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THE EFFECTS OF SURFACE LAYERS ON THE ULTRASONIC SCATTERING FROM SPHERES

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1.INTRODUCTION

The applications of ultrasound in inhomogeneous media are becoming increasingly widespread, with measurements of attenuation and scattering proving of value in a number of different contexts, such as food systems[1,2],biomedical systems[3,4],and sewage[5].The purposes of the investigations range from extending our fundamental understanding[5], to applied engineering problems such as process control[6].

It is with engineering applications in mind that this work has been carried out. Instead of using the usual approach of analysing the structure of the spectra of a particular system with the aim of identifying the mechanisms contributing to the structure, it was decided to gain an idea of the general effect of the physical parameters of the different media in the system on the backscattered compressional wave (this being the most readily measured parameter).

Previous work has been carried out on the backscattering of many different materials for spheres immersed in water[7]. Subsequently the attenuation was examined for a variety of solid and liquid spherical inclusions suspended in liquids[8]. Here the study is extended by the addition of a boundary (shell) layer around the inclusion. Two extreme cases are examined, the first with a thin gaseous (air) shell, and the second with its physical properties only slightly different from the media inside and outside the shell.

2.ANALYSIS AND PARAMETERS

The model used to calculate the backscattering was developed by Anson and Chivers[9]. This is a shell model permitting the inclusion of viscoelastic effects and thermal waves in the calculations. The basic equations used in the model were those developed by Hay and Burling[10]. For the calculations the model needs several parameters: the longitudinal and shear wave speeds with their corresponding attenuations are needed for the solid phases; whilst for liquids the shear wave is set to zero with viscosity replacing the shear wave attenuation. In addition the density , thermal conductivity, specific heat at constant pressure, and coefficient of thermal expansion are needed for both liquid and solid phases.

All calculations were carried out as a function of acoustic radius, ka, this being determined from the wave number of the matrix and the radius of the inner sphere. For lossless materials the analysis can be scaled entirely as a function of ka. For lossy materials it is necessary to identify the actual value of the radius, a, that is used. For the present calculations it was 20μm (for the innermost sphere.)

3.GROSS SHELL MODEL

This first set of calculations was directed at an attempt to determine the general effect of a thin gaseous shell separating various media whose acoustic properties differ greatly from those of the shell. The shell properties used are those listed for air in reference [8].

Three types of systems were analysed as indicated in the following subsections.

3.1 SOLID/AIR/SOLID

The specific systems investigated were: Cu/Air/Cu, Cu/Air/Glass, Al/Air/Glass, Mg/Air/Glass, Polyethylene/Air/Glass, and Mg/Air/Al In addition the backscattering was calculated with no air gap and, for air spheres in place of the shell and inner sphere, for comparison with the shell calculations.

The spectra were calculated for the range 0<ka<6 with steps of 0.05, the thickness of the air layer was varied from 0.1nm to 10nm. Glass and Polyethylene were chosen for their lossy behaviour,and especially Polyethylene for its low acoustic impedance compared to the lossless metals considered. Emphasis was placed on the low ka range because of its practical importance and also because the higher ka ranges of the backscattering spectra have a complex shape due to overlapping resonances.

3.2 SOLID/AIR/LIQUID.

Polyethylene/Air/Pentane, Polyethylene/Air/Diodomethane, Al/Air/Pentane, Al/Air/Diiodomethane were the four systems studied with the same conditions as were used for solid/air/solid. The materials chosen represented a low density lossy solid (polyethylene), a lossless metal (Al), a low density liquid (pentane) and a high density one (diiodomethane), and were felt to give a reasonably wide coverage of possible systems

3.3 LIQUID/AIR/SOLID

The same materials as were used in section 3.2 were used for this type of system, the main change being the exchange of positions of the liquid and solid.

4.RESULTS AND DISCUSSION.

4.1 SOLID/AIR/SOLID

At the lower limit of the layer thickness used (0.1nm) the spectra for the metal/air/glass [fig.1] systems do not approach those expected for any of the metal/glass systems [fig.2]. This indicates that the introduction of even a very thin gaseous layer has a very significant effect on the system i.e the boundary conditions at the outer radius of the shells are similar to that of a free surface.

As the air gap is increased to 10nm the spectra in all cases (including the system with ployethylene) approach those for the relevant solid/air systems, an example of which is shown in Figure.3. The only case where any resemblance was found between the spectrum with a 0.1nm layer and that for the relevant solid/solid system, was for polyethylene/air/glass.

It was noted that for the Cu/Air/Glass and the Cu/Air/Cu systems, the backscattered spectra were nearly identical. This shows that the outer boundary has the most significant effect on the backscattering. This is hardly surprising given the sharp contrast in acoustical properties between a lossless metal and a lossy gas!

4.2 SOLID/AIR/LIQUID

4.2.1 Al/Air/Liquid; For an Aluminium matrix the backscattering is largely the same as that calculated for an air sphere of 20μm radius for air layers greater than 1nm in thickness [Figure 4]. Even at an air gap of 0.1nm [Figure.4 curve A] the sharp resonant structure shows a strong overlying similarity to that for an Al/air system.

The effect of changing the liquid in the core medium is not particularly large - the difference in properties between Pentane and Diiodomethane has a much smaller effect on the backscattering compared with the primary effect of a thin air shell. It is only at a thickness of 0.1nm that the resonances of the liquid core medium begin to show a significant effect. However the spectra for both Al/Pentane [Figure.5] and Al/Diiodomethane are very different to those for the spectra obtained with a 0.1nm air layer included (see for example curve A of fig.4).

4.2.2 Polyethylene/Air/Liquid; The results for this system are very different from those with Aluminium as the matrix medium [c.f. Figure 6 and Figure 4].For Polyethylene/Air/Diiodomethane, with a 0.1nm thick air layer, the spectrum is similar to that for a Polyethylene/Diiodomethane system [Figure 7]. There are two main groups of resonances centred around $1.0<ka<2.5$ and $3.5<ka<5.0$. The effect of the air layer [Figure 7 curve A] is to make these resonance peaks much sharper, and larger in magnitude. With a 10nm thick boundary layer, the spectrum is essentially that for an air sphere in a Polyethylene matrix, revealing a surprisingly rapid change from the two extreme cases of solid/liquid to solid/air. With a 1nm layer the spectrum shows features of the other two, tending to the solid/air system for higher ka values.

Polyethylene/Air/Pentane shows the same general features as the Polyethylene/Air/Pentane system, i.e the spectrum changes rapidly from the solid/liquid curve to a solid/air spectrum as the shell thickness is changed from 0.1nm to 10nm.

4.3 LIQUID/AIR/SOLID

In all the four cases studied, with a 0.1nm layer, the backscattered curve is approximately that for the relevant liquid/solid interface with only small shifts in the positions and sizes of the resonances.

However, when the layer is set at 10nm, the spectrum has the approximate shape of the curves expected for liquid/air systems. Above ka=1.5 the curve is flat and has a value approximately equal to one [curve C in Fig.8]. Whereas the liquid/air spectra continue to rise sharply as ka decreases (the lowest limit of ka was 0.05), in the system with the boundary layer included, there is a sharp double resonance in the region of ka=0.5 dropping rapidly to near zero below this. Presumably the height, width and structure of this feature depend on the properties of the matrix and the core medium.

When the thickness of the boundary layer is set to 1nm, the backscattering shows features in common with both spectra. This was most clearly shown for the Diiodomethane/Air/Aluminium system [Figure 8 curve B].

4.4 SUMMARY

a) The outer boundary in general has the most significant effect on the shape of the backscattered spectra. The inner boundary only has a significant effect when the boundary layer is extremely thin (of the order of 0.1nm), or the difference in properties across the outer boundary are not too great compared to those of the inner boundary, e.g. Pentane/Air/Aluminium. In these cases for a shell of 0.1nm, it is the inner boundary which has the most significant effect on the scattering.

b) In general above a thickness of 10nm the air shell looks like an air sphere with no core at all.

5. SUBTLE SHELL MODEL.

Using the same choice of a sphere of 20μm radius, the backscattering was calculated for a liquid/liquid/liquid system. The properties of the boundary layer were not greatly different from those of the matrix or of the inclusion. The intention was to determine the thickness of the shell for which the shell has a significant effect on the backscattering.

Three different systems were studied;Water/Anilene/Water, Water/Morpholine/Water and Hexanol/Morpholine/Hexanol. The second and third systems were chosen to examine the effect of having similar acoustic impedances for the media, but different thermal properties, (Water/Morpholine/Water), and the effect of similar thermal properties but different acoustic impedances.

As in the "gross shell" model the backscattering was calculated over the range 0<ka<6 with various thicknesses for the boundary layer, between 0.1nm and 30μm (with the internal sphere's radius still kept constant at 20μm).

For all three systems, layer thicknesses less than 1μm produce backscattering curves which exhibit the same form. This is not particularly remarkable since the properties of all the liquids used are similar. The differences only really become noticeable when the shell has the thickness of 10μm, i.e when it is of the same order of magnitude as the radius of the internal sphere.

It was noted that for shells whose thickness was greater than 1μm there was a shift in the position of the resonances to higher values. This was quite marked for 10μm shell. Below 0.1μm shell thickness this shift is negligible, if indeed it exists at all. As has been stated above, the curves obtained for shells of 0.1μm thickness or less were very similar, differing only in the values of the magnitudes of the resonance peaks. It seemed reasonable to believe that this was indeed the same curve with just a different scaling factor related to the thickness of the shell. Taking the Hexanol/Morpholine/Hexanol system a scaling factor was used based on the ratio of the size of the second maximum for pairs of curves

This re-scaling was carried out for the curves obtained for the Hexanol/Morpholine/Hexanol system and good agreement was found between all the curves for different shell thicknesses until the re-scaled curve for a 0.1μm layer was superimposed on that for the 1μm layer. Here there were slight differences in the positions of the resonance peaks although the heights agreed quite well. The upper limit at which the shell thickness causes shifts in the positions of the resonances is around 0.1μm. A graph of the height of the second resonance (this being a convenient arbitrary point) was plotted as a function of shell thickness, for both Water/Anilene/Water and Hexanol/Morpholine/Hexanol systems on a log-log scale. As expected a straight line was obtained.

For larger values of shell thickness a scaling factor was applied to the ka values, this was defined as the ratio of the positions of the second minimum, (this being a well defined arbitrary feature.) The graph for the Hexanol/Morpholine/Hexanol system with a 1μm shell was re-scaled and overlaid on that for a shell of 10μm thickness. The two curves coincide up to about ka=5, but diverge in both terms of the positions and the heights of the resonances. As a generalisation, the shell affects the size and to a lesser extent the positions of features in the backscattered spectra, but does not appear to alter the basic form of the scattering. This generalisation is only valid up to a certain limit, which in this particular case is around a shell thickness of approximately 10μm.

As further evidence, a graph was plotted of the position of the second minimum against shell thickness. The curve obtained approaches a limiting value of ka approximately equal to 4.41, when the shell thickness reaches approximately 30μm. If the backscatter calculated for a 50μm Morpholine sphere in a Hexanol matrix is compared with a 30μm Morpholine shell round a 20μm Hexanol sphere, it is apparent that the two curves are very similar. The major difference is that the the shell system has a larger second resonance. This shows that the limiting value of the ka of the second minimum as a function of shell thickness identifies the region where the shell system appears to the acoustic wave to be effectively a sphere of the shell material.

It was also of interest to discover whether the backscattering depended on the absolute thickness of the shell, or only on the relative thickness with respect to the size of the inner sphere. The calculation was run with the internal radius set to 200μm (c.f. 20μm) and shell thickness of 0.01μm (c.f. 1nm) and 100μm (c.f. 10μm). When these curves were overlaid with those calculated for the 20μm sphere-based system there were no obvious differences. This suggests that the backscattering is determined by the ratio of the shell to sphere radius.

We summarise the results obtained for the "subtle shell" model;
a) The shell becomes significant in its effect when its thickness is of the same order of magnitude as the size of the core medium.

b) The backscattered spectra, as a function of ka, for various shell thicknesses, are basically the same curve with different scaling factors on the axes. These scaling factors are related primarily to the shell thickness.

c) There is a limiting (maximum) value of ka for the position of the second minimum as a function of the shell thickness. For a 20μm sphere in the Hexanol/Morpholine/Hexanol system this value is approximately 4.41 and is reached at a shell thickness of 30μm.

d) At a shell thickness of 30μm the system is showing features characteristic of a 30μm radius Morpholine sphere suspended in a Hexanol matrix. This suggests that a changeover point has been reached where the system "looks" like a sphere rather than a shell.

e) The scattering curve depends on the relative size of the shell and the internal sphere, not just on the absolute value of the shell thickness.

6. CONCLUSION.

Systematic investigations are needed to validate some of the generalisations suggested by the specific studies reported here, which nevertheless find application in a wide variety of diagnostic applications.

7. REFERENCES.

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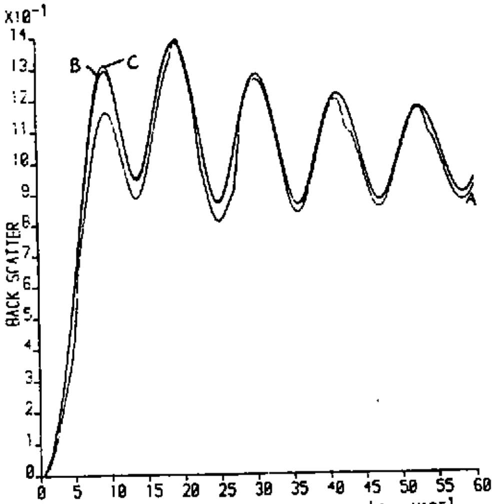

Fig1. Cu/Air/Glass with different shell thicknesses. A:0.1nm; B:1nm; C:10 nm

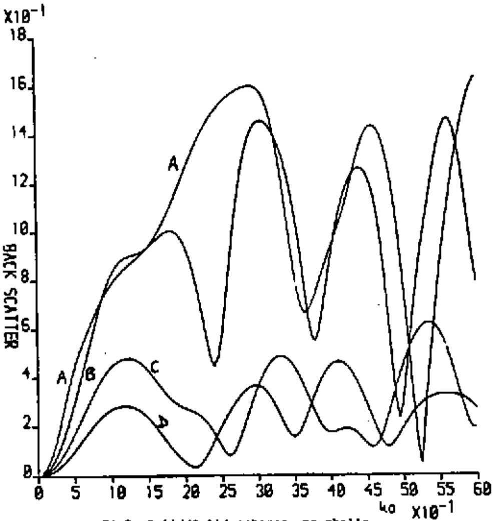

Fig2. Solid/Solid spheres, no shells.
A: Polyethylene/Glass; B: Cu/Glass
C: Mg/Al; D:Mg/Glass.

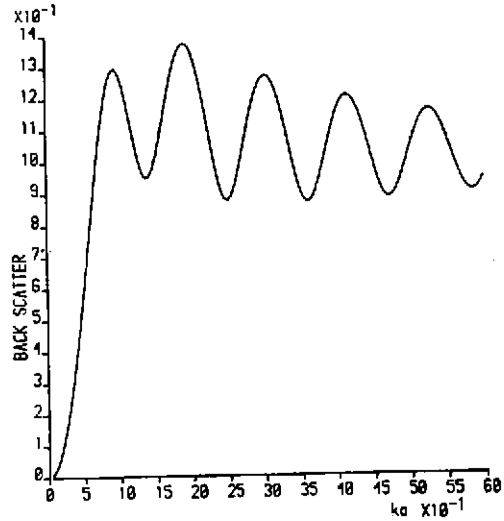

Fig3. Gaseous sphere in solid.
(Co matrix, air sphere).

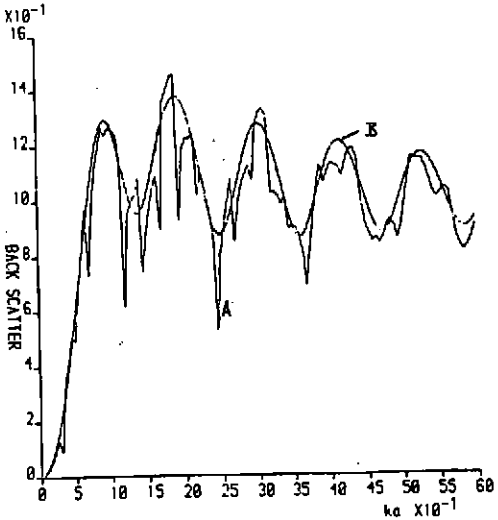

Fig4. Al/Air/Pentane for different shell thicknesses. A:0.1nm; B:10nm

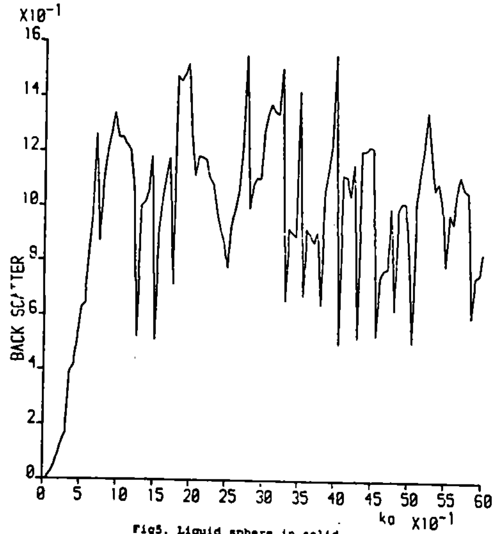

Fig5. Liquid sphere in solid.
(Al matrix, Pentane sphere)

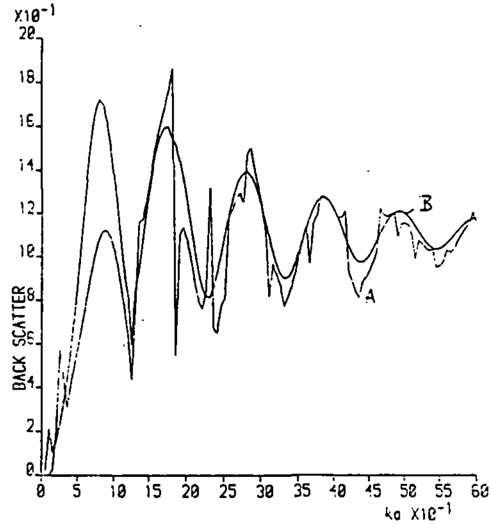

Fig6. Polyethylene/Air/Diiodomethane
for different shell thicknesses.
A:0.1nm; B: 10nm

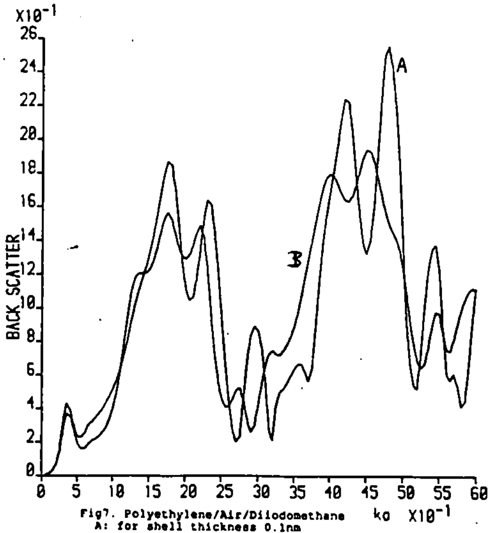

Fig7. Polyethylene/Air/Diiodomethane
A: for shell thickness 0.1nm
B: with no shell

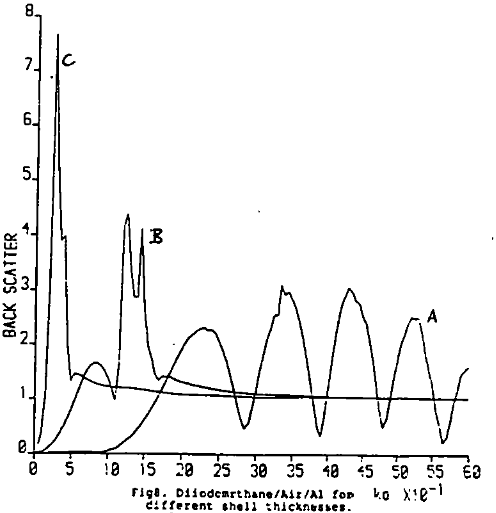

Fig8. Diiodcmrthane/Air/Al for
different shell thicknesses.
A: 0.1nm; B: 1nm; C: 10nm.