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Modified log-normal particle size distribution in  
acoustic spectroscopy

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## Modified log-normal particle size distribution in acoustic spectroscopy

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### Abstract

The modified log-normal distribution is a smooth continuous distribution depending on four parameters: median size, standard deviation, minimum size and maximum size. This distribution introduced by Kottler [J. Franklin Inst. 250 (1950) 339] and Irani [J. Phys. Chem. 63 (1959) 1603] gives an opportunity to restrict the particle size range. This distribution is asymmetrical on the logarithmic scale of particle sizes.

The modified log-normal distribution is especially useful for acoustic spectroscopy because sound attenuation is very sensitive to the presence of large particles. Particles with size above 3  $\mu\text{m}$  cause significant scattering losses. As a result, sound attenuation spectra show a sharp growth within a sound frequency range 10–100 MHz when large particles are present.

The high sensitivity to large particles is a very useful feature of acoustic spectroscopy allowing one to detect these particles. This feature requires a special approach allowing the characterization of large particles with a high accuracy. Regular log-normal distribution tends to overestimate the fraction of large particles and underestimate the median size.

Experimental tests have been performed with silicon particles dispersed in water. The particle size distribution has been measured independently by X-ray sedimentation and by an acoustic spectrometer. This test shows that the log-normal distribution provided by the acoustic spectrometer deviates from the sedimentation data. Implementation of the modified log-normal distribution resolves this discrepancy. © 1998 Elsevier Science B.V.

### 1. Introduction

The biggest advantage with respect to acoustic spectroscopy, relative to other spectroscopic methods, is the ability to characterize intact concentrated dispersed systems. The frequency dependence of the sound attenuation is the normal experimental output of this measurement. This frequency spectrum can then be converted to the particle size distribution.

This conversion procedure requires a theory for sound propagation through the dispersed system.

The theory for the dilute case has been created by Epstein and Carhart [1] and Allegra and Hawley [2] (“ECAH” theory). The validity of the theory is restricted because of neglect of particle–particle interaction.

A theory for the concentrated case is available for small particles with a high density contrast when viscous losses are the dominant mechanism of the sound attenuation [3]. There are also several versions of the multiple scattering theory which are important for the concentrated system containing large particles [4]. The Waterman–Truell version [5] is the best known.

These theoretical models make it possible to

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calculate attenuation spectra if the particle size distribution is known. The goal of acoustic spectroscopy is the reverse: calculation of the particle size distribution when attenuation spectra are known. This is a so-called "ill-posed problem" which occurs because of possible multiple solutions. There are two general approaches to resolving problems like these.

Approach 1 is the simpler and older one. The distribution of the predefined shape is used in the optimization procedure. The goal of the optimization procedure is to determine the parameters of the distribution which give the best fit for the experimental spectra. Log-normal distributions are typically tested.

Approach 2 applies no restrictions on the shape of the distribution. The sophisticated mathematical procedure allows calculation of the particle size distribution from the integral equation. A superposition assumption justifies the use of a certain integral equation, usually a Fredholm equation of the first kind [6,7]. This approach seems superior because it aims to yield the full "real" distribution.

The basic difference between these two approaches is the number of adjustable parameters used to fit experimental spectra. Approach 1 requires only two adjustable parameters (median size and standard deviation) when a log-normal distribution is tested. The number of adjustable parameters required for Approach 2 is much larger. These presentative full particle size distributions usually contain about 20 fractions. The weight of each fraction is an unknown adjustable parameter.

Acoustic spectra do not provide enough information to determine uniquely all of the adjustable parameters required by Approach 2. Therefore, Approach 2 is not viable for acoustic spectroscopy and other techniques based on the macroscopical measurements of the intact dispersed system. As a consequence, the modern electroacoustic spectrometer [8,9] utilizes Approach 1.

Approach 1 yields the approximate particle size distribution. This approximation is adequate when the predefined distribution shape is correct. However, the shape of the real particle size distribution often deviates from the log-normal; the

distribution might be truncated intentionally or naturally, depending on the processing history of the material. The predefined shape must then be modified in order to improve performance of the characterization.

The "modified" log-normal distribution suggested by Kottler [10] and Irani [11] is a good candidate for a distribution with asymmetric shape. The number of adjustable parameters increases to four (median, maximum and minimum sizes, standard deviation), which is much less than in the case of Approach 2.

The goal of this paper is to demonstrate that acoustic spectroscopy is capable of characterizing an asymmetrical particle size distribution using a "modified" log-normal distribution. In order to achieve this goal we chose an aqueous dispersion of commercial silicon powder. The full particle size distribution was determined by the X-ray sedimentation (XRS) technique. This method is able to provide the full distribution because it essentially separates particle fractions based on their settling rate in a gravitational field according to Stokes' law. This makes XRS quite different to acoustic spectroscopy, which deals with intact systems and the resulting combined signal coming from all fractions simultaneously.

The acoustic attenuation spectra of the concentrated silicon dispersion were measured with the PenKem-8000 Acoustophor.

## 2. Theoretical background

### 2.1. Mechanisms of sound attenuation

There are six known loss mechanisms of the sound interaction with a dispersed system: (1) viscous; (2) thermal; (3) scattering; (4) intrinsic; (5) structural; (6) electrokinetic.

(1) The viscous losses of the acoustic energy occur due to the shear wave generated by the particle oscillating in the acoustic pressure field. These shear waves appear because of the difference in the densities of the particles and medium. This density contrast causes the particle motion with respect to the medium. As a result the liquid layers

in the particle vicinity slide relative to each other. This sliding non-stationary motion of the liquid near the particle is referred to as a “shear wave” [1,2].

(2) The reason for the thermal losses is the temperature gradients generated near the particle surface. These temperature gradients are due to the thermodynamic coupling between pressure and temperature.

(3) The mechanism of the scattering losses is quite different for the viscous and thermal losses. The acoustic scattering mechanism is similar to light scattering. Acoustic scattering does not produce the dissipation of acoustic energy; particles simply redirect a part of the acoustic energy flow, and as a result this portion of the sound does not reach the sound transducer.

(4) The intrinsic losses of the acoustic energy occur due to the interaction of the sound wave with the materials of the particles and medium as homogeneous phases.

(5) The oscillation of the network of inter-particle links in a structured dispersed system causes structural losses. Thus, this mechanism is specific for structured systems.

(6) Oscillation of the charged particles in the acoustic field leads to the generation of an alternating electrical field and, consequently, alternating electric current. As a result, a part of the acoustic energy transforms to electric energy and then irreversibly to heat.

Only the first four loss mechanisms (viscous, thermal, scattering and intrinsic) make a significant contribution to the overall attenuation spectra in most cases. Structural losses are significant only in structured systems, which require a quite different theoretical framework. Finally, the contribution of electrokinetic losses to the total sound attenuation is almost always negligibly small [12] and will be neglected.

The general theory taking into account all four main mechanisms exists only for dilute systems when particle–particle interaction is not important. It is called the “ECAH” theory, as previously mentioned.

Only three mechanisms (viscous, scattering and intrinsic) are important for silicon particles. The

viscous and intrinsic losses will be calculated using theory developed in our previous paper [3]. This theory takes into account hydrodynamic particle–particle interaction and is valid for concentrated systems. The contribution of the scattering losses will be calculated following McClements [4] using Waterman–Truell theory [5] with lossless scatters.

## 2.2. Particle size distribution

Attenuation of sound in the dispersed system depends on the volume fraction of the dispersed phase. This means that the most reliable output of acoustic spectroscopy is particle size distribution on a weight or volume basis. Particle size data may then be recalculated in terms of either surface area (area basis) or number density (number basis). However, particle size distribution on either an area or number basis are less reliable.

The particle size distribution may be presented either as a cumulative distribution, as a differential distribution, or both. Cumulative distribution on a weight basis  $W_i$  is equal to a weight fraction of the particles with diameters smaller or equal to  $d_i$ . The weight fraction is determined relative to the total weight of solid phase. Differential distribution on a weight basis  $w_i$  defines a weight fraction of the particles with diameter values in the range  $(d_i + d_{i-1})/2$  to  $(d_{i+1} + d_i)/2$ . This weight fraction is equal to the product of  $w_i$  and the width of the above-mentioned diameter values range  $\Delta d_i = (d_{i+1} + d_{i-1} - 2d_i)/2$ . The width of the first range and of the last range is specially defined as  $\Delta d_1 = (d_2 - d_1)/2$  and  $\Delta d_N = (d_N - d_{N-1})/2$ , where  $N$  is a conventional number of fractions. The dimension of the parameter  $w_i$  is reciprocal length. The geometrical meaning of this definition is that the  $i$ th weight fraction is equal to the area of the  $i$ th column on the differential distribution histogram.

The particle size distribution may be presented either on a linear or on a logarithmic scale of the particle diameter. The definitions given above are valid for the linear scale. The relationship between differential distributions on these two scales is

given as

$$w_i(d_i) = w_i[\log(d_i)] \frac{\Delta \log(d_i)}{\Delta d_i}$$

The PenKem-8000 output particle size distribution is a differential distribution on a weight basis and logarithmic scale, i.e. it is  $w_i(\log(d_i))$ . This distribution is referred to as PSD and is normalized.

Four special distributions are used by the PenKem-8000: log-normal, bimodal, modified log-normal and modified bimodal. Only two distributions (log-normal and modified log-normal) are of interest in this paper.

The log-normal distribution is defined as a normal distribution of decimal logarithm in particle diameter. It is defined on a weight basis using a logarithmic scale.

$$w_i = \frac{\Delta d_i}{\Delta \log(d_i) \sigma_{\log} \sqrt{2\pi}} \exp \left\{ -0.5 \left[ \frac{\log(d_i) - \log(d^{\text{median}})}{\sigma_{\log}} \right]^2 \right\}$$

where

$$\sigma_{\log} = \sum_{i=1}^N w_i[\log(d_i)] \Delta \log(d_i) [\log(d_i) - \log(d^{\text{median}})]^2$$

A modified log-normal distribution is used when values of either  $d_{\min}$  or  $d_{\max}$  or both are known and restrict the values of particle size. This distribution is calculated according to the expression

$$w_i = \frac{\Delta d_i}{\Delta \log(d_i) \sigma_{\log} \sqrt{2\pi}} \exp \left[ -0.5 \left( \frac{M}{\sigma_{\log}} \right)^2 \right]$$

where

$$M = \log \frac{(d_i - d_{\min})(d_{\max} - d_{\min})}{d^{\text{median}}(d_{\max} - d_i)}$$

An example of the log-normal and modified log-normal distributions with the same median size (1  $\mu\text{m}$ ) and standard deviation (0.2) is shown in Fig. 1.

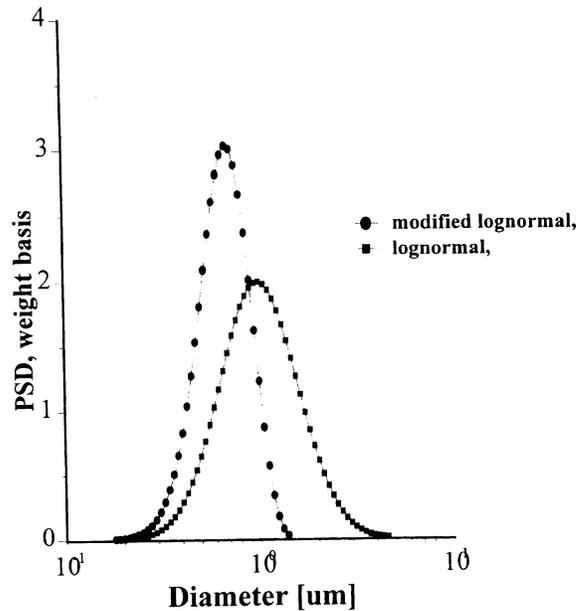


Fig. 1. Log-normal and modified log-normal distributions with the same median size (1  $\mu\text{m}$ ) and the same standard deviation (0.2). The maximum size of the modified log-normal distribution is restricted to 2  $\mu\text{m}$ .

It is important to mention that the median size of the modified log-normal distribution does not coincide with 50% size on the cumulative curve. The term median is somewhat misleading in this case; the name is inherited from the log-normal distribution. This parameter is mathematically the same for the two distributions. The meaning of this parameter is different.

### 3. Preparation silicon dispersion

A commercial jet-milled silicon powder (HQSilgrain, Elkem Materials Inc., Pittsburg, PA) was used to prepare suspensions for acoustic and sedimentation studies. This material is stated by the manufacturer to be less than 10  $\mu\text{m}$  particle size, and is characterized by a truncated particle size distribution. Suspensions were prepared by dispersing silicon powder in deionized water adjusted to  $\text{pH } 8.5 \pm 0.2$  (at 25°C) using a high intensity ultrasonic probe. All pH adjustments were made with 10 wt.% NaOH solution. Samples

for XRS and acoustic spectroscopy were prepared at 2.5 wt.% and 10.9 wt.% respectively, and aged 24 h to fully develop the necessary surface charge for dispersion. Homogeneous-phase density for this powder was determined by He pycnometry to be  $2.31 \pm 0.01 \text{ g cm}^{-3}$ . The uncertainty interval represents the standard deviation of three independent measurements.

#### 4. Measurement technique

The particle size distribution of the silicon powder suspension was determined by X-ray gravitational sedimentation using a Sedigraph 5100 (Micromeritics, Norcross, GA).<sup>1</sup> A resolution value of  $\pm 0.05 \mu\text{m}$  is commonly associated with this technique. Two runs were performed on separate suspensions to estimate the measurement uncertainty as a function of the cumulative percentile distribution. This data is shown in Table 1.

The attenuation spectrum was measured with the Acoustophor PK-8000. This instrument operates in the frequency range from 1 MHz to 100 MHz with a sample volume of at least 100 ml.

Table 1  
Cumulative size distribution for silicon dispersions measured by X-ray gravitational sedimentation

Weight fraction (%)	Diameter ( $\mu\text{m}$ )		
	Run 1	Run 2	Mean value
10	0.88	0.84	$0.86 \pm 0.03$
20	1.63	1.54	$1.59 \pm 0.06$
30	2.34	2.20	$2.27 \pm 0.10$
40	3.00	2.84	$2.92 \pm 0.11$
50	3.62	3.45	$3.54 \pm 0.12$
60	4.23	4.07	$4.15 \pm 0.12$
70	4.89	4.72	$4.81 \pm 0.11$
80	5.62	5.46	$5.54 \pm 0.11$
90	6.62	6.45	$6.54 \pm 0.12$

<sup>1</sup> Certain trade names and company products are mentioned in the text or identified in illustrations in order to adequately specify the experimental procedure and equipment used. In no case does such identification imply recommendation or endorsement by National Institute of Standards and Technology; neither does it imply that the products are necessarily the best available for the purpose.

The variable gap technique makes it possible to exclude calibration procedures.

Analysis software converts attenuation spectra into the particle size distribution, minimizing the difference between theoretical and measured attenuation spectra. The absolute error of the theoretical fit is the measure of this difference. In the case of the log-normal distribution the absolute error is minimized by adjusting median size and standard deviation of the log-normal distribution. In the case of the "modified" log-normal distribution the absolute error is also minimized by adjusting median size and standard deviation, but assuming a priori known values for minimum and/or maximum sizes.

Statistical uncertainty intervals reported for measured size data represent the estimated standard deviation of the mean at a series of independent observations. This interval, termed the *standard uncertainty*, is intended to identify and quantify random effects in the measurement.

The uncertainty associated with the experimental determination of acoustic attenuation was estimated by linear regression analysis of the measured total attenuation vs. the length of the gap between the transducer and receiver. An expanded uncertainty was then defined as the standard deviation multiplied by a coverage factor,  $k=3$ .

The uncertainty, or relative error, between theoretical fits and experimental attenuation spectra was estimated by dividing the sum of the differences for each frequency by the sum of the experimental values.

#### 5. Results

The particle size distribution measured with XRS (Run 1 is used as an example) is shown in Fig. 2. It is apparent that this distribution is truncated near  $10 \mu\text{m}$ ; it is definitely not log-normal. The cumulative mass distribution for each run, along with the calculated mean and sample standard deviation, is given in Table 1.

Attenuation spectra measured with the Acoustophor PK-8000 are shown in Fig. 3. Three runs are represented. Runs 1, 2 and 3 were analyzed on different instruments and separately pre-

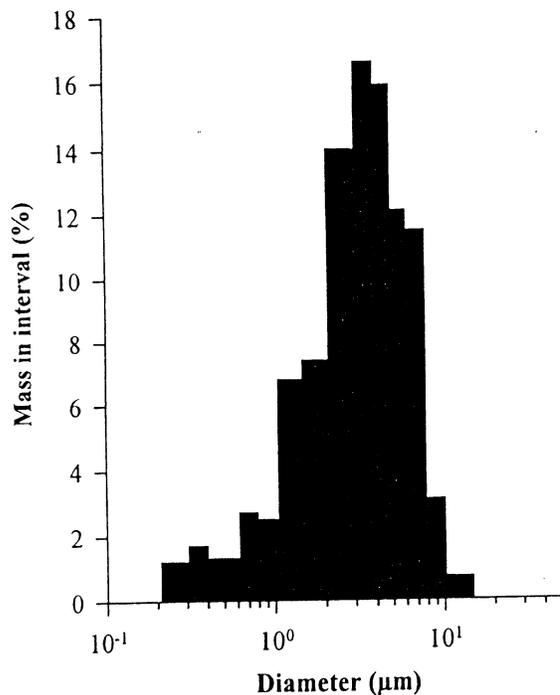


Fig. 2. The full particle size distribution measured by XRS analysis of a 2.5 wt.% silicon dispersion in water. Data correspond to Run 1.

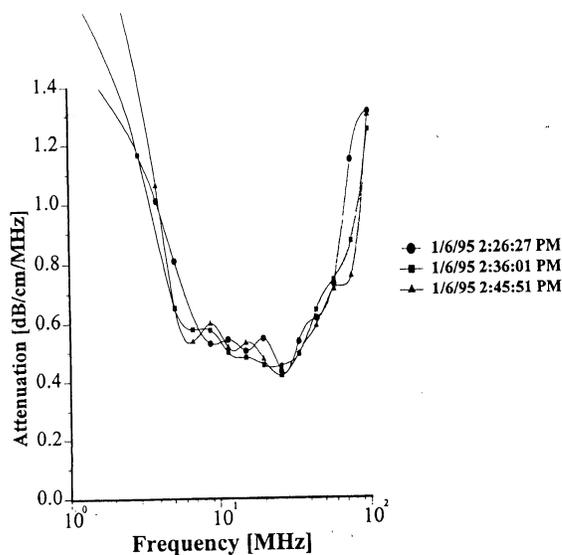


Fig. 3. Acoustic attenuation spectra measured with a PenKem-8000 Acoustophor for a 10.9 wt.% silicon dispersion in water.

pared samples. It is seen that reproducibility is quite good.

The log-normal distribution providing the best theoretical fit to the measured attenuation spectra is shown in Fig. 4 (Run 1 is used as an example). Corresponding theoretical and measured attenuation spectra are shown in Fig. 5. Cumulative log-normal distributions for each run are listed in Table 2.

## 6. Discussion

It is seen that log-normal distribution (Fig. 4, Table 2) differs significantly from the real particle size distribution (Fig. 2, Table 1). First of all, the median size is  $2.89 \pm 0.11 \mu\text{m}$  for the log-normal compared to  $3.54 \pm 0.12 \mu\text{m}$  determined by XRS. However, this log-normal distribution provides a theoretical attenuation spectra which gives a satisfactory fit to the measured attenuation spectra (Fig. 5, Table 3).

The "modified" log-normal distribution shown in Fig. 6 provides theoretical attenuation spectra fitting measured data equally well (Table 3). However, it is seen that this "modified" log-normal yields a distribution that is much more similar in

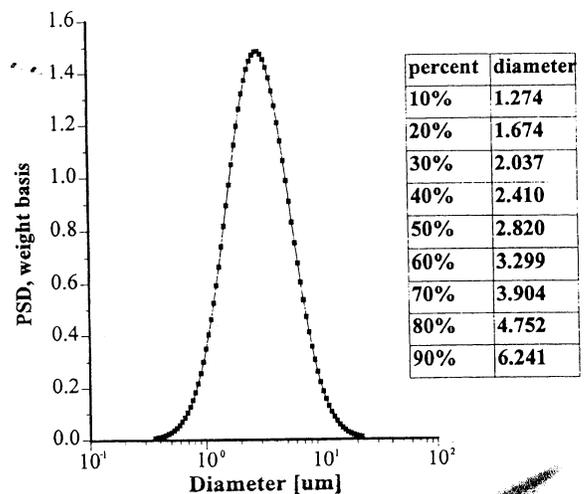


Fig. 4. The log-normal distribution providing the theoretical attenuation spectra with the best fit to the measured attenuation spectra. Data correspond to Run 1.

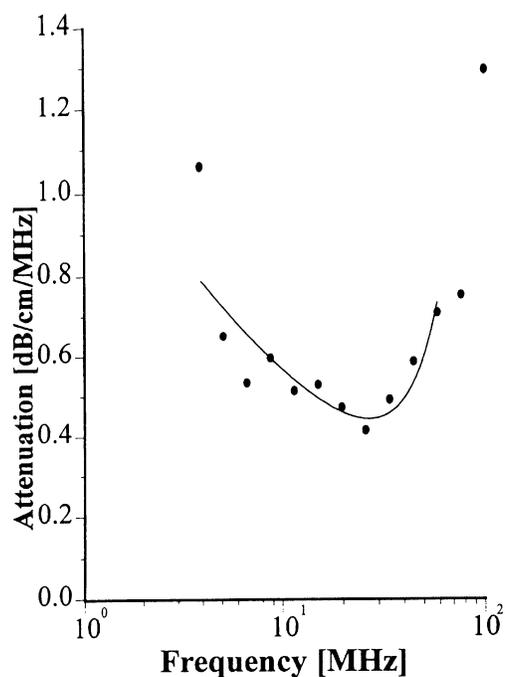


Fig. 5. Theoretical and measured attenuation spectra. Theoretical spectra correspond to the log-normal distribution shown in Fig. 4.

form and mean size to the XRS distribution (Fig. 2). This means that acoustic spectroscopy is able to accurately characterize dispersed systems

Table 3

Experimental and theoretical fitting errors associated with acoustic attenuation measurements

Run	Experimental error (%)	Log-normal fit error (%)	Modified log-normal fit error (%)
1	5	9.3	9.1
2	4.9	10.1	9.7
3	1.7	5.7	5.8

with asymmetrical particle size distributions. A priori information regarding the minimum and/or maximum sizes is necessary.

The maximum particle size has been restricted to 10  $\mu\text{m}$  for the "modified" log-normal distribution. This means that only 2.3% of particles mass has been truncated from the log-normal distribution shown in the Fig. 4. This small change causes a relatively large change in the corresponding median size and, perhaps more significantly, in the overall shape of the distribution.

The reason of this strong non-linearity is a dominant contribution of the scattering losses for large particles at high frequency. The relative contribution of the viscous and scattering losses to the total attenuation spectra is shown on the Fig. 7 for the log-normal distribution from Fig. 4.

Table 2

Cumulative size distributions calculated for the log-normal and modified log-normal functions providing the best fit to the measured acoustic attenuation spectra

Weight fraction (%)	Diameter ( $\mu\text{m}$ )							
	Log-normal				Modified log-normal			
	Run 1	Run 2	Run 3	Mean value	Run 1	Run 2	Run 3	Mean value
10	1.38	1.27	1.30	$1.32 \pm 0.05$	0.97	0.95	0.96	$0.96 \pm 0.01$
20	1.80	1.67	1.70	$1.72 \pm 0.07$	1.59	1.50	1.50	$1.53 \pm 0.05$
30	2.17	2.03	2.05	$2.08 \pm 0.08$	2.19	2.01	2.03	$2.07 \pm 0.09$
40	2.58	2.42	2.43	$2.48 \pm 0.09$	2.81	2.53	2.58	$2.64 \pm 0.15$
50	3.01	2.82	2.83	$2.89 \pm 0.11$	3.50	3.09	3.19	$3.26 \pm 0.21$
60	3.49	3.29	3.30	$3.36 \pm 0.11$	4.28	3.74	3.87	$3.96 \pm 0.28$
70	4.08	3.84	3.86	$3.93 \pm 0.13$	5.13	4.48	4.67	$4.76 \pm 0.33$
80	4.91	4.65	4.64	$4.73 \pm 0.15$	6.06	5.38	5.57	$5.67 \pm 0.35$
90	6.41	6.13	6.07	$6.20 \pm 0.18$	7.20	6.50	6.71	$6.80 \pm 0.36$

The best fit "modified" log-normal distribution is shown in Fig. 6. The value of the minimum size was 1 nm. The value of the maximum size was 10  $\mu\text{m}$ . Cumulative "modified" log-normal distributions for each run are listed in this table.

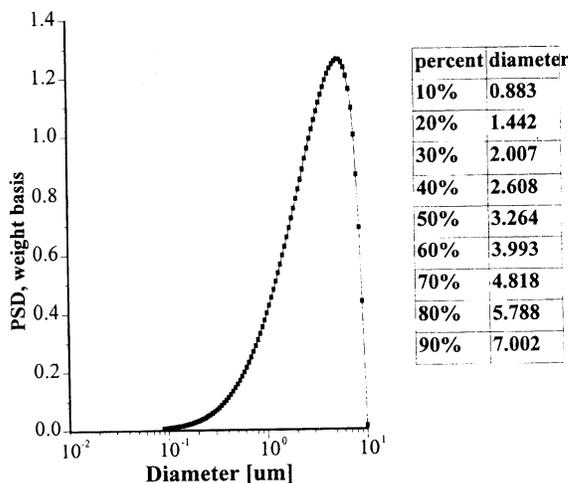


Fig. 6. The modified log-normal distribution providing the theoretical attenuation spectra with the best fit to the measured attenuation spectra. Maximum particle size is restricted to 10  $\mu\text{m}$ . Data correspond to the Run 1.

It is clearly seen that scattering losses dominate in the high frequency range.

The aggregation process is one of the important cases where the modified log-normal distribution can be useful. Aggregation leads to an increase of the particle size. The minimum size would be

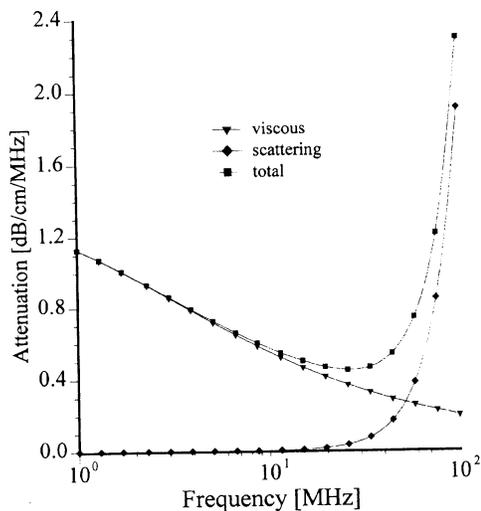


Fig. 7. Total theoretical attenuation, viscous and scattering losses corresponding to the log-normal distribution shown in Fig. 4.

expected to remain more or less constant, representing the primary particles; it may increase eventually, but definitely does not decrease. This minimum size can be defined for the stable system, and used as a priori information for a “modified” log-normal distribution when an aggregated system is characterized.

Abrasive and polishing materials is another example where the “modified” log-normal distribution is very useful. Special efforts are usually made to remove the particles exceeding some specified size. Thus, the particle size distribution in dispersions of abrasive and polishing materials are truncated at some known maximum size. Acoustic spectroscopy makes it possible to monitor effectively the presence of the large particles due to the dominant effect of scattering losses. This means that acoustic spectroscopy, in conjunction with the “modified” log-normal distribution, could be a useful tool for quality control of abrasive and polishing materials.

## 7. Conclusions

Acoustic spectroscopy requires a “modified” log-normal distribution in addition to the standard log-normal function in order to characterize systems with truncated particle size distribution. The error resulting from the search for the median particle size in a fitting procedure using only the log-normal distribution might easily reach 50% and more. The modified log-normal distribution described in this paper is especially effective when large particles above 3  $\mu\text{m}$  are truncated. These particles are expected to cause strong scattering losses, and acoustic spectra reflect the presence of these large particles through the scattering losses contribution to attenuation.

The modified log-normal distribution requires a priori information regarding minimum and/or maximum sizes. Information about minimum size is available in the case of an unstable aggregating system from an analysis of the stable dispersion. Abrasive and polishing materials is another important example when a priori information regarding maximum size is available.

The combined methodology described herein

was satisfactorily tested using a commercial silicon powder dispersed in aqueous media with a maximum size truncated at 10  $\mu\text{m}$ . Results were compared to the size distribution determined independently by XRS.

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