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(54) Title: DETERMINING THE SIZE DISTRIBUTION OF PARTICLES IN A FLUID

(57) Abstract

In this embodiment the size distribution of particles in a fluid is determined by performing the following steps: (a) passing a plurality of ultrasonic beams through the fluid, wherein the beams have respective frequencies (f_1, f_2,...f_n); (b) obtaining a velocity measure of the beams in the fluid; (c) obtaining an ultrasonic velocity spectrum as a function of particle size for the fluid; (d) determining from the attenuation of gamma-rays the density of the fluid; and (e) calculating the particle size distribution for the fluid from the velocity measure, the velocity spectrum, and the suspension density which were respectively obtained in steps (b), (c) and (d).

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TITLE: DETERMINING THE SIZE DISTRIBUTION OF PARTICLES IN A FLUID

The present invention relates to a method and apparatus for

determining the size distribution of particles in a fluid.

5   The term fluid is intended to include both a liquid and the solid particles

contained in that liquid. Examples include slurries and other suspensions.

The invention has been developed primarily for use with opaque slurries in the

mineral and paint industries and will be described hereinafter with reference to that

application. However, the invention is not limited to these particular fields of use and

10   can also be used to determine size distribution of particles in other suspensions and in

other industries such as the chemical, pigment, pharmaceutical, food and cosmetic

industries, amongst others. The invention is particularly applicable to on-line particle

size analysis for the control of grinding, crystallisation, precipitation and

agglomeration processes.

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It is preferable to use on-line particle size analysers to negate the need for side stream or dilution of the fluid being analysed. Hitherto, such analysers have made use of either the attenuation experienced by one or more ultrasonic beams directed through the fluid, laser scanning or laser diffraction.

5   It is an object of the present invention, at least in the preferred embodiment, to provide a method and apparatus for determining the size distribution of particles in a fluid using a velocity measure of a plurality of ultrasonic beams at different frequencies in said fluid.

According to one aspect of the invention there is provided a method for

10   determining the size distribution of particles in a fluid, the method including the steps of:

(a)   passing a plurality of ultrasonic beams through the fluid, wherein the beams have respective frequencies f_1, f_2,.....f_n;

(b)   obtaining a velocity measure of the beams in the fluid;

15   (c)   obtaining an ultrasonic velocity spectrum as a function of particle size for the fluid; and

(d)   calculating the particle size distribution for the particles in the fluid from the velocity measure and the velocity spectrum which were respectively obtained in steps (b) and (c).

20   Preferably, the ultrasonic velocity spectrum is obtained either by prior experiment or calculation.

Preferably, step (c) includes transforming the velocity spectrum into a linear form. More preferably, step (d) includes an inversion of the linear form.

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Preferably also, the method includes the steps of:

(e)   passing gamma rays through the fluid;

(f)   determining from the attenuation of the gamma-rays the density of the fluid; and

5   (g)   calculating the particle size distribution of the particles in the fluid from the velocity measure, the velocity spectrum, and the density measure respectively obtained during steps (b), (c) and (f).

Preferably step (g) includes an inversion of the linear form to give the particle size distribution in a certain size range. In an alternate embodiment, the amount of

10   particles outside the inversion size range is also calculated.

Preferably also, step (b) includes determining the difference in velocity of the beams having frequencies f_i and f_{i+1}. In alternative embodiments, however, step (b) includes determining the velocity of each of the beams.

According to another aspect of the invention there is provided an apparatus for

15   determining the size distribution of particles in a fluid, the apparatus including:

transducer means for passing a plurality of ultrasonic beams through the fluid, wherein the beams have respective frequencies $f_1, f_2....f_n$;

first processor means for obtaining a velocity measure of said beams in the fluid;

20   second processor means for obtaining the ultrasonic velocity spectrum as a function of particle size for the fluid; and

third processor means responsive to the first and second processor means for calculating the particle size distribution for the fluid.

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Preferably, the first, second and third processors are incorporated in a single processor.

A preferred embodiment of the invention will now be described, by way of example only, with reference to the accompanying drawings in which:

5   Figure 1 is a graph illustrating the variation of velocity of an ultrasonic beam through a fluid containing particles of various sizes as calculated using a theoretical model;

Figure 2 is an apparatus according to the present invention as used during testing;

10   Figure 3 is the results of the inversion procedure for 1.0μm and 1.5μm diameter test silica suspensions using the apparatus and method according to the invention;

Figure 4 illustrates the measured velocity spectrum giving rise to the results in Figure 3;

15   Figure 5 illustrates the results for the 1.0μm diameter test silica sample with and without a density correction;

Figure 6 illustrates the particle size distribution for an oversize coarse SiC sample;

Figure 7 shows the measured and theoretical values of velocity as a function of

20   frequency for two TiO_2 suspensions; and

Figure 8 illustrates the particle size distribution obtained by the invention for two TiO_2 suspensions.

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In this embodiment the size distribution of particles in a fluid is determined by

performing the following steps:

        (a)   passing a plurality of ultrasonic beams through the fluid, wherein the

beams have respective frequencies $f_1, f_2, ...f_n$;

5   (b)   obtaining a velocity measure of the beams in the fluid;

        (c)   obtaining an ultrasonic velocity spectrum as a function of particle size

for the fluid;

        (d)   determining from the attenuation of gamma-rays the density of the

fluid; and

10   (e)   calculating the particle size distribution for the fluid from the velocity

measure, the velocity spectrum, and the suspension density which were respectively

obtained in steps (b), (c) and (d).

In this embodiment the velocity measure is the differences between velocities

at different frequencies. This is particularly advantageous because the changes in

15   velocity are small compared to the total velocity. Moreover, compared to total

velocity measurements, velocity differences are less sensitive to temperature changes

and air bubbles in the fluid.

For the fluids under consideration, the difference in velocity is best expressed

by the matrix equation:

20      $D = RM$ (1)

where $D=\{d_1, ...d_m\}$ whose elements d_m represent the measured velocity

differences between each pair of frequencies f_i and f_{i+1}. Additionally, $R=\{r_1, ...r_n\}$

whose elements r_n represent the volume fraction of particles in the fluid in each

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successive size range, while M is a matrix of calculated velocity differences whose

elements are given by:

$$m_{ij}=V(f_{j+1}, a_i,\rho)-V(f_j, a_i,\rho) \tag{2}$$

In equation 2, V is the velocity calculated in step (b) above, ρ is the total

volume fraction of solids as measured by a gamma-ray attenuation gauge, and a_i is the

radius of the ith size bin. In addition, there is the important constraint that:

$$\rho = \sum_{i=1}^{n} r_i \tag{3}$$

During step (d) the matrix equation is inverted to determine the values of

$r_1,....r_n$. As with most indirect methods of measurement, the results of direct inversion:

$$R = DM^{-1} \tag{4}$$

can be unstable and sensitive to small measurement errors. However,

satisfactory results are obtained with the use of inversion algorithms such as that

described by Twomey, S. in "Introduction to the mathematics of inversion in remote

sensing and indirect measurements", Developments in Geomathematics 3, Elsevier

Amsterdam 1977, the contents of which are incorporated herein by way of cross

reference. Twomey describes various techniques for inverting such equations without

introducing unwanted oscillations into the solution. An iteration technique due to

Chahine and described in Twomey was chosen for the present work, although

alternative techniques are available. Iteration is carried out until the RMS error

between the experimental and calculated results is similar to the measurement error.

The adopted technique has the advantages of automatically ensuring that all elements

of R are positive and allowing normalisation to ensure that equation 3 is satisfied after

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each iteration. The fraction of particles outside the size range accessible to the range

of frequencies used may also be inferred using the gamma-ray density measurement as

will be discussed in more detail below.

The range of particle sizes accessible to this method can be calculated using

5   various theoretical models such as those described by Harker, A. and Temple, J. in

"Velocity and attenuation of ultrasound in suspensions of particles in fluids" J. Physics

D: Applied Physics 21, 1576-88 and by McClements, D.J. and Povey, M.J.W., in

"Ultrasonic Velocity as a Probe of Emulsions and Suspensions", Advances in Colloid

and Interface Science 27, 285-316, the contents of both being incorporated herein by

10   way of cross reference. The theoretical model of Harker and Temple (1988) has been

used in this embodiment due to a combination of accuracy and ease of use.

Figure 1 shows the calculated ultrasonic velocity as a function of frequency for

suspensions of various particle sizes. To interpret the curves in Figure 1 note that the

velocity of ultrasound in slurries depends on the inertia of particles in the fluid under

15   the influence of the ultrasonic wave. An ultrasonic wave passing through a fluid

causes a periodic displacement of the fluid. At low frequencies very small particles

tend to move in phase with the fluid so the velocity in the slurry of the ultrasound may

differ considerably from the velocity in the pure suspending fluid. As particle size and

ultrasonic frequency increases, the particles tend to lag more and more behind the

20   movement of the fluid and the ultrasonic velocity approaches that of the suspension

acting as a uniform fluid. There is a transition frequency range between complete

entrainment and no entrainment of the particles. The centre frequency of this

transition region shows a significant decrease with increasing particle diameter.

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Figure 1 shows that, for silica in water, ultrasonic velocity measurements in

the frequency range 10 kHz to 50 MHz can be used to determine the particle size

distribution of particles in the range of about 0.1 to 20 microns. It is the range in the

frequency of ultrasound velocity measurements that restricts the range of the

5   calculated particle size distribution. It is feasible to extend the technique to coarser

particles by measuring at lower frequencies and to finer particles by using higher

frequencies.

Figure 2 shows a slurry measurement chamber 1 used in the testing of the

invention. The chamber 1 contains the suspension 2 to be measured, which is a

10   dispersion of unknown particle size and concentration. An agitator 3 (which may be a

stirrer) is employed to keep the dispersion suspended and homogenous. An additional

ultrasonic disperser may be used to keep the particles in suspension. In industrial

applications, chamber 1 may be replaced by a pipe through which the slurry of interest

is pumped.

15   This embodiment of the invention uses six pairs of piezoelectric transducers 4

to transmit and receive ultrasound of specific frequency. Alternatively, use can be

made of lesser number of broadband ultrasonic transducers. Transducers 4 are aligned

with the faces highly parallel and a Matec Instruments MBS 8000 was used to

generate pulsing signals and to receive and amplify the transmitted signals. The

20   transducers were excited in toneburst mode and the transit time of the pulse measured.

Calibration was performed by measuring the transit time of ultrasonic pulses in water

at different temperatures.

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A gamma-ray attenuation density gauge 5 is used to measure the solids weight fraction of the slurry. A collimated 370 MBq (10 mCi) [137] Cs gamma-ray source 6 is used and gamma-ray transmission detected with a 51x51mm NaI(T1) detector and photomultiplier 7. The density may be derived from the gamma-ray transmission, as

5   is known.

The experimental data is then inverted to give a particle size distribution. In some circumstances other inversion methods such as a correlation technique or a full non-linear inversion procedure are preferable.

To further illustrate the method according to the invention a specific example

10   of silica suspensions in water is given. More particularly, two standard suspensions of spherical silica particles of known particle size were measured, one with a mean diameter of 1.0 microns and the other with a mean diameter of 1.5 microns. The size distribution of these samples was supplied by the manufacturer and confirmed by measurements on a laser diffraction particle size analyser. The theoretical model of

15   Harker and Temple (1988) was used to provide a velocity spectrum. For volume fractions less than 15% by volume the model is approximately linear and so no perturbation scheme was necessary.

The results of the inversion procedure for the 1.0 micron and 1.5 micron diameter silica suspensions are shown in Figure 3. The method shows good

20   discrimination between the two suspensions. The volume density for both solutions was measured to be approximately 10%. For the nominal 1.0 micron sample the mean particle size was measured to be 0.94 micron whereas for the 1.5 micron distribution it was measured as 1.43 micron. These results represent good agreement between the

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present method and laser diffraction. The measured width of the size distributions

shown in Figure 3 is relatively large because of the experimental errors in velocity

determination and because of the small number of frequencies used.

The experimental data for the two suspensions is graphed in Figure 4 (dots for

5   the 1.0 micron sample and squares for the 1.5 micron sample) along with the velocities

calculated for the particle size distributions of Figure 3 (unbroken lines). The

inversion method has produced an accurate result for the particle size despite errors in

the individual measurements shown in Figure 4. This favourable result is achieved in

part due to the use of the density gauge which places a tight constraint on the

10   inversion. Without use of the density measurement the invention can still provide

useful results in some cases, as could be determined by those skilled in the art. Figure

5 shows the inversion for the 1.0 micron silica sample with and without the constraint

in place. Spurious high concentrations of fine particle size (less than 0.5 microns) are

generated in the unconstrained case while the RMS error of the fit to the ultrasonic

15   velocity data is improved. This illustrates the importance of the density constraint in

improving the resolution of the inversion technique. The choice of inversion method

is also important in producing a good inversion as it limits the degree of oscillation

present in the solution.

In the above example, the particle size of the silica corresponds to the central

20   part of the frequency range covered by the measurements. In order to evaluate the

method on a sample containing out of range particles, a coarse SiC sample with a

mean diameter of approximately 20 microns was used. Measurements were again

made at the same frequencies as for the silica. Figure 6 shows the results of inverting

that data. The program correctly assigns almost all the material to the coarse end of the particle size spectrum.

One application of the ultrasonic velocity technique is in the improved control of dispersion in the paint and pigment industry. Dispersion is aimed at breaking down

5 agglomerations of the 0.2 micron diameter TiO_2 particles. After dispersion the majority of particles are agglomerations of a few basic units. Two TiO_2 dispersions were provided by industry, one well and the other poorly dispersed. Measurement of these dispersions also provides a test of the invention in conditions where the particle size distribution is polydisperse unlike those in the test cases described above.

10 In the present work, measurements were made on the two dispersions at a solid content of 2.3% by volume. Figure 7 shows the measured and theoretical values of velocity as a function of frequency. The results show discrimination of the two samples. In the present case, high accuracy velocity measurements were made over the frequency range 0.25 to 3.5 MHz. Therefore, the present measurements are not

15 very sensitive to particles of diameter less than about 0.3 microns or greater than about 10 microns.

Application of an inversion algorithm to the data shown in Figure 7 yields the particle size distributions shown in Figure 8. The d50 for particles finer than 2.0 microns is 0.38 microns for the well dispersed sample and 0.53 microns for the poorly

20 dispersed sample. The fraction of particles greater than 2.0 microns estimated by the present method and is 0.33 for the poor sample and 0.16 for the good sample.

Measurements were made of both dispersions by optical diffraction techniques. The dispersions had to be diluted to 0.004% to allow sufficient optical transmission.

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Stirring was kept to a minimum in the measurement and no ultrasonic dispersion was used to avoid breaking up the agglomerates. The d50 measured by the optical method was 0.36 micron for the poor and 0.35 micron for the good dispersions. The optical and ultrasonic techniques display good agreement for the well dispersed sample not

5 for the poorly dispersed sample. This can be explained, however, by the extreme dilution which causes the agglomerates in the poorly dispersed sample to a break up. In any event this illustrates the disadvantage of prior art methods such as optical diffraction techniques.

Alternatively, information about the particle size distribution can be presented

10 in a simpler fashion. For the above TiO_2 example, a parameter R may be defined as

$$R=(V_{1.0}-V_{.5})/(V_{3.5}-V_{2.25}) \tag{5}$$

where V_x is the ultrasonic velocity at x MHz. As particle size changes, the curve in Figure 1 moves either to the left or right changing the ratio R quite strongly. This ratio is insensitive to temperature and concentration fluctuations and therefore

15 may be used as a figure of merit for dispersion. The value of R was measured as 1.40 ± 0.05 for the poor dispersion and 0.90 ± 0.05 for the good dispersion. Such a single parameter provides a simple criteria for the adequacy of the dispersion process.

Although the invention has been described with reference to a particular example, it will be appreciated by those skilled in the art that it may be embodied in

20 many other forms.

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CLAIMS:-

1.   A method for determining the size distribution of particles in a fluid, the

method including the steps of:

        (a)   passing a plurality of ultrasonic beams through the fluid, wherein the

5   beams have respective frequencies $f_1, f_2, \dots f_n$;

        (b)   obtaining a velocity measure of the beams in the fluid;

        (c)   obtaining an ultrasonic velocity spectrum as a function of particle size

for the fluid; and

        (d)   calculating the particle size distribution for the particles in the fluid.

10   from the velocity measure and the velocity spectrum which were respectively obtained

in steps (b) and (c).

2.   A method according to claim 1 wherein the ultrasonic velocity spectrum is

obtained either by prior experiment or calculation.

3.   A method according to claim 1 wherein step (c) includes transforming the

15   velocity spectrum into a linear form.

4.   A method according to claim 3 wherein step (d) includes an inversion of the

linear form.

5.   A method according to claim 1 including the steps of:

        (e)   passing gamma rays through the fluid;

20   (f)   determining from the attenuation of the gamma-rays the density of the

fluid; and

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(g)   calculating the particle size distribution of the particles in the fluid

from the velocity measure, the velocity spectrum, and the density measure

respectively obtained during steps (b), (c) and (f).

6.   A method according to claim 5 wherein step (g) includes an inversion of the

linear form to give the particle size distribution in a certain size range.

7.   A method according to claim 6 wherein the amount of particles outside the

inversion size range is also calculated.

8.   A method according to any one of the preceding claims wherein step (b)

includes determining the difference in velocity of the beams having frequencies f_i and

f_{i+1}.

9.   A method according to claim 1 wherein step (b) includes determining the

velocity of each of the beams.

10.   An apparatus for determining the size distribution of particles in a fluid, the

apparatus including:

transducer means for passing a plurality of ultrasonic beams through the fluid,

wherein the beams have respective frequencies $f_1, f_2....f_n$;

first processor means for obtaining a velocity measure of said beams in the

fluid;

second processor means for obtaining the ultrasonic velocity spectrum as a

function of particle size for the fluid; and

third processor means responsive to the first and second processor means for

calculating the particle size distribution for the fluid.

11.   An apparatus according to claim 10 wherein the first, second and third

processors are incorporated in a single processor.

Figure 1
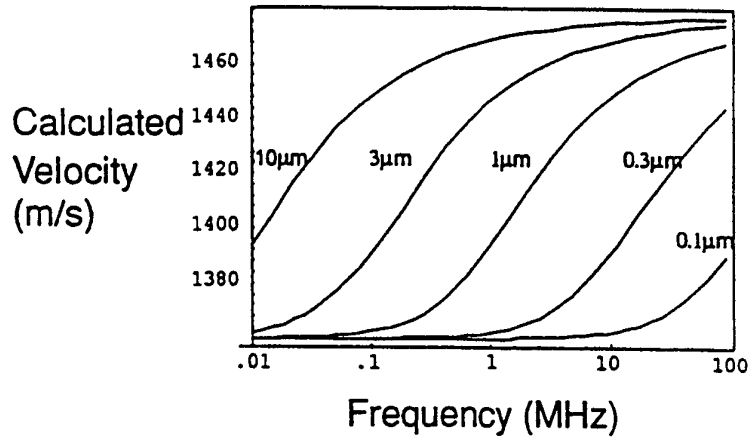
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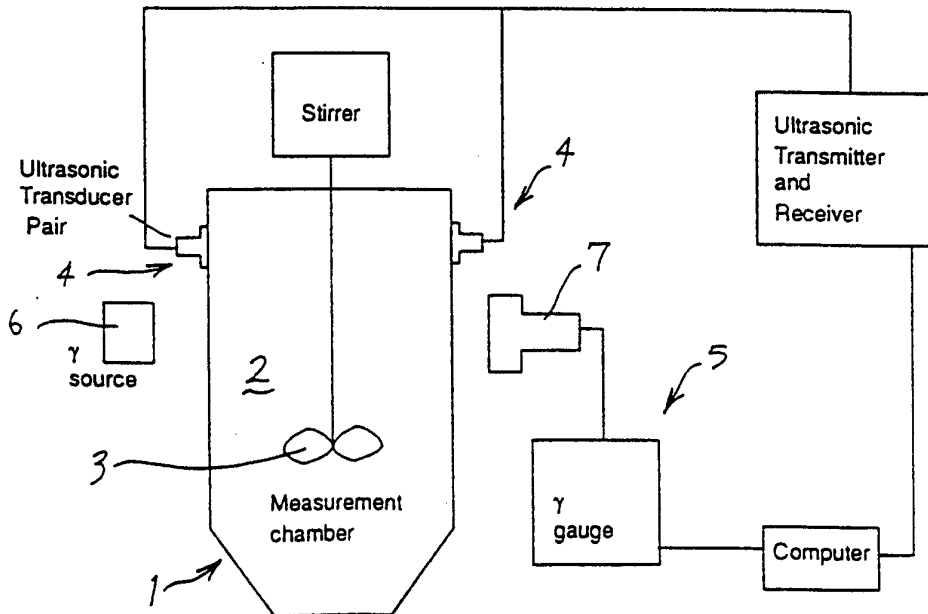
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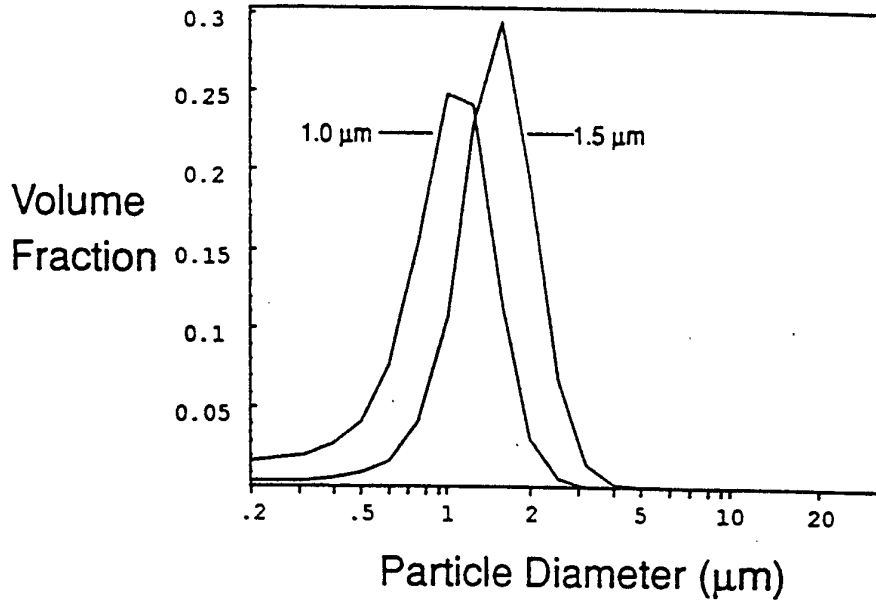
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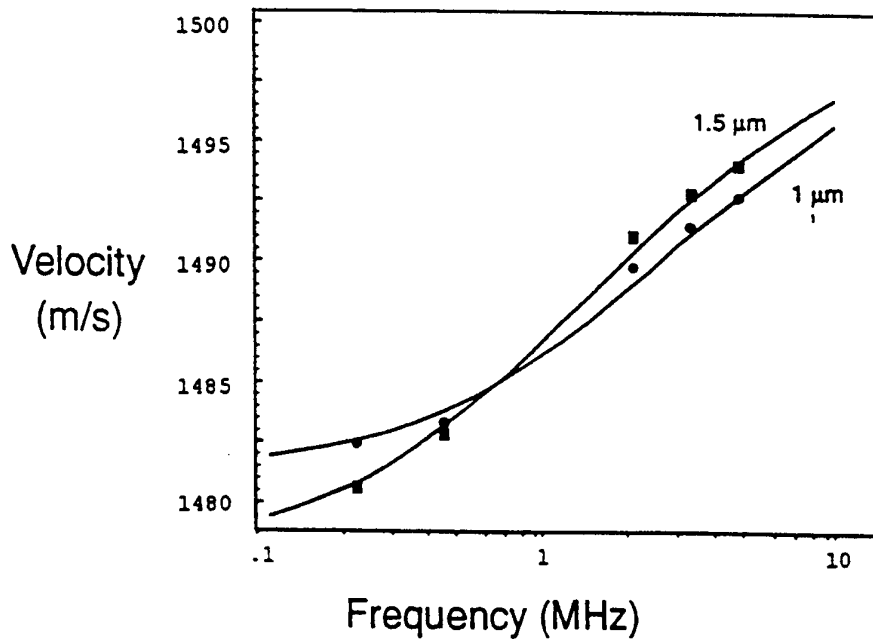
Figure 5
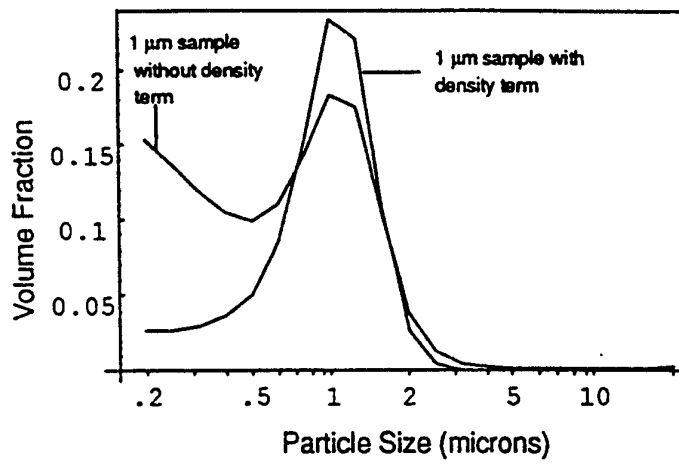
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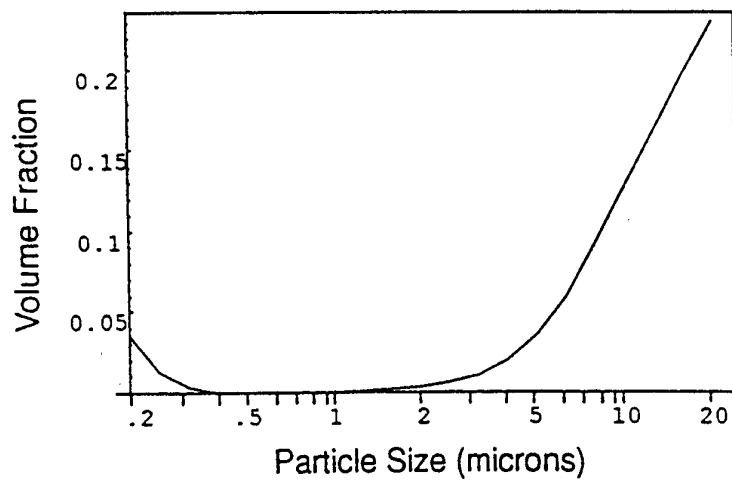
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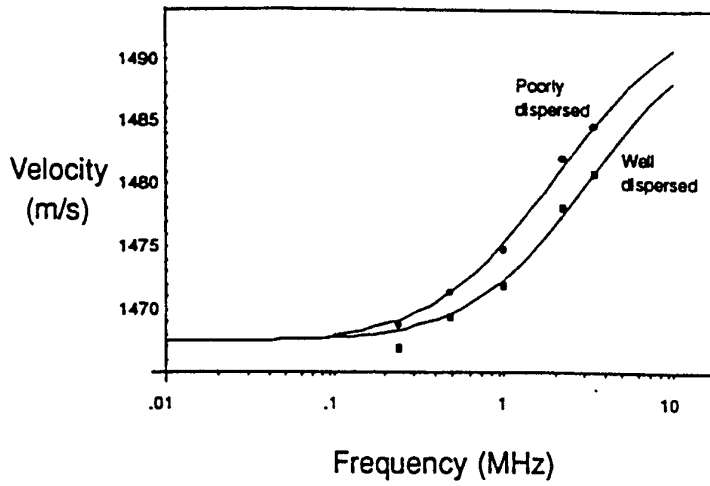
Figure 8
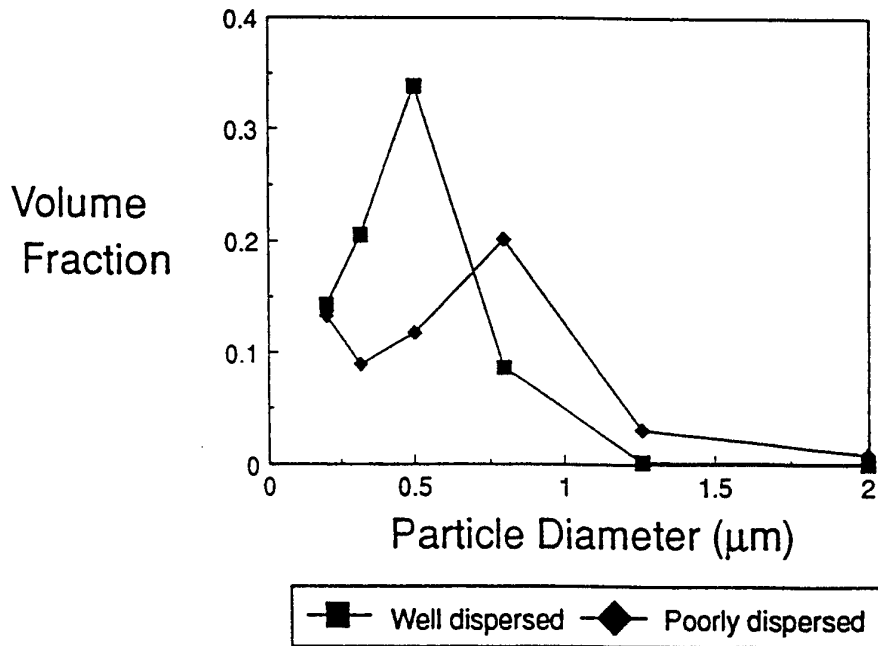
A.   CLASSIFICATION OF SUBJECT MATTER

Int Cl[6:]   G01N 15/02, 29/02, 29/18

According to International Patent Classification (IPC) or to both national classification and IPC

B.   FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC : G01N 15/02, 15/10, 23/12, 29/02, 29/18

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
AU : IPC as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
DERWENT, JAPIO

C.   DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 44596 A (INSTITUTET FOR VATTEN-OCH LUFT) 27 January 1982. See page 6 line 27 to page 7 line 4	1, 8-10
A	US 4283953 A (PLONA) 18 August 1981. See Abstract	1, 10
A	US 5121629 A (ALBA) 16 June 1992. See Abstract	1-10

[X] Further documents are listed in the continuation of Box C   [X] See patent family annex

*	Special categories of cited documents:	"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A"	document defining the general state of the art which is not considered to be of particular relevance		
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"O"	document referring to an oral disclosure, use, exhibition or other means		
"P"	document published prior to the international filing date but later than the priority date claimed	"&"	document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
12 July 1996	**23 JUL 1996**

| Name and mailing address of the ISA/AU AUSTRALIAN INDUSTRIAL PROPERTY ORGANISATION PO BOX 200 WODEN ACT 2606 AUSTRALIA   Facsimile No.: (06) 285 3929 | Authorized officer **STEPHEN CLARK** Telephone No.: (06) 283 2164 |

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Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
	C (Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT	
A	GB 1485750 A (NATIONAL RESEARCH DEVELOPMENT CORPORATION) 14 September 1977. See lines 41 to 73	1, 10
A	Patent Abstracts of Japan, P-413, page 106 JP 60-147607 A (HITACHI SEISAKUSHO) 3 August 1985. See Abstract	1, 10
A	Patent Abstracts of Japan, C-1195, page 8, JP 6-22956 A (YOKOGAWA MEDICAL SYST) 1 February 1994. See Abstract	1, 10
A	US 3529153 A (ZIMMERMAN) 15 September 1970. See Abstract	5

This Annex lists the known "A" publication level patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent Document Cited in Search Report		Patent Family Member						
EP	44596	JP	57086026	SE	8005251			
US	4283953	AU	52183/79	BR	7907044	CA	1150814	
		DE	2943304	EG	14417	FR	2440561	
		GB	2034888	IT	1124839	MY	179/85	
		NL	7907836	NO	793458	PH	22913	
US	5121629	EP	605409	WO	9107646			

END OF ANNEX