of 100 mm, i.e., 100-200 mm, 200-300 mm, 300-400 mm, and 400-500 mm. The average value obtained and its variation was \(c_w = 1480.76\pm0.55\) m s\(^{-1}\), which is about 1.6 m s\(^{-1}\) less than the value taken from Del Grosso and Mader. As discussed in chapter 5, if a wrong velocity in water is used when the substitution method is employed (section 4.2, eq.(4.2)) a wrong velocity in the specimen will result.
FIGURE 6.1.1: Preliminary results of velocity in sample as a function of frequency at transducer separation $Tx-Rx = 200$ mm (×) and $Tx-Rx = 500$ mm (○). The sample was positioned halfway between the transducers. (a) sample 1, and (b) sample 20.
FIGURE 6.1.ii: See figure 6.1.i. (c) sample 2, and (d) sample 27.
It is necessary, therefore, to measure the velocity in water under the same conditions (temperature and pressure) and transducer separation (because of diffraction effects) that the measurements in the specimen will be carried out. For details on these conditions the reader is referred to chapter 5.

In the new set of measurements, there are velocity hops, but they are not particularly confined to 3 MHz and to a particular sample. They occur in various frequencies and are, in most cases, just about covered by the error bars. Therefore, it is rather doubtful whether they are physically significant. Only two cases were not covered by the calculated error bars: samples 1 and 2 at 1.5 MHz and 2 MHz.

Experiment 4 of table 5.1 was then performed. The technique of measurement used was the same as for experiments 2 and 3 described above. Only the lower and upper limit of the frequency range were investigated, i.e., at 1.5 MHz and at 8 MHz, for both Tx-Rx = 200 mm and Tx-Rx = 500 mm. Sample 20 was used in this experiment and its relative position between the transducers was varied. The data presented in figure 6.2 show the measured velocity in the sample as a function of the distance between the transmitter and the first interface water/sample. Figure 6.2.a and 6.2.b show the data at 1.5 MHz and 8 MHz, respectively. The data for both Tx-Rx separation of 200 mm and 500 mm are presented together for ease of comparison.

It can be seen in figure 6.2, that the measured velocity in the sample did not change with its relative position in the acoustic field. However, a higher velocity was observed at Tx-Rx = 500 mm than at 200 mm for both 1.5 MHz and 8 MHz. It was thought that two factors contributed to this difference in velocity, namely diffraction effects and temperature variation in water, since it is believed that no other parameter had been changed. From eq. (4.2), if the velocity in water
FIGURE 6.2: Ultrasonic velocity in sample 20 as a function of its relative position between the transducers at the frequencies of (a) 1.5 MHz and (b) 8 MHz. Transducer separation Tx-Rx = 200 mm (x) and Tx-Rx = 500 mm (□).
is under estimated, the resulting velocity in the sample will also be
under estimated.

From the studies presented in chapter 5, the velocity in water with
the transducer separation of 200 mm was about 1.2 m s\(^{-1}\) greater than the
velocity at 500 mm separation at the frequency of 1.5 MHz, and about
0.5 m s\(^{-1}\) at 8 MHz. For a sample of thickness 29.21 mm, the increase in
velocity that would then be observed is about 0.57 m s\(^{-1}\) at 1.5 MHz and
about 0.24 m s\(^{-1}\) at 8 MHz, which are in the right sense but about half
the differences which were experimentally observed. Diffraction would
account for positive velocity hops only. The results of experiments 2,
3 and 4 had agreed with this direction of velocity hop, with the
exception of sample 27 at the lower end of the frequency range.

The magnitude of this positive velocity hop in sample 20 varied
from experiments 2 and 3 (figure 6.1.b) to the experiment 4 (figure
6.2). This is particularly noticeable on the measurements at 1.5 MHz.
Temperature variations between one set of measurements and another
could have been responsible for that extra variation. The diffraction
effects are expected to be the same on any occasion, whereas
temperature variation could have been larger on one occasion than on
another. Even if the temperature had varied by 1°C, the diffraction
alone would not account for that difference in velocity in the sample.

The velocity in water was measured only once (at 8 MHz) and
thereafter the temperature was monitored to 0.1°C accuracy. Although
the water had been stirred, there could have been some gradient of
temperature within the water tank during the measurements.

No other effects were thought to provide with an explanation for
the discrepancies of that sort of magnitude in velocity in the sample.
6.3 - FINAL RESULTS

In the light of the preliminary results discussed in chapter 5 and in the previous section of the present chapter, it was decided to place the receiver at 500 mm from the transmitting transducer, where the presence of approximately plane waves can be assumed within the whole range of frequency of interest. The theory of multiple scattering of waves used to predict the ultrasonic propagation in the specimens assumes the incidence of plane waves. Therefore, to have approximately plane waves entering the samples, this was placed at the farthest possible position from the transmitter with the existing experimental arrangement i.e., about 300 mm.

The experimental results, presented in figures 6.3 to 6.7, were obtained with the time interval averaging technique described in section 4.4.2. The theoretical model prediction (chapter 8) is plotted together with the experimental data in figures 6.3 to 6.7. The theoretical prediction of the phase velocity and group velocity are represented by the solid and the dashed line, respectively, according to the measured volume concentration of glass beads (or scatterers - see table 3.2.b). This is summarised in table 6.1 for a quick reference guide.

Some general comments on these experimental results are given in this chapter. A further discussion is given in chapter 8, to facilitate comparisons with the theoretical results and the attenuation.
TABLE 6.1

Guide to figures 6.3 to 6.7.

ultrasonic velocity in sample

<table>
<thead>
<tr>
<th>symbol</th>
<th>measured</th>
<th>phase</th>
<th>group</th>
</tr>
</thead>
<tbody>
<tr>
<td>x</td>
<td>✓</td>
<td></td>
<td></td>
</tr>
<tr>
<td>solid line</td>
<td>✓</td>
<td></td>
<td></td>
</tr>
<tr>
<td>dashed line</td>
<td>✓</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

KEY WORDS:
- measured: experimental data.
- phase and group: theoretical prediction.
FIGURE 6.3: GROUP I. Predicted phase velocity (solid line) and group velocity (dashed line) compared with measured velocity (x) in samples.
FIGURE 6.4.a: GROUP II. Predicted phase velocity and group velocity compared with measured velocity in samples. (see table 6.1).
FIGURE 6.4.b: GROUP II. (See figure 6.4.a and table 6.1).
FIGURE 6.5.a: GROUP III. Predicted phase velocity and group velocity compared with measured velocity in samples. (see table 6.1).
FIGURE 6.5.b: GROUP III. (See figure 6.5.a and table 6.1).
FIGURE 6.6. Predicted phase velocity and group velocity compared with measured velocity in samples.
(see Table 6.1).
FIGURE 6.6.c: GROUP IV. (See figure 6.6.a and table 6.1).
FIGURE 6.6.d: GROUP IV. (see figure 6.6.a and table 6.1).
FIGURE 6.7.a: GROUP V. Predicted phase velocity and group velocity compared with measured velocity in samples (see Table 6.1).
FIGURE 6.7.c: GROUP V. (see figure 6.7.a and table 6.1).
FIGURE 6.7.d: GROUP V. (see figure 6.7.a and table 6.1).
FIGURE 6.7.e: GROUP V. (see figure 6.7.a and table 6.1).
6.4 - ERROR ANALYSIS

The error bars indicated on the measured velocity in the samples (figure 6.3 to 6.7) were determined from the formulation of the most probable error (Topping, 1979). This approach of determining the magnitude of errors of measurements in velocity involved each separate measured physical quantity, i.e., errors related with: the dimension of the sample; the time of flight with the sample in position in the acoustic beam; the time of flight in water alone; and the velocity of ultrasound in water.

Taking equation (4.2) for the velocity of ultrasound in the sample immersed in water, "the most probable error" in \( c_s \) is given by:

\[
\varepsilon_s^2 = \left( \frac{\partial c}{\partial x} \right)^2 \varepsilon_x^2 + \left( \frac{\partial c}{\partial t_{ws}} \right)^2 \varepsilon_{t_{ws}}^2 + \left( \frac{\partial c}{\partial t} \right)^2 \varepsilon_t^2 + \left( \frac{\partial c}{\partial c} \right)^2 \varepsilon_c^2 \]  

(6.1)

where \( \varepsilon_x, \varepsilon_{t_{ws}}, \varepsilon_t, \varepsilon_c \) are respectively the errors in the dimension of the sample (see section 3.2); in the time of flight with the sample intervening the acoustic beam; in the time of flight in water alone; and in velocity in water.

By solving the derivatives of equation (4.3) one can write

\[
\varepsilon_{c_s} = \left[ \frac{1}{(t_{ws} - t)^2 \varepsilon_x^2 + \varepsilon_{t_{ws}}^2 (\varepsilon_x^2 + \varepsilon_t^2) + (c/c)^4 \varepsilon_c^2} \right]^{1/2} \]  

(6.2)
6.5 - GENERAL COMMENTS

It can be seen very clearly in figures 6.3 to 6.7, that a fall in the measured velocity is present at 2.5-3 MHz, the magnitude of which varied from sample to sample, but all of them in the same direction. An explanation to that fall in the measured velocity is given below.

With the received signal displayed on a delayed time base of an oscilloscope, it was observed that its first cycle had a constant period throughout the frequency range applied from the wavefunction generator. It was also observed that the frequency of this first cycle of the received signal was approximately 7 MHz, which is approximately the central frequency of the transducers. An observation was also made on the transmitting signal and no clear distinction (in the period of oscillation) was found, except that the leading peak had a higher amplitude than the rest of the exciting tone burst.

To identify the origin of this high frequency component of the start of the received pulse, the output of the wavefunction generator was switched to zero volts, but the gate was still operated. By doing so, both transmitted and received signals exhibited a pulse of two cycles of approximately 7 MHz at the same time at which the gate was activated, i.e., at the opening and closing of the gate. When the pulse generator (used to gate the wavefunction generator) changed its state, it was detected by the transmitting transducer (by means of electrical leakage) emitting two transient pulses of wide frequency spectra, one at the tip and the other at the tailing of the gate duration. Subsequently, these wide band pulses were transmitted through the water and detected at the receiving transducer. The peak amplitude of the received signal was approximately 34 mV. With the insertion of a sample in the acoustic path, those wide band pulses were
attenuated as a result of the acoustic propagation properties of the sample.

When the wavefunction generator was switched on, constructive and destructive interference occurred between its sinusoidal tone-burst signal and those wide band pulses, leading to a change in phase of the second cycle of the train of sine wave signal. The counter timer had been arranged to ignore the first cycle of the received tone burst and to stop at the threshold level of 40 mV on the second cycle (see section 4.4.2). It was realised that sudden changes of velocity with frequency in the region of 3 MHz could be due to that effect on the phase of the signal. The size of that effect on velocity in the sample was estimated (from the period of the transient pulses) to be of the order of magnitude of 1 m s⁻¹. A manual measurement, i.e., ignoring the counter timer and reading the time difference (time of flight with and without a sample) directly from the oscilloscope screen confirmed the order of magnitude of that effect on velocity in sample 1. That problem could perhaps have been avoided if a zero-crossing detector set to a pre-determined value from the leading cycle position of the tone burst was available. Nevertheless, having in mind the problem, its origin and effects on the results, the entire investigation of velocity in the whole set of samples as a function of frequency proceeded.

The velocity in water, required by eq.(4.2), was measured according to the piecewise method in intervals of 100 mm each (see section 5.3.1). The error in velocity in water, i.e., \( \xi_c \) was obtained from equation (5.12). It was observed that the major contribution to errors of measurements of velocity in the sample was from temperature fluctuations (random error) in the water bath.

An effort was made to keep the temperature in the water bath within the accuracy of the ultrasonic thermometer used, i.e., 20.0±0.1°C (see
section 4.3.6). However, on some occasions the room temperature was higher (about 25°C) or lower (about 11°C) than the desirable value, which consequently caused instability in the water bath temperature. This lead to relatively large fluctuations in measurements of the time of flight ($3 \text{ ns} \leq t \leq 25 \text{ ns}$).

A better thermal insulation of the acoustic tank and/or an air conditioning system in the laboratory would be adequate to increase the precision of measurements.

The measurements on each sample, over the entire range of frequency of interest take about one hour to be completed. This means that fine temperature control in the acoustic tank was made a difficult task, even when covering the whole tank with expanded polystyrene shields. The sensitivity of the results to temperature instability is relatively large (see section 4.3.5). Unfortunately, the water circulator and stirrer had to be switched on and off very regularly, leading to relatively large random errors in the results.

The results are further discussed in chapter 8.
CHAPTER 7

ATTENUATION RESULTS
CHAPTER 7

ATTENUATION RESULTS

7.1 - INTRODUCTION

Attenuation of the ultrasonic waves in materials was found to be easier to measure than velocity. Some preliminary measurements were made regarding the effects of diffraction on attenuation measurements on the specimens. The final results and analysis of attenuation of ultrasound in the whole batch of samples have shown that the attenuation of ultrasound is a reliable index for characterization of a two phase medium, although the velocity is also needed for a full ultrasonic characterization. The results are presented in this chapter, accompanied by some general comments. Their discussion will be given in chapter 8, where comparisons with the theoretical predictions and with the velocity results are made.

7.2 - PRELIMINARY RESULTS

With the use of the numerical model approach of Bacon & Chivers (1981) discussed in chapter 5, the effects of diffraction in silicone rubber and in water were separately computed for comparisons. Figure 7.1 illustrates the computed magnitude of the effects of diffraction (phase shift and loss) in water and in silicone rubber RTV-602, at the frequency of 1 MHz, for a pair of identical transducers (one as transmitter and one as receiver) of radius 6.35 mm. Because of the lower velocity in the silicone rubber, compared with water, there
FIGURE 7.1: Theoretical phase angle and relative loss of signal as a function of transducer separation due to diffraction in distilled water and in silicone rubber RTV-602. Radius of transducers $a = 6.35$ mm, frequency $f = 1$ MHz. $d/\lambda \approx 8.6$ in water and $d/\lambda \approx 12.5$ in RTV-602.
results a phase lag in the wave propagation. In measuring attenuation in a specimen, the results that are obtained are not only due to the inherent properties of the specimen but also due to diffraction effects. An experiment was performed to verify the effects of diffraction phenomena on the attenuation measurements in a pure silicone rubber specimen. Samples 1 and 20 were used for this experiment, which employed the swept frequency technique (described in section 4.4.4) and the substitution method outlined in section 4.2. The geometrical dimension of the sample blocks facilitated this study. Attenuation measurements were obtained along the width of sample 1 (29.18 mm) and the length of sample 20 (48.54 mm), in turn, with both transducers in contact with the sample. The results are presented in figure 7.2.

It can be clearly seen that sample 20 has a lower absorption than sample 1, over the entire frequency range investigated. The disparity between the measured attenuation in samples 1 and 20 at frequencies above 8 MHz is due to the low signal to noise ratio for sample 20, where an extra rubber length was present, leading to a higher loss of signal. The trough observed at approximately 8.5 MHz is believed to be an electronic artefact inside the spectrum analyser signal processor as a result of the poor signal to noise ratio. The effect of the trough and the low signal to noise ratio will be also seen on the final results for the whole set of samples. On each graph (see figures 7.2 to 7.8) an arrow has been used to indicate the frequency above which the signal to noise ratio may be beginning to be sufficiently low to prejudice the value of the measurements. The experimental data above the frequency indicated with the arrow should therefore be disregarded.

Note the peaks and bulges on the frequency range below 6 MHz, particularly more pronounced for the closer transducer separation. At
FIGURE 7.2: Diffraction effects on measurement of attenuation along width of sample 1 (Δ) and length of sample 20 (×). Desregard the results at frequencies above the position of the arrow (sample 20) - see text. The error bar represents the order of magnitude of reproducibility of measurements.
these transducer separations and at the frequencies where the occurrence of peaks and bulges was observed, troughs in diffraction loss occur in water (see figure 7.1 and section 5.6). Therefore, the lag in phase in the silicone rubber, when compared with water, can give rise to misinterpretation of attenuation measurements. Corrections are consequently necessary. From the data of figure 7.1, it was estimated that in the case of a sample of 30 mm thick, the magnitude of the diffraction correction was about -0.2 dB (at 1 MHz) for the transducers separated by a distance equal to the thickness of the sample, i.e., Tx-Rx = 30 mm. The corrections are easier to make if the sample (of finite dimension) is inserted in the far field of the transmitter at whichever frequency applied, where no peaks or troughs take place, since the magnitude of the peaks and troughs due to diffraction are dependent on how the transducer responds to the applied signal (see section 5.6).

The investigation on the effect of diffraction upon attenuation measurements was continued at transducer separations of approximately 200 mm and 500 mm. As in the velocity study at these transducer separations (chapter 6), the relative position of the sample between the transducers was varied. Samples 1 and 20 were used for this purpose, with the waves travelling along their width. The results are presented in figure 7.3. Table 7.1 indicates the configuration of transducer separation and the approximate position of the sample, relative to the transducers, as a guide to figure 7.3.
TABLE 7.1
Configurational guide to figure 7.3.

<table>
<thead>
<tr>
<th>symbol</th>
<th>Tx-Rx (mm)</th>
<th>Tx-Sz₁ (mm)</th>
<th>Rx-Sz₂ (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>○</td>
<td>200</td>
<td>0; 20; 100</td>
<td>270; 150; 70</td>
</tr>
<tr>
<td>+</td>
<td>500</td>
<td>0; 20; 100; 470; 450; 370; 200; 300; 400 270; 170; 70</td>
<td></td>
</tr>
<tr>
<td>□</td>
<td>200</td>
<td>170</td>
<td>0</td>
</tr>
<tr>
<td>◊</td>
<td>500</td>
<td>470</td>
<td>0</td>
</tr>
</tbody>
</table>

symbol: symbol used in figure 7.3.
Tx-Rx: approximate transducer separation.
Tx-Sz₁: approximate distance between the transmitting transducer and the first surface of the sample.
Rx-Sz₂: approximate distance between the receiving transducer and the second surface of the sample.

For purposes of clarity, the results for the sample either in physical contact with the transmitter or with the receiver at both Tx-Rx separation of 200 mm and 500 mm were shifted by 3 dB cm⁻¹ in figure 7.3. At the lower frequency range (below 6 MHz) a higher attenuation was observed when the sample was in contact with either transducer than when completely surrounded by water. This may be explained by the change in transmission and reflection coefficient originating from a change in the characteristic impedance of one of the media, i.e., the incident or the emerging medium. The characteristic acoustic impedances of the transducers are not known to the author. Note also that a bulge in the attenuation occurs below 6 MHz and transducer separation of 200 mm. It appears that diffraction effects are responsible for this excess attenuation (compared with Tx-Rx = 500mm) as in the case of figure 7.2. As the frequency was increased, the attenuation results in the samples, for each transducer separation, gradually became indistinguishable, as far as the relative
FIGURE 7.3: Diffraction effects on measurements of attenuation in samples 1 and 20. The results represented by the symbols □ and ◊ were shifted by 3 dB cm⁻¹ for clarity (see text and table 7.1). The error bar indicates the order of magnitude of the reproducibility of measurements.
position of the sample and transducer separations are concerned. However, as previously pointed out, for frequencies above 8 MHz, there is a distinct difference between the results for the two transducer separations. At the Tx-Rx = 500 mm configuration, the signal to noise ratio is reduced. With the insertion of the sample in the acoustic path, the signal to noise ratio is reduced even further, causing a poor amplitude resolution in the signal processing circuitry of the spectrum analyser. This was due to the diffraction losses in addition to the intrinsic absorption of both media.

From the above experiments and results, measurements of attenuation, when using comparative methods, should be taken in the far field of the acoustic beam, where diffraction corrections are easier to make and of minor magnitude.

7.3 - FINAL RESULTS

The attenuation results for all samples, obtained with the various techniques of measurements used (chapter 4) are covered within this section. The symbols used in figures 7.4 to 7.8 are related with each individual technique, as specified in table 7.2. The theoretical data are plotted together with the experimental data. The solid line represents the theoretical prediction computed according to the measured concentration of glass beads in suspension in the matrix. For a more detailed discussion on the composition of the samples and on the techniques used, the reader is referred to chapters 3 and 4, respectively. The theory of multiple scattering of waves used is discussed in chapter 2, and its numerical solution is discussed in chapter 8. For convenience, the results are discussed in chapter 8, but some general comment with respect to the measurements are made in the following sections.
**TABLE 7.2**
Guide to figures 7.4 to 7.8

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>*</td>
<td>digitised signal technique (wide band excitation)</td>
</tr>
<tr>
<td>□</td>
<td>digitised signal technique (tone burst excitation)</td>
</tr>
<tr>
<td>△</td>
<td>time delay spectrometry technique (hydrophone as receiver)</td>
</tr>
<tr>
<td>▽</td>
<td>time delay spectrometry technique (pair of transducers of identical characteristics)</td>
</tr>
<tr>
<td>○</td>
<td>swept frequency technique (Tx-Rx = 200mm)</td>
</tr>
<tr>
<td>+</td>
<td>swept frequency technique (Tx-Rx = 500mm)</td>
</tr>
<tr>
<td>solid line</td>
<td>theoretical prediction</td>
</tr>
<tr>
<td>↓</td>
<td>indication of frequency above which the signal to noise ratio is sufficiently low to prejudice the measurements (data should be disregarded)</td>
</tr>
<tr>
<td>I</td>
<td>estimated error bar of measurements at 2 MHz (shown in figure 7.4)</td>
</tr>
</tbody>
</table>
FIGURE 7.4: GROUP I. Theoretical and measured attenuation as a function of frequency (see table 7.2).
FIGURE 7.5.a: GROUP II. Theoretical and measured attenuation as a function of frequency (see table 7.2).
Figure 7.5.b: Group II. (see figure 7.5.a and table 7.2).
FIGURE 7.6.a: GROUP III. Theoretical and measured attenuation as a function of frequency (see table 7.2).
FIGURE 7.6.b: GROUP III. (see figure 7.6.a and table 7.2).
FIGURE 7.7.a: GROUP IV. Theoretical and measured attenuation as a function of frequency (see table 7.2).
FIGURE 7.7.b: GROUP IV. (see figure 7.7.a and table 7.2).
FIGURE 7.7.c: GROUP IV. (see figure 7.7.a and table 7.2).
FIGURE 7.7.e: GROUP IV. (see figure 7.7.a and table 7.2).
FIGURE 7.7.f: GROUP IV. (see figure 7.7.a and table 7.2).
FIGURE 7.8.a: GROUP V. Theoretical and measured attenuation as a function of frequency (see table 7.2).
FIGURE 7.8.b: GROUP V. (see figure 7.8.a and table 7.2).
FIGURE 7.8.c: GROUP V. (see figure 7.8.a and table 7.2).
FIGURE 7.8.d: GROUP V. (see figure 7.8.a and table 7.2).
FIGURE 7.8.e: GROUP V. (see figure 7.8.a and table 7.2).
FIGURE 7.8.f: GROUP V. (see figure 7.8.a and table 7.2).
7.4 - ERROR ANALYSIS

7.4.1 - TRANSMISSION COEFFICIENT

When a progressive plane wave in fluid medium impinges on the boundary of a contiguous second medium a reflected wave is generated in the first medium and a transmitted wave in the second medium. The ratio of the respective intensities and pressure amplitudes of the reflected and transmitted waves to those of the incident wave depend on the characteristic impedances of the two media and on the angle of incidence of the incident wave. Kinsler and Frey (1962) discuss various situations related to the transmission phenomena. Here, only two relevant cases will be considered, for a parallel sided specimen of finite thickness, immersed in a liquid (water). The two cases are discussed below.

7.4.1.1 - PULSED WAVES

Consider the transmission of a pulsed plane wave from water through a parallel sided specimen S at normal incidence and into water again, as illustrated by figure 7.9. Consider the duration of the pulse to be sufficiently short to ensure any multiple reflections do not interfere among themselves in any of the media. Assume that the initial wave is travelling in the positive direction, with transmitted amplitude \( A_0 \) at the zero position. The characteristic acoustic impedances of water and of the specimen are \( Z_j = \rho_j c_j \) and \( Z_2 = \rho_2 c_2 \) respectively, where \( \rho_i \) is the density of the medium \( i \) and \( c_2 \) is its phase velocity.
FIGURE 7.9: Transmission loss of a parallel sided specimen for short pulsed waves at normal incidence (not to scale).
Considering the absorption (or the attenuation) coefficients $\mu_1$ in water and $\mu_2$ in the specimen, the transmitted acoustic amplitude $T_1$ and $T_2$ can be written (extending Kinsler & Frey, 1962) as:

$$T_1 = A_0 \cdot \exp(-\mu_1 \cdot x_1) \cdot \frac{2 \cdot Z_2}{Z_1 + Z_2}$$  \hspace{1cm} (7.1)$$

and

$$T_2 = T_1 \cdot \exp(-\mu_2 \cdot l) \cdot \frac{2 \cdot Z_3}{Z_3 + Z_2}$$  \hspace{1cm} (7.2)$$

where $x_1$ is the position of the first interface water/specimen with respect to the transmitting transducer $T_x$, $l = x_2$ is the thickness of the specimen, and $Z_3 = Z_1$.

The incident amplitude on the surface of the receiver $Rx$, located at $x_3$ from the specimen/water interface, is

$$A_r = T_2 \cdot \exp(-\mu_3 \cdot x_3)$$  \hspace{1cm} (7.3)$$

Substituting eq.(7.1) and (7.2) in eq.(7.3), and making $\mu_3 = \mu_1$, one can write

$$A_r = A_0 \cdot \exp[-\mu_1 \cdot (x_1 + x_3)] \cdot \frac{4 \cdot Z_1 \cdot Z_2}{(Z_1 + Z_2)} \cdot \exp(-\mu_2 \cdot l)$$  \hspace{1cm} (7.4)$$

Rearranging eq.(7.4), the absorption (or the attenuation) in the specimen can be written in the form of

$$\mu_2 = -\frac{1}{l} \cdot \ln \left[ \frac{A_r}{A_0 \cdot \exp[-\mu_1 \cdot (x_1 + x_3)]} \right] - \frac{1}{l} \cdot \ln \left[ \frac{(Z_1 + Z_2)^2}{4 \cdot Z_1 \cdot Z_2} \right]$$  \hspace{1cm} (7.5)$$

which gives the results in units of Np/length.

The second term of eq.(7.5) is called the transmission loss, T.L.,

$$T.L. = -\frac{1}{l} \cdot \ln \left[ \frac{(Z_1 + Z_2)^2}{4 \cdot Z_1 \cdot Z_2} \right] \quad [\text{Np/length}] \quad (7.6.a)$$

or

$$T.L. = -8.686 \cdot \frac{1}{l} \cdot \ln \left[ \frac{(Z_1 + Z_2)^2}{4 \cdot Z_1 \cdot Z_2} \right] \quad [\text{dB/length}] \quad (7.6.b)$$

The term $4 \cdot Z_1 \cdot Z_2 / (Z_1 + Z_2)^2$ is known as the ultrasonic transmission coefficient, $\alpha_r$, which indicates the factor by which the amplitude of
a plane wave passing at normal incidence is reduced due to reflections at the two interfaces.

If the transmission loss and absorption in water are negligible, equation (7.5) becomes identical to equation (4.4).

The transmission loss given by equation (7.6.b) was calculated for samples 1, 20, 4, 8, 26, and 34, for comparison with effects introduced by the scattering characteristics present in each of these samples. As velocity dispersion occurs in the samples (see chapters 6 and 8), their transmission coefficient will be frequency dependent. The velocity as a function of frequency in each of these samples, were obtained from the numerical solution of the theoretical model discussed in chapter 8, and used for the computation of the transmission loss given by equation (7.6.b). The results are presented in figure 7.10.

The transmission loss due to the different characteristic acoustic impedances of the two media (water and sample) is very small compared with the attenuation in the sample. For example, in the case of the clear rubber (samples 1 and 20) the transmission loss at 1 MHz was about one sixth of the measured loss of signal (absorption and reflection losses together). As the frequency was increased, the transmission loss became relatively smaller.
FIGURE 7.10: Transmission loss at interfaces for sufficiently short tone burst as a function of frequency at normal incidence for samples 1, 20, 4, 8, 26 and 34.
7.4.1.2 - CONTINUOUS WAVES

Reflection and transmission characteristics for the case of continuous waves through a specimen are relatively more complicated functions than for short pulses.

The model used for the computation of the transmission characteristics as a function of angle of incidence was based on the formulation given by Barnard et al. (1975). This model had been previously used by Dr. L.W. Anson, who kindly let his computer programme be used for the present analysis.

The case of a single solid specimen surrounded by liquid media is shown in figure 7.11. The incident pressure amplitude is $P_i$, the reflected amplitude is $P_r$, and the amplitude of the wave after transmission through the specimen is $P_t$. The complex transmission coefficient for a general case is given by

$$T = \frac{2 N Z_3}{M(Z_3 + Z_1) + i[(N^2 - M^2)Z_3 + Z_1]} \tag{7.7}$$

where

$$M = \frac{Z_2 \cos^2 2\psi_2 \cot P + Z_{5l} \sin^2 2\psi_2 \cot Q}{Z_3}$$

$$N = \frac{Z_2 \cos^2 2\psi_2 + Z_{5l} \sin^2 2\psi_2}{Z_3 \sin P \sin Q}$$

$$Z_\ell = \frac{\lambda_\ell c_\ell}{\cos \theta_\ell}$$

$$P = -\alpha_\ell l = -k_\ell l \cos \theta_\ell$$

$$Q = -\beta_\ell l = -k_\ell l \cos \psi_\ell$$

The subscript 1, 2 and 3 correspond to the numbered medium indicated in figure 7.11. The $z$ component of the propagation constant for the longitudinal waves is $\alpha$, while $\beta$ represents a like quantity for shear waves. The longitudinal and shear wave propagation constants are given by $k_\ell$ and $K_\ell$, respectively, and defined as $k_\ell = \omega/c_{L2}$ and $K = \omega/c_{S2}$, where $\omega$ is the angular frequency, $c_{L2}$ and $c_{S2}$ are the
FIGURE 7.11: Transmission characteristics for continuous waves as a function of angle of incidence on one-layer problem (after Barnard et al., 1975).
longitudinal and the shear wave velocities in the specimen of thickness \( l \).

The absorption coefficients in neither the water nor the sample were accounted for in these calculations. To account for absorption in the silicone rubber sample, if the absorption in water is considered negligible, the wave propagation constants, \( k_2 \) and \( K_2 \), would have to be made complex numbers. Therefore, the media used in these calculations were not realistic but can provide with an idea of the effect of angle of incidence of continuous waves on the specimen.

The \( x \) component of the propagation constants is \( \sigma \). These quantities may be mathematically related to the vectors \( \alpha_2 = k_2 \cos \theta_2 \); \( \beta_2 = K_2 \cos \phi_2 \) and \( \sigma = k_1 \sin \theta_1 = \alpha_1 = \beta_1 \).

The definition of \( \sigma \) is also a mathematical expression for Snell's law. \( \theta_2 \) and \( \phi_2 \) are the angles formed in the specimen by the intersection of longitudinal and shear wave vectors with the normal to the interface.

The velocities in the specimen were separately computed by the model discussed in chapter 2 and used in the computation of the transmission coefficient given by equation (7.7). Medium 1 and 3 were both considered to be water with \( c_1 = c_3 = 1480 \text{ m s}^{-1} \). Figure 7.12.a and 7.12.b show the results obtained from the calculations of the transmission loss in the decibel scale (20 \( \log T \)) for the measured and the nominal thicknesses of sample 1 (pure silicone rubber), respectively 29.175 mm and 30 mm, at the frequencies of 1 MHz, 3 MHz, 5 MHz and 8 MHz. For clarity, the results are presented shifted by 1 dB at each frequency investigated.

Note the clear difference in the results caused by changes from measured to nominal thickness (figure 7.12.a and 7.12.b), particularly at 8 MHz. At this frequency and normal incidence of waves a
FIGURE 7.12: Transmission loss at the interfaces for continuous waves as a function of angle of incidence at the frequencies indicated (absorption coefficient not accounted for). (a) Considering the measured thickness, and (b) the nominal thickness of sample 1 (thickness = width of sample).
transmission loss of approximately 0.5 dB is predicted for sample 1 if it had the nominal thickness, while it is 0 dB when considering the measured thickness. The oscillations in the transmission loss as a function of angle of incidence obtained at each frequency are due to reverberation of the waves inside the specimen, causing constructive and destructive interference upon emergence into water. It is expected that these oscillations would be somewhat reduced if absorption in the sample had been entered into consideration.

In pulse methods, the effect of absorption and the effect of reflections can be easily separated. In the case of continuous waves, both effects are inter-related. As a consequence of this inter-relationship one may not be able to separate them in a clear way. Unfortunately, time did not allow this study to be continued, as modifications in the computer programme would be necessary.

7.4.2 - ERRORS OF MEASUREMENTS

Upon consideration of sufficiently short pulsed waves and negligible absorption of ultrasound in water, the attenuation in the specimen given by equation (7.5) can be written as:

$$ \mu = -\frac{1}{l} \ln\left(\frac{A_r}{A_o}\right) - \frac{1}{l} \ln(\frac{1}{\alpha \tau}) $$  \hspace{1cm} (7.8)

From equation (7.8), "the most probable error" (Topping, 1979) in the attenuation in the specimen, $\xi_\mu$, is given by:

$$ \xi_\mu^2 = \left(\frac{\partial \mu}{\partial l}\right)^2 \xi_l^2 + \left(\frac{\partial \mu}{\partial A_r}\right)^2 \xi_{A_r}^2 + \left(\frac{\partial \mu}{\partial A_o}\right)^2 \xi_{A_o}^2 + \left(\frac{\partial \mu}{\partial \alpha \tau}\right)^2 \xi_{\alpha \tau}^2 $$  \hspace{1cm} (7.9)

where $\xi_l$, $\xi_{A_r}$, $\xi_{A_o}$ and $\xi_{\alpha \tau}$ are respectively the errors in the dimension of the specimen, in the received amplitude of signal with the specimen present in the acoustic field, in the received amplitude without the specimen in the field, and in the transmission coefficient at the
By solving the derivatives in equation (7.9), one can write:

$$\mathcal{E} = \pm \frac{1}{l} \left[ \left( \ln \left( \frac{A_r}{A_o} \right) + \ln \left( \frac{1}{\alpha_r} \right) \right)^2 + \frac{\xi_r^2}{A_r^2} + \frac{\xi_{\alpha r}^2}{A_o^2} + \frac{\xi_{\alpha \alpha r}^2}{\alpha_r^2} \right]^{1/2} \quad (7.10)$$

which gives the value in Neper per unit length. To obtain the value in the decibel scale, one needs to multiply equation (7.10) by the factor of 8.686.

From the calculations of the error bars on the attenuation measurements, it was found that they did not vary significantly from sample to sample. The major sources of errors of measurements were found to be the diffraction in the lower end of the frequency range and the precision of data reading from the XY-recorded data chart (when applicable). Therefore, for reference purposes, the estimated error bars were only drawn at 2 MHz in figure 7.4 (the two clear silicone rubber samples). The errors of measurements are further discussed in the following section.

7.5 - GENERAL COMMENTS

The results obtained with the time delay spectrometry (T.D.S.) technique had shown a higher attenuation than with the other techniques. This is particularly noticeable in the lower and the higher end of the frequency range used. The results obtained with the digitised pulse (wide band and tone burst excitation) gave the lowest attenuation. The results obtained by the use of the swept frequency technique lay between the T.D.S. and the digitised pulse techniques. The peaks in attenuation at frequencies below 1 MHz, measured with the T.D.S. technique, are believed to be associated with the equipment constraints.
A possible reason for the T.D.S. technique giving the highest attenuation for all groups of samples in most of the measurements, is that the samples were not aligned by the minimum time of flight but by the maximum received signal amplitude (see section 4.3.2) at the intermediate frequency of approximately 5 MHz. The transmission coefficient is dependent on the angle of incidence, as discussed in section 7.4.1. In that section, only two cases were considered, i.e., short pulsed waves and continuous waves, with no absorption or attenuation present. It is believed that both the time delay spectrometry and the swept frequency techniques would give a transmission coefficient somewhat between the two cases considered for the computation, because of the filter band-width used for the frequency span and sweep rate chosen, and the inherent absorption (or attenuation) in the specimens. Some calculations were carried out by Chivers et al. (1985) and it was concluded that it was unlikely to get any reflection through but the initial transmission signal. The attenuation in the specimen can effectively eliminate (or at least reduce considerably) the reflected waves, if any present. A good experimental evidence supporting this is that no (or in some cases negligible) interference patterns were shown in the frequency spectra of the specimens.

Although in the T.D.S. experiment the samples had been oriented in the acoustic field by the maximum received signal amplitude, the path length was longer than the width of the samples. Unfortunately, precise corrections to this were not possible because the angle formed by the orientation of each sample with respect to the acoustic axis was not recorded for two reasons: firstly, the very short time available for the experiment to be entirely performed (part of a series of experiments with the technique, Chivers et al., 1985); and secondly,
the lack of a calibrated rotational holder for the specimen. Only a visual estimate was made, being of the order of magnitude of 5° to 15°, and varied from sample to sample.

According to some theoretical calculations (without considering attenuation in the specimen), the dependence of the transmission coefficient upon the angle of incidence and frequency is very small compared to the attenuation itself. Nevertheless, the extra path, longer than the width of the sample, gave rise to a higher measured attenuation. By using the Snell's law of refraction of plane waves, if the angle of incidence was 15°, it was estimated that the extra path would be only about 0.05 cm longer than the width of the specimens (about 3 cm). This extra path would cause the measurement of attenuation at 5 MHz to be about 0.1 dB cm\(^{-1}\) greater in the clear silicone rubber. At 9 MHz, the worst case, it would be expected to be 0.25 dB cm\(^{-1}\) (and not about 1.5 dB as measured), which was of the same order of magnitude of error of reading the data from the XY-recorded data chart, i.e., ±0.3 dB cm\(^{-1}\). Considering the T.D.S. technique as pulsed, the magnitude of the transmission loss at 15° and 9 MHz would be approximately 0.09 dB cm\(^{-1}\), while at 0° it would be about 0.08 dB cm\(^{-1}\). Thus, both effects together, i.e., transmission loss and mis-orientation of the sample did not account for having measured a much higher attenuation. In section 7.2, it was shown that diffraction effects would account for an error not greater than 0.2 dB (at 1 MHz) in the measurement of attenuation. The diffraction effects were easily indentified in other experiments (using other measurement techniques) for about the same transducer separation as used for the T.D.S. experiment. However, the diffraction effects were not clearly evident to account for a higher measured attenuation throughout the frequency range investigated with the T.D.S. technique. Some other
effect (or effects) must have been responsible for having obtained this higher attenuation in all samples. The T.D.S. measurements of attenuation in the samples were carried out at about 16.8°C, whereas the other techniques were at 20°C. Chivers & Bacon (1983) had found a temperature slope of absorption in silicone rubber RTV-602 to be approximately \(-1.25 \text{ dB cm}^{-1} \text{°C}^{-1}\). The largest difference of the absorption results in either sample 1 or 20, between the measurements with the T.D.S. and the digitised pulse techniques (the largest difference) was about 1.5 dB cm\(^{-1}\) (at 8 MHz). If that figure of temperature dependence given by Chivers & Bacon (1983) is correct, this difference should then be about 4 dB cm\(^{-1}\). However, from the present results no definite conclusion can be drawn on this matter, because of the different procedure of alignment of the sample in the acoustic beam.

The bulges in attenuation in all samples in the range of 2-6 MHz, measured with the swept frequency technique and transducer separation of 200 mm, were originated from diffraction effects, as concluded from the experiments related with figures 7.2 and 7.3, and discussed in section 7.2.

The results are further discussed in the following chapter.
CHAPTER 8

DISCUSSION
CHAPTER 8

DISCUSSION

8.1 - NUMERICAL MODEL.

Although the derivation of Waterman & Truell's (1961) formulation of multiple scattering of waves is complicated (see chapter 2), the results of this derivation are mathematically relatively simple and easily programmed in a computer as a numerical model. A computer programme that calculates the effects of multiple scattering of waves on velocity and attenuation on the basis of Waterman & Truell's formulation was already available. That computer programme was written in standard fortran IV by Dr. L.W. Anson, who kindly provided it for use in the study reported here. The original form of Dr. Anson's programme was then adapted by the author of this thesis to model the inhomogeneous systems used for the present investigation. The adaptation involved introducing the frequency dependent phase velocities in the constituent materials of the composites (silicone rubber and glass) which were determined from the experimental data of the frequency dependent absorption, by utilising the Kramers-Kronig relationship between phase velocity and absorption (see sections 8.2 and 8.3). The calculation of the group velocity in the composites was also introduced into the programme. The programme was also modified to allow interactive access by the user, in the form of a menu chart. The input parameters for the programme were the characteristics of wave propagation in the constituent materials of the composites, which are
the frequency dependent longitudinal and shear velocity and absorption (determined by the Kramers-Kronig relationship method - sections 8.2 and 8.3), the densities of the constituent materials of the samples (section 3.4), the volume concentration of the glass beads (column 4 of table 3.2.b), and the histograms of the size distribution of glass beads, shown in figure 3.2, section 3.3.

Some preliminary computational studies were carried out comparing the results of velocity and attenuation with the first two terms considered and with all terms considered in equation (2.8). For the materials used (glass beads embedded in a silicone rubber matrix) and the volume concentration of up to 5% of scatterers, no significant difference between the results was observed in the frequency range of interest, i.e., 0 to 10 MHz, for all four distribution of scatterer sizes. There is no evidence in the literature that multiple scattering has a significant effect at the concentrations used in this investigation.

There are several parameters involved in the theoretical calculation of the scattering effects and some of them are not precisely known. For instance, it was observed that by small variations in the longitudinal and shear absorption in the glass beads, both the phase velocity and the group velocity in the composite were affected. Particularly, the group velocity would exhibit even sharper variation on frequency dependence, if the shear absorption in the beads was nil. This is further discussed in the subsequent sections of this chapter.

The results of this numerical solution of the analytical approach, for the specimens of chapter 3, are presented in chapters 6 and 7, where the experimental results of velocity and attenuation as a function of frequency are shown. A good agreement was found in the
comparison of the theoretical predictions with the experimental observations, as discussed in section 8.4.1.

The computational model was further modified to calculate the velocity and attenuation as a function of concentration at a given frequency. The results of this computation are presented and discussed in section 8.4.2.

8.2 - ABSORPTION COEFFICIENT

The formulation of the theoretical model discussed in chapter 2 requires a knowledge of the longitudinal and the shear components of the absorption coefficient in the matrix as well as in the scattering material embedded in the matrix, in addition to the same components of the wave velocity.

If the shear absorption coefficient in the material of interest is not readily available, it can be estimated from a relationship between the longitudinal absorption coefficient, $\alpha_L$, and the ratio of the corresponding wave velocities, $c_s$ and $c_L$, as given by Madigosky & Warfield (1984):

$$\alpha_s = \frac{\alpha_L}{2} \left( \frac{c_L}{c_s} \right)^2 \quad (8.1)$$

In the case of rubber-like material, the velocity ratio may be as large as 10 or 100 (see appendix B). Consequently, even a small longitudinal absorption will give rise to a large shear absorption, as was observed experimentally by Madsen et al. (1983). In the case of glassy material, the velocity ratio is low (about 2) and the longitudinal absorption is about the same order of magnitude as the shear absorption. Table 8.1 tabulates the relevant quantities for the materials in question.
The frequency dependence of the absorption coefficients of the constituent materials of the composites were assumed to be in the form
\[ \alpha = a f^b \]  \hspace{1cm} (8.2)
where \( a \) and \( b \) are constants and \( f \) is the frequency (Wells, 1977; Williams, 1983). With the frequency in the megahertz scale, \( a \) is the absorption coefficient at 1 MHz.

The value of the constants \( a \) and \( b \) for the silicone rubber RTV-602 were empirically determined from the experimental data of figure 7.3 (sample 1 and 20), by drawing a straight line through the data points drawn on a log-log scale. With the available data of the longitudinal and shear velocities and longitudinal absorption coefficient in the RTV-602 (see chapter 6, chapter 7 and appendix B), the shear absorption coefficient was subsequently estimated from equation (8.1) at the frequency of 1 MHz. The value of the constant \( b \) for the shear waves was assumed to be the same as for the longitudinal waves. The values of longitudinal and shear velocities and the values of \( a \) and \( b \) for both longitudinal and shear absorption in the silicone rubber RTV-602 are given in table 8.1.

The longitudinal absorption coefficient in the glass beads (scattering material) was assumed to be the one given by Kaye & Laby (1973) and considered to be linear with frequency (\( b = 1 \)) within the range used (0-10 MHz). No data on shear absorption in glass was found in the literature. Therefore, its value had to be estimated. Equation (8.1) was used for this estimation, as in the case of the silicone rubber, giving the value indicated in table 8.1. The longitudinal and shear velocities in the glass, indicated in table 8.1, were also extracted from Kaye & Laby (1973).
TABLE 8.1
Parameters for equation (8.1) and (8.2).

<table>
<thead>
<tr>
<th>material</th>
<th>$c_L$ (m s$^{-1}$)</th>
<th>$c_S$ (m s$^{-1}$)</th>
<th>$a$ (dB cm$^{-1}$)</th>
<th>$b$</th>
</tr>
</thead>
<tbody>
<tr>
<td>RTV-602</td>
<td>1020.6</td>
<td>40</td>
<td>0.5</td>
<td>162.8</td>
</tr>
<tr>
<td>GLASS</td>
<td>5660</td>
<td>3420</td>
<td>0.017</td>
<td>0.023</td>
</tr>
</tbody>
</table>

8.3 - VELOCITY DISPERSION

In an absorbing (or attenuating) medium, the frequency dependent absorption (or attenuation) coefficient is related to the frequency dependent phase velocity. Therefore, one can determine the dispersion of the waves from measurements of the frequency dependent absorption (or attenuation) and vice-versa. In an acoustic system, a method that can be used to determine one of the frequency dependent parameters from the measurements of the other is to use the so called Kramers-Kronig relationship between absorption (or attenuation) coefficient and phase velocity provided that the conditions of causality and linearity are satisfied, i.e., that the effects do not precede their causes, and that to some approximation the response of the system is linearly proportional to the stimulus (O'Donnell et al., 1978; O'Donnell et al., 1981; Beltzer et al., 1983). The Kramers-Kronig relationship can be shown to yield a description of the nearly local ultrasonic attenuation-dispersion relationship in the absence of rapid variations with frequency such as those associated with a sharp resonance (O'Donnell et al., 1981).

The relationship between phase velocity and absorption (or attenuation) coefficient can be used in the following manner to determine the dispersion of the waves:
\[ c(\omega) = c_0 + \frac{2c^2}{\pi} \int_{\omega_0}^{\omega} \frac{\alpha(\omega') \ d\omega'}{\omega'} \]  

(8.3)

where \( c_0 \) is the phase velocity at the angular frequency \( \omega_0 \), and \( \alpha(\omega') \) is the absorption (or attenuation) at the angular frequency \( \omega' \).

The Kramers-Kronig relations method was used to determine the longitudinal and shear phase velocities in the basic materials used in the specimens of inhomogeneous viscoelastic media, i.e., in silicone rubber and in glass. The value of \( c_0 \) in the silicone rubber RTV-602 was obtained by averaging nine extrapolated values of zero frequency velocity of nine samples investigated, i.e., the two pure silicone rubber samples (1 and 20), and seven other samples containing low concentration of beads (samples 9, 10, 11, 14, 15, 16 and 17). The average value obtained was \( c = 1019 \pm 1 \text{ m s}^{-1} \). The zero frequency shear velocity in the silicone rubber was estimated to be 40 m s\(^{-1}\) (see appendix B). The dispersion in glass is very small and, therefore, its zero frequency velocities were assumed to be those given in table 8.1, which were extracted from Kaye & Laby (1983). For reference purposes, the calculated elastic constants of the samples and of their constituent materials are given in appendix B. The absorption coefficient used in equation (8.3) for each material and mode of propagation are given in table 8.1, section 8.2.

Equations (8.1), (8.2) and (8.3) defined the absorption and dispersion in the basic constituent materials of the specimens, based on experimental observations. These equations were implemented in the computational model described in section 8.1, which used the analytical approach discussed in chapter 2.
8.4 - ANALYSIS OF RESULTS

8.4.1 - FREQUENCY DEPENDENCE

GENERAL COMMENTS

The results in discussion here are presented in chapter 6 and 7, concerning respectively the velocity and the attenuation in all the inhomogeneous viscoelastic specimens studied as a function of frequency. Some general comments with respect to the experimental data are given in those two chapters. For reference purposes, the mean values of the propagation parameter \( \kappa_a \), concerning each group of bead sizes, are given in appendix C.

As reported in chapter 3, there is an uncertainty in the measured concentration of glass beads in the samples (see table 3.2.b). In some cases, errors in measured concentration can significantly affect the calculated velocity and attenuation. The effect of variation in the percentage volume concentration upon the calculated velocity and attenuation is presented in figures 8.1 and 8.3. From these figures, the possible variation in velocity and attenuation due to the uncertainty in the concentration can be estimated.

From the results of the numerical model, the velocity appears to be a more complicated function of frequency than the attenuation. The attenuation has a general trend to increase with frequency, whereas the velocity can either increase or decrease, depending on the size of the scatterers compared with the wavelength and on their concentration. The frequency dependent attenuation can in some cases exhibit a plateau, or a bulge in other cases. The results obtained seem to follow a feasible physical explanation given by Morse & Ingard (1968). When the wavelength of an incident wave is much greater than the bead
radius, the scattered wave spreads more or less isotropically. This causes a rapid increase in the frequency dependent attenuation and velocity. As the frequency is increased, forward scattering becomes dominant, i.e., in the direction of the incident wave. Less interaction between scattered waves consequently results, and less energy is deflected from the original incident wave. In this transition, the attenuation may present a plateau region, i.e., frequency independent, as predicted by the numerical model and experimentally observed.

The inclusion of glass beads increases the moduli in silicone rubber. At the same time, the density is increased at a greater rate than the moduli. This results in a decrease in velocity at very long wavelength (see appendix B). As the frequency is increased, a quick rise in phase velocity occurs, after which its frequency dependence is varied according to the bead size and concentration. It can continue increasing, although at a much lower rate, or present a series of maxima and minima.

From the definition of group velocity it was expected that sharp variations in its frequency dependence would result in the numerical computation of the theoretical model. In the numerical model, the magnitude of these variations are related to the frequency step used and to the precision of the computed complex wave propagation constant in the composite. The computer programme was written in standard fortran IV and this language allows only single precision on complex numbers (accuracy of 7 decimal digits on PRLME computers), leading to possible numerical artefacts. The frequency step used was 75 kHz. A shorter frequency step would be preferable, but this would considerably increase the time of computation (average of about 5 minutes real time per sample). Examples of these sharp variations in group velocity are
seen in figures 6.3 to 6.7 in the frequency range of 0.1 to 0.4 MHz (lower frequency for the largest of the beads and higher frequency for the smallest of the beads). In particular, sample 4 also showed these variations at about 5.8 MHz and 5.9 MHz. However, some local and smooth variation in the group velocity as a function of frequency, presenting smooth peaks and dips, are not related to numerical artefact in the computation, but to the composite themselves. Variations of this kind, including plateaus, are also observed in the curves of phase velocity and attenuation, agreeing with the experimental results. The magnitude of these variations are enhanced with the increase of concentration of scatterers, but remain at the same frequencies. As the size of the scatterers is reduced, the peaks, dips and plateaus occur at higher frequencies. It is thought that these local variations in velocity and attenuation as a function of frequency in the composite are related to resonance effects taking place in the glass beads, but are not sharp because of the absorption in the silicone rubber matrix, and the distribution of bead sizes. Gaunaurd & Barlow (1984) compared hypothetical elastic and viscoelastic media containing a distribution of air-cavities, and found that the presence of viscosity broadens the resonance peaks and dips, and reduces their amplitude. They also found that viscosity slightly shifts the resonance peaks from their values in elastic media.

By definition, the group velocity has a general trend to be higher than the phase velocity if the phase velocity increases with frequency. In most cases the experimental data (including error bars) lie on the phase velocity curves. In the 3 cases (samples 3, 4 and 26) that there was significant difference between the data points and both theoretical curves, calculations were performed and indicated that any concentration variations implicit in column 5 of table 3.2.b would not
affect the theoretical curves sufficiently to produce better agreement. The reader is referred to figure 8.1 to acquire a knowledge of the order of magnitude of variation that the error in concentration would give in the theoretical curves of phase velocity.

A potential cause for the significant difference between the experimental data points and the theoretical predictions of velocity was thought to be the instability of the temperature in the water bath. The acoustic velocity is very sensitive to environmental conditions such as temperature and pressure. For more accurate results, the measurements should have been taken at less temperature disturbance (within at greatest ±0.05°C), since the pressure was kept constant. A discussion concerning the problems encountered during the measurements of velocity is given in chapter 6.

In the remainder of this section each group of samples (described in chapter 3) is discussed separately.

**GROUP I: figures 6.3 and 7.4**

The concept of velocity dispersion is directly related to the absorption (or attenuation) coefficient of the propagating medium, as discussed in section 8.3. A good agreement for dispersion in the pure silicone rubber was obtained between experimental observations and the theoretical prediction based on the Kramers-Kronig relationship between phase velocity and absorption. The measurements showed a higher absorption in sample 1 than in sample 20. This was also observed in the preliminary results presented in figures 7.2 and 7.3.

**GROUP II: figures 6.4 and 7.5**

As expected, the attenuation increased with the concentration. Plateaus and bulges in attenuation and in phase velocity were observed
at the same frequencies that peaks were observed in the calculation of the group velocity, the magnitude of which increased with the concentration. The peaks, bulges and plateaus in velocity and attenuation appear at nearly regular frequency intervals, i.e., at 2.6 MHz, 4.9 MHz, 7.1 MHz. (The sharp variation in the calculated group velocity at about 0.1 MHz at any of the samples, and those at 5.8 MHz and 5.9 MHz for sample 4 are discussed in the first part of this section). These peaks, bulges and plateaus suggest resonance taking place in the glass beads, but absorbed by other mechanisms regulated by the silicone rubber matrix. The origin of the small peaks at intermediate frequencies between the occurrence of the large peaks in group velocity are not precisely known. However, it was noticed that slight changes in the longitudinal and shear absorption coefficients in the glass beads affect the magnitude of the peaks and dips in the computed group velocity. They can be either enhanced or smoothed out.

**GROUP III: figures 6.5 and 7.6**

Similar effects to group II were observed in group III. As the size of the beads was reduced from group II to group III, the largest peaks in the calculated group velocity occurred at higher frequencies (4.9 MHz and 9.4 MHz). (The sharp peak at about 0.2 MHz is discussed in the first part of this section). A particular reference is made to sample 8. This sample is the only case in the set of 32 samples where the attenuation calculated from the theory differs significantly from the measurements. Therefore, it seems more reasonable to believe that the theory is valid for this specimen and that the discrepancy must lie in the measured concentration, i.e., the concentration and its error bar for this sample were under-estimated.
GROUP IV: figures 6.6 and 7.7

The group velocity exhibits bulges and one clear trough in the vicinity of 6 MHz. (The sharp peak in group velocity at about 0.3 MHz is discussed in the first part of this section). For a high concentration of beads, the phase velocity passes through a maximum at about 1 MHz, followed by a steady slow drop in magnitude, converging to a constant value. The attenuation consistently increases as the concentration of scatterers is increased. However, as the attenuation increases with the frequency, a poorer signal to noise ratio is achieved and the measurements consequently started to fail (indicated by the arrows on the graphs).

GROUP V: figures 6.7 and 7.8

In this group of samples, the effects of scattering were similar to the ones in group IV. (The very sharp peak observed in the calculation of the group velocity appeared at a slightly higher frequency than in the case of group IV, i.e., at about 0.4 MHz, and a discussion related to this is given in the first part of this section). The lowest level of the trough in group velocity was shown at about 8.3 MHz.

8.4.2 - CONCENTRATION DEPENDENCE

GENERAL COMMENTS

The variation of phase velocity and attenuation as a function concentration at selected frequencies are discussed in the present section only on the basis of the theoretical predictions, since comparisons with the experimental observations have already been made as a function of frequency for all available concentrations.
Approximate values of the propagation parameter $ka$ corresponding to each group of beads and frequencies investigated are tabulated in appendix C for reference purposes.

The formulation of multiple scattering of waves of Waterman & Truell (1961), used in this investigation, has been found valid for concentrations below 15% by volume for lead spheres in epoxy matrix (Sayers & Smith, 1983). The largest concentration of scatterers used in the present investigation was 5% volume, which is well within the safe limit of validity of the theory. The velocity dispersion and the absorption coefficients in both silicone rubber matrix and in the glass beads have also been accounted for.

**Phase Velocity**

Figures 8.1 and 8.2 show the theoretical variation of velocity as a function of the concentration of glass beads in suspension in the silicone rubber RTV-602 matrix at selected frequencies. The abscissa of figure 8.1 is the concentration of beads in terms of percentage volume, whereas in figure 8.2 it is in terms of the absolute number of beads per cubic centimeter, i.e., the number density of scatterers in the matrix.

The phase velocity is shown to be linearly dependent with concentration for any of the frequencies considered. Figure 8.1 shows that there are points of intersection, at which for a given percentage by volume concentration, the same value of velocity is achieved at different frequencies. Similarly, at certain frequencies, the velocity is independent of concentration, as indicated by the dashed-line for each group number. In figure 8.2, the Roman figure indicates the group number of the glass bead diameter, whereas the subscript corresponds to the frequency in megahertz. The velocity can be increased with a
FIGURE 8.1.a: Theoretical ultrasonic phase velocity as a function of volume concentration of glass beads embedded in silicone rubber matrix (RTV-602) at the frequencies and group number indicated.
GROUP IV: Frequencies: 1, 2, 3, 4, 6, 8, 10 MHz.

FIGURE 8.1.b: (see figure 8.1.a).
FIGURE 8.2: Theoretical phase velocity as a function of the number density of glass beads embedded in the silicone rubber (RTV-602) matrix. The Roman figures correspond to the group number of bead size, and the subscripts indicate the frequency in megahertz.
decrease of the diameter of the scatterers, although not in a simple manner. For groups II and III, the velocity, at any of the frequencies considered, decreases linearly as the number density of the glass beads is increased. A higher velocity is observed for group III than for group II, and the negative slope is made smaller as the frequency is increased. Groups IV and V show even higher velocity than groups II and III, and at the same time are less sensitive to the number density of beads. However, group IV shows a higher velocity than group V at 1 MHz, but lower at 4 and 8 MHz. At 1 MHz, the velocity increases with the number density for either group IV or V, whereas at 8 MHz a decrease in velocity results from an increase in the number density.

ATTENUATION

Figure 8.3 shows the attenuation as a function of the volume fraction of glass beads scatterers embedded in the silicone rubber RTV-602 matrix at the same frequencies used for the computation of the phase velocity, for all four groups of bead sizes. The attenuations represented by the dashed line are at the same frequencies which gave velocity independent of concentration.

As for the velocity, the attenuation of ultrasound in a random distribution of glass beads scatterers in RTV-602 is linear with the concentration. However, the attenuation is always increasing with increasing concentration, because of the increase in the scattering of waves.

For a given group distribution of glass bead scatterer size, as the frequency is increased, the slope of the attenuation with the concentration is increased. No clear inter-relationship was found between the groups of scatterer sizes.
FIGURE 8.3.a: Theoretical attenuation as a function of volume concentration of glass beads embedded in silicone rubber matrix (RTV-602) at the frequencies and group number of bead size indicated. The dashed lines correspond to the frequency at which the phase velocities were independent of concentration.
FIGURE 8.4: Theoretical attenuation as a function of the number density of glass beads embedded in the silicone rubber (RTV-602) matrix. The Roman figures correspond to the group number of bead sizes, and the subscripts indicate the frequency in megahertz.
A similar graphical representation of the velocity, is presented for the attenuation as a function of the concentration of the scatterers in figure 8.4, where the absolute number of scatterers per unit volume (number density of scatterers) is used as the abscissa. As for the graphical representation of the phase velocity, the Roman figures correspond to the group number and the subscript to the frequency in megahertz. For a given frequency, the attenuation slope with the number density of scatterers is decreased as the scatterer size is made smaller.
CHAPTER 9

CONCLUSION

9.1 REVIEW

The investigation of the propagation of ultrasound in inhomogeneous viscoelastic materials reported here has been introduced in chapter 1 of this thesis. It was centered on a series of specimens of silicone rubber matrix (RTV-602) with a range of randomly distributed glass beads in suspension, as models of inhomogeneous viscoelastic media. The specimens were classified into five groups: one group of specimens of clear silicone rubber plus four other groups according to the bead size range distribution (mean diameter: 582 μm, 317 μm, 101 μm and 63.8 μm). The nominal size of the specimens was 3 cm x 4 cm x 5 cm. The fractional volume concentration of the glass beads in suspension in the silicone rubber matrix was estimated from the measurement of the effective densities of the constituent materials of the specimens and of the specimens themselves.

An extended investigation was centered on the effects of diffraction on velocity and attenuation measurements. The effects of diffraction in distilled water were compared with plausible mathematical models of transducers.

Measurements of ultrasonic velocity and attenuation in the specimens were made by using the substitution method (relative to water) in association with through-transmission techniques. Various experimental arrangements were used for the measurement of velocity and
attenuation as a function of frequency (up to 10 MHz), with the specimen immersed in a distilled water bath, at approximately constant temperature and pressure. The results of velocity were obtained by measuring the time of flight of the second cycle of ultrasonic tone bursts by using the time interval averaging technique. The attenuation measurements were made with the following techniques: digitised peak amplitude of wide band pulses; digitised peak amplitude of tone bursts; time delay spectrometry; and simple swept frequency technique. The only set of measurements obtained with a receiving transducer different in design from the transmitting one, i.e., a small receiver hydrophone of 1 mm in diameter, was made with the time delay spectrometry technique, for comparison with a set of measurements obtained with the same technique and experimental arrangements, but employing a pair of identical transducers (one as transmitter and the other as receiver). The experimental results were compared with a theoretical prediction approach, which involved the combination of the multiple scattering of waves of Waterman & Truell (1961) and the Kramers-Kronig relationship between phase velocity dispersion and absorption coefficient in the constituent materials of the specimens. The relationship between the phase-velocity dispersion and the absorption coefficients in the constituent materials of the specimens (glass and silicone rubber) were based on empirical observations.

The size distribution and the fractional volume concentration of the glass beads in suspension in the silicone rubber matrix were used in the computation of the effect of the presence of the beads (i.e., the scatterers) on the ultrasonic wave propagation.
To overcome some problems associated with experimental constraints, some preliminary studies were necessary. Among them, there were some very specific studies related with diffraction effects on measurements of velocity and attenuation. It was found that it does not matter where the specimen is placed in the acoustic field, but that it does matter where the receiving transducer is located with respect to the transmitting transducer. Both velocity and attenuation measurements can be affected.

When using a comparative method (or the substitution method) for measurements of velocity of ultrasound in a specimen of interest, it is important that the velocity of the ultrasonic waves in the reference medium (for instance distilled water) is measured and not taken from a reference table. If a wrong velocity in the reference medium is used, there will result a wrong velocity in the specimen. Diffraction is one of the causes of obtaining wrong values of velocity.

A detailed experimental study was dedicated to the quantification of diffraction effects in water alone, which found very good agreement with the theoretical predictions for both phase velocity and attenuation. For \( d/\lambda < 50 \), where \( d \) is the diameter of the transducer and \( \lambda \) is the wavelength, the measured phase velocity is greater than the plane-wave velocity. The phase velocity monotonically (but non-linearly) increases as the distance between the transmitter and the receiver is decreased. As the ratio \( d/\lambda \) is increased, the measured phase-velocity approximates the plane-wave value, becoming independent of the transducer separation. The approximate plane-wave value of velocity in distilled water was always found to be slightly lower than the values normally found in the literature. From the measurements of loss of amplitude of signal due to diffraction, it was found that the
7.5 MHz transmitting transducer used did not behave like a piston source for frequencies below 4 MHz, but more like a plate, the exciting function of which appeared to have varied with the frequency. From this experiment, it was also found that the absorption coefficient in distilled water was slightly greater than the values normally found in the literature.

The computations of the numerical solution of the theoretical approach of multiple scattering of waves in the specimens as a function of frequency were carried out on the basis of the estimated fractional volume concentration of the glass beads in suspension in the silicone rubber matrix. With these values of volume concentration, the results of velocity and attenuation predicted by the theoretical approach were shown to be in very good agreement with the experimental ones. However, in some cases, difficulties were encountered on the quantitative comparison of the velocity results due to experimental constraints like temperature fluctuations. Nevertheless, the experimental results of velocity have shown a good qualitative agreement with the theoretical prediction of the phase velocity. Only one case showed a significant discrepancy in the attenuation, which may have been an indication that the measured concentrations in that specimen was under-estimated. The results show that the measurement of attenuation is a very reliable method of determining the approximate concentration of spherical particles, or the average diameter of particles in composites.

The scattering effects may be seen in both velocity and attenuation. However, the presence of scatterers affects the velocity in a more complicated manner than the attenuation. The velocity may increase or decrease with frequency, depending on the size and concentration of beads in suspension. Peaks, bulges, dips and plateaus
were, therefore, observed in the velocity as a function of frequency. The attenuation has a general trend to increase with frequency. Bulges, dips and plateaus were also observed in attenuation at the same frequencies as for the velocity. These were indications of resonances taking place in the glass beads, but being absorbed by the viscoelastic properties of the silicone rubber matrix.

The velocity can either increase or decrease linearly with the volume concentration, depending on the size of the beads and the frequency. At certain frequencies, however, the phase velocity was shown to be independent of the volume concentration, i.e., insensitive to scattering. The attenuation was also shown to be a linear function of volume concentration, but always increasing with increasing volume concentration. This indicates that the scattering probability increases with an increasing concentration of beads, for the concentration ranges studied here. The slope of this linear increase was dependent on frequency and on the size of beads.

The slope of the linear function of attenuation versus number density of scatterers (beads) decreased with decreasing scatterer size for all of the frequencies investigated. However, the slope of the velocity as function of the number density was either negative or positive, depending on the frequency and size of the beads.

From the theoretical graphical representation of the variation of velocity and attenuation as a function of concentration of particles, it is then possible, and easy, to determine the concentration of particles in a given specimen, by a simple comparison with the measured velocity and attenuation, provided that the size distribution of the particles is known. Similarly, the average size distribution can be determined if the concentration is known. Although the attenuation was found to be a more reliable acoustic parameter of measurement than the
velocity, both parameters are needed for a full characterization of the scattering and absorbing media. From graphical functions like the ones described above, it is also possible to estimate the magnitude of error bars in the determined concentration (or size) from the estimated error bars of measurements of the acoustical properties of the specimen. These type of graphical functions are therefore very useful 'tools' in non-destructive testing for materials characterization.

This thesis provides what is believed to be the first proof that the theory of multiple scattering of waves of Waterman & Truell (1961), in combination with the Kramers-Kronig relationship between absorption and phase velocity in the constituent materials of the composite, is valid for a solid viscoelastic matrix with elastic spherical scatterer inclusions. Thus the theory can now be used with confidence to: [a] describe the ultrasonic propagation in inhomogeneous viscoelastic materials; [b] determine (non-destructively) the concentration, or size of scatterers, or other unknowns like bulk and shear moduli; and [c] design composite materials for specific applications. An example of such specific applications is the design of biological tissue models for use in assessment of ultrasonic scanners or training of personnel (Chivers et al., 1983). Other examples can be listed as the design of materials with: [a] low/high acoustical attenuation; and [b] acoustic impedance matching (or mismatching). Applications of these are encountered, for instance, in the design of backing of transducers, and design of acoustic chambers.
9.3 - SUGGESTIONS FOR FURTHER WORK

The investigation of diffraction effects on measurements of velocity and attenuation in the finite specimens immersed in water lacked proper theoretical prediction. The theoretical analysis of phase-shift and loss was made on the basis of an infinite continuous medium. It is believed that, in principle, the insertion of a finite lossy specimen of different acoustic properties from the surrounding medium would change diffraction effects upon the emergence of the waves. The presence of a finite specimen of such a nature may effectively act as an acoustic lens in the field. Therefore, a theoretical and experimental study concerning this problem is worthy of investigation, with specimens of different geometrical shape and dimensions, placed at different positions in the acoustic field.

After the measurement of effective density of the specimens (for subsequent determination of concentration of beads in the specimens) it was realised that the measurements of volume of the specimens could have been taken ultrasonically. The measurements of volume of the specimens were taken from the water displacement method (section 3.5) read from a measuring cylinder accurate to ±0.1 cm³. An ultrasonic transducer operating at a sufficiently high frequency (d/λ >.50 to avoid effects of diffraction) can be used to measure the volume of water displacement from measurements of transit time of an echo-signal from the water/air interface. Shortage of time did not allow the volume measurements to be re-taken by using this method. The transducer may be coupled to the bottom end of the measuring cylinder of figure 3.3, or even better, to a beaker directly linked to the specimen/water container. Calibrations can be made before or after the immersion of the specimen in the water container, by immersing blocks of known volume. A plot of transit time versus water volume
displacement can be drawn and subsequent volume of specimens can be easily obtained. The final measurements and the calibration should be carried out at the same water temperature. Alternatively, a liquid in which the dependence of the ultrasonic velocity upon temperature is less than that of water, and that the viscosity is also low, may be used instead of water. The precision of the measurements will depend on the transit time measurement device; on the alignment of the acoustical axis of the transducer perpendicular to the water/air interface; the calibration procedure; and temperature fluctuations.

The experiments performed for the investigation reported in this thesis were all made at the temperature of about 20°C. The temperature dependence of velocity and attenuation in the specimens used are unknown, and therefore, they should receive some attention.

Although some evidence exists (in the literature) that the multiple scattering of waves formulation of Waterman & Truell (1961) is valid for volume concentration of scatterers of at least 15% (for elastic materials) it is not yet very clear which parameters define this limit. They are thought to include the scattering strength. As far as the author is aware, no work concerning viscoelastic materials has been done to date. Therefore, it is not yet known how high the concentration would have to be for that formulation no longer to be physically meaningful. Therefore, this determination is worthy of investigation. The largest concentration of glass beads in silicone rubber matrix investigated in the presently reported research work was just below 5% by volume.

In this work, only specimens with glass beads in silicone rubber received attention, as models of inhomogeneous viscoelastic media. Therefore, it is suggested that other materials should also be investigated, and it is not known how applicable the Waterman
& Truell's formulation is to, for instance, biological materials and many geological materials. An investigation on these type of materials would be, potentially, of great use for diagnostic purposes.
APPENDIX A

FREQUENCY SPECTRA OF THE TRANSDUCERS
The frequency spectra of each pair of identical transducers of nominal frequency 1 MHz, 2 MHz and 7.5 MHz were measured by means of the swept frequency technique described in sections 4.4.4 and 5.6, with the transducers immersed in water.

The 1 MHz and the 2 MHz transducers were manufactured by T.M.S. (presently Systems & Instrumentation). They were unfocussed, and had 15 mm diameter piezoelectric ceramic elements (ref. No.: M1, 15E, serial No. 41032 and 61906; and M2, 15E, serial No. 61907 and 61908). The 7.5 MHz transducers were manufactured by Panametrics Acuscan Immersion Transducers. They were unfocussed and had 12.7 mm diameter piezoelectric ceramic elements (ref. No. V-320, serial No. 39651 and 39652).

The frequency spectra of the pair of 7.5 MHz transducers were obtained in connection with the study of diffraction phenomena presented in chapter 5, and they are illustrated in figure 5.12. The reader is referred to that chapter for the necessary information concerning these transducers. As a point of interest, the output power generated by either of these transducers was measured by Dr. L.W. Anson, with the use of a radiation force balance. The maximum output power was about 7 mW at 7.5 MHz with an excitation voltage of approximately 6 V peak-to-peak (Anson, 1985).

The frequency spectra of the other two pairs of transducers, i.e., the 1 MHz and the 2 MHz transducers, are presented in this appendix for
FIGURE A.1: Frequency spectra of a pair of T.M.S. transducers immersed in water at a separation of 220mm. Diameter of transducers 15mm. Nominal frequency: (a) 1 MHz., and (b) 2 MHz. The temperature of water at which the spectra were obtained and the serial No. of the transducers are indicated in the figures.
purpose of documentation. The frequency spectrum of each pair of transducers was obtained with the transducers immersed in a water bath at a separation of 220 mm. No diffraction correction and no compensation for the absorption in water were made. The temperature of water was 19.8°C and 18.5°C for the spectra of the 1 MHz and the 2 MHz transducers, respectively. The frequency spectra are presented in figure A.1.a and A.1.b. The frequency response of each pair was also measured by exchanging the transmitter as receiver and vice-versa, and no difference in spectrum was observed.

Because diffraction correction and compensation for the absorption coefficient in water were not made, the quality factor \( Q = \frac{f_0}{\Delta f_{3dB}} \) of the transducers could only be roughly estimated from the frequency spectra obtained. They were \( Q(1 \text{ MHz}) = 6.4, \ Q(2 \text{ MHz}) = 7.2 \) and \( Q(7.5 \text{ MHz}) = 1.7 \), where the number in brackets indicate the pair of transducers concerned.
APPENDIX B

ELASTIC CONSTANTS OF THE SAMPLES
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The elastic constants of solid substances are manifestations of intermolecular forces and, as a consequence, they are related to other physical quantities. Both the bulk and shear moduli of an anisotropic solid can be determined by measuring the velocity of the longitudinal and shear elastic waves. In a two-phase medium, once the elastic constants of each component are known, the effective elastic constants and velocities in the composite can be calculated.

The effective bulk modulus of the composite, $B^*$ (the asterisk does not denote the complex conjugate but the composite), is related to each component material of the two phase-medium by

$$\frac{B^* - B}{3B^* + 4G} = \frac{V}{3B^* + 4G} - \frac{B^1 - B}{3B^1 + 4G}$$  \hspace{1cm} (B.1)

where $B$ and $B^1$ are respectively the bulk modulus of the matrix and of the inclusion; $G$ is the shear modulus of the matrix and $V$ is the fractional volume concentration of the inclusions (Kuster & Toksoz, 1974).

Similarly, the effective shear modulus of the composite, $G^*$, is given by

$$\frac{G^* - G}{6G^*(B + 2G) + G(9B + 8G)} = \frac{V}{6G^*(B + 2G) + G(9B + 8G)} - \frac{G^1 - G}{6G^1(B + 2G) + G(9B + 8G)}$$  \hspace{1cm} (B.2)

where $G^1$ is the shear modulus of the inclusion material and the other symbols are signified in the previous paragraph.
The corresponding longitudinal and shear wave velocities are

\[ c_{l*} = \left[ \frac{B^* + 4.3 \, G^*}{\rho^*} \right]^{1/2} \]  

(B.3)

and

\[ c_{s*} = \left[ \frac{G^*}{\rho^*} \right]^{1/2} \]  

(B.4)

where \( \rho^* \) is the density of the composite.

The bulk and shear moduli of the composites (the samples described in chapter 3) were calculated at the static state, i.e., at zero frequency.

The value of the static longitudinal velocity of the silicone rubber RTV-602 matrix was obtained from the average of nine experimental data (of nine samples in chapter 6) extrapolated to zero frequency. The two pure silicone rubber samples and seven other samples containing low concentration of beads in suspension, i.e., samples 1, 20, 9, 10, 11, 14, 15, 16 and 17, were taken for this purpose. The average value of the static longitudinal velocity encountered was 1019 ± 1 m s\(^{-1}\).

The shear velocity in soft rubber is generally found in the literature to be much lower than the longitudinal velocity (a ratio of 10 to 100). Measurements of shear velocity in rubber materials very difficult because of their high shear attenuation. Krevelen & Hoftyzer (1976) give the shear modulus of natural rubber as \( G = 3.5 \times 10^5 \) Pa, which gives a shear velocity \( c_s \approx 20 \) m s\(^{-1}\), and silicone rubber (without specification of type) as \( G = 1.5 \times 10^5 \) Pa yielding \( c_s \approx 12 \) m s\(^{-1}\). The only other published data concerning silicone rubber material, which the author is aware of, is that of Madsen et al. (1983). The silicone rubber that they investigated was the RTV-615 which has a slightly higher longitudinal velocity (about 1025 m s\(^{-1}\) at 1 MHz) and approximately the same density (about
1.02 g cm\(^{-3}\)) of RTV-602. However, the RTV-615 has a higher longitudinal attenuation (about 1.3 dB cm\(^{-1}\) at 1 MHz) than the RTV-602 (0.5 dB cm\(^{-1}\) at 1 MHz). From the study of Madsen et al. (1983) on the RTV-615, the static shear velocity was found to be approximately 65 m s\(^{-1}\). Consequently, from the above data found in the literature, it was then estimated that the shear velocity of the RTV-602 is of the order of magnitude of 40 m s\(^{-1}\).

With these values of static longitudinal and shear velocity in the matrix, its bulk and shear moduli were calculated to be $B = 1.057 \times 10^9$ Pa and $G = 1.632 \times 10^6$ Pa, respectively.

The inclusions used in the construction of the samples were glass beads. From the informations given by the suppliers catalog, the beads were made from soda glass and lead glass. The longitudinal velocity of ultrasound measured in some specimens of soda glass varied from 5620 m s\(^{-1}\) to 5790 m s\(^{-1}\). The nearest value of longitudinal velocity found in the literature (Kaye & Laby, 1973) was that of crown glass, i.e., $c = 5660$ m s\(^{-1}\). The shear velocity in crown glass is $c = 3420$ m s\(^{-1}\). These velocities were taken as reference because of the intermediate density of crown glass (2.6 g cm\(^{-3}\)) between those of the glass beads (see section 3.4). The bulk and shear moduli of the glass beads were subsequently calculated and are given in table B.1. The group number in table B.1 refers to the size distribution and density of the glass beads as conventioned in chapter 3.
TABLE B.1
Elastic constants of the glass beads.

<table>
<thead>
<tr>
<th>group No.</th>
<th>bulk modulus $10^6$ Pa</th>
<th>shear modulus $10^6$ Pa</th>
</tr>
</thead>
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<td>II</td>
<td>4.79</td>
<td>3.41</td>
</tr>
<tr>
<td>III</td>
<td>4.76</td>
<td>3.39</td>
</tr>
<tr>
<td>IV</td>
<td>4.75</td>
<td>3.38</td>
</tr>
<tr>
<td>V</td>
<td>4.06</td>
<td>2.88</td>
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</table>

Various values of velocity and absorption in glass were tested in the theoretical model of Waterman & Truell (1961) discussed in chapter 2, and no significant difference in the results of phase velocity and attenuation in the samples were noticed. The same test was performed for the shear velocity and shear absorption in the matrix, and changes started to be noticed only for $c_s > 100$ m s$^{-1}$ and $\alpha_s < 26$ dB cm$^{-1}$ (at 1 MHz), respectively. From the information obtained from the literature these values were not considered realistic for this kind of silicone rubber.

With the above parameters on the silicone rubber RTV-602 matrix and on the glass beads inclusion, the bulk and shear moduli, and corresponding velocities, were determined for all samples under investigation. The calculation was made for the measured volume concentration of glass beads embedded in the silicone rubber matrix (column 4 of table 3.2.b). With the inclusion of glass beads in silicone rubber, the moduli are increased. As the density is at the same time increased, at a greater rate than the moduli, there results a decrease in velocity. The relevant data are presented in table B.2.
### TABLE B.2 **

Elastic constants and static velocities of samples.

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<tr>
<th>group</th>
<th>samp No.</th>
<th>bulk modulus (x10^9 Pa)</th>
<th>shear modulus (x10^6 Pa)</th>
<th>static long. velocity (m s^-1)</th>
<th>static shear vel. (m s^-1)</th>
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<td>1.615</td>
<td>1019.0</td>
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* from estimated effective concentration (see table 3.2.b and 3.2.c).
** The figures were approximated after calculations.
APPENDIX C

$\kappa$ VALUES AT CERTAIN FREQUENCIES
APPENDIX C

ka VALUES AT CERTAIN FREQUENCIES

With the presence of velocity dispersion in the silicone rubber RTV-602 matrix, the propagation parameter $ka$ is not linear with frequency, where $k = 2\pi / \lambda$ and $a$ is the mean radius of the scatterer. Nevertheless, for purpose of reference, the approximate values of $ka$ at certain frequencies are given in table C.1, for each group of mean bead size. The Roman figures in table C.1 indicate the group number.

<table>
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<th>$f$ (MHz)</th>
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<th>$ka_{IV}$</th>
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REFERENCES


