Neural Network Particle Sizing in Slurries by Reflectance Spectroscopy

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Measuring concentration and size of solids in suspension is important in many industries. Even though techniques based on optical transmission measurements have been well developed, they are not always successful in practical applications because low concentration suspensions are needed. A method developed determines particle-size distribution and concentration from reflection measurements in concentrated suspensions using neural networks with particle concentrations up to 10% volume fraction. Based on measured optical reflectance spectra of suspensions with known particle-size distributions and concentrations, a neural network was trained to identify particle-size distribution and volume fraction of suspensions. Training is a time-consuming process requiring presentation of many spectra and their corresponding particle-size distributions and volume fractions to the neural network, but once concluded satisfactorily, the neural network can be used to predict the particle-size distribution and volume fraction of high concentration suspensions rapidly in-situ.

Introduction

The measurement of the concentration and size of solid particles in a suspension is important in many industries such as manufacturing pigments, pharmaceuticals, cosmetics, and foods. Furthermore, accurate control of product particle size is a significant feature of many material specifications.

Methods to recover particle-size distribution from spectral extinction measurements are well developed (Melik and Fogler, 1983; Wilkinson and Waldie, 1991; Gordon et al., 2000; Li and Wilkinson, 2001a,b). The light absorption characteristics of a particulate suspension can be determined from transmission measurements between an emitter and receiver separated by a short distance. However, transmission measurements are impractical for high concentration suspensions. At present, transmission measurements can be used to measure particle-size distribution at particle concentrations up to only about 3% by volume.

Alternatively, a reflection fiber optic probe makes *in-situ* measurements possible at a high particle concentration. Incident light from the delivery fiber is scattered and also partly absorbed. The reflected light intensity received by the collection fiber is a function not only of the particle concentration, but also of numerous factors such as size, shape, refractive index of particles, and geometrical configuration of optical fibers. In a dilute suspension, each particle may be regarded as an isolated scatter of light and reflection by the suspension is simply the sum of the individual particle contributions. The reflected intensity depends on the wavelength of the radiation and on the particles' size, shape and refractive index and is well described by Mie theory (Kerker, 1969). When particle volume concentration is greater than approximately 1%, single particle scattering theory is no longer valid and multiple scattering occurs. Three approaches to describe multiple scattering problems have been developed: analytical theories, transport theory, and the Monte Carlo method (Ishimaru, 1978; Groenhuis et al., 1983; Bergougnoux et al., 1996). Unlike Mie theory for single scattering, these models for multiple scattering are still far from practical application. An alternative approach to using reflection measurements for particle-size distribution and concentration determination is first to calibrate the fiber sensor by known samples. Then, the calibrated sensor is subsequently used to measure unknown suspensions. Optical fiber sensors based on light reflection by

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particles are widely used to measure the volume fraction of suspensions (Kineke and Sternberg, 1992; Conner and De Visser, 1992; Bergougnoux et al., 1999), prior to obtaining the particle-size distribution by other means. Other works based on light reflection have determined the particle mean size of suspensions (Bemer, 1978; Bos and Heerenns, 1982; Heffel et al., 1996), prior to finding the suspension concentration. The ability to measure concentration and particle-size distribution in concentrated suspensions at sizes from 0.1 to 0.5 micron has been developed using photon migration techniques by Sevick-Muraca and coworkers (Jiang et al., 1997; Shinde et al., 1999). However, it is still an open problem to determine particle size and volume fraction in high concentration suspensions simultaneously by reflectance spectroscopy.

In this work, a method to determine the particle-size distribution and volume fraction of concentrated suspensions using neural networks from reflection measurements has been investigated. To simplify the problem, we assume that the particles are spherical and obey the log normal distribution function, which is characterized by the particle geometric mean size and standard deviation. The range of particle concentrations investigated is up to 10% volume fraction. Training is a time-consuming process requiring presentation of many sets of spectra and their corresponding particle-size distributions and volume fractions to the neural network, but once concluded satisfactorily, the neural network can be used to predict the particle-size distribution and volume fraction in high concentration suspensions *in-situ* essentially immediately. Based on measured optical reflection spectra of suspensions with known particle-size distributions and concentrations, a neural network has been trained by an effective training algorithm, the Levenberg-Marquardt (LM) algorithm (Hagan and Menhaj, 1994), to identify the particle-size distribution and volume fraction of suspensions.

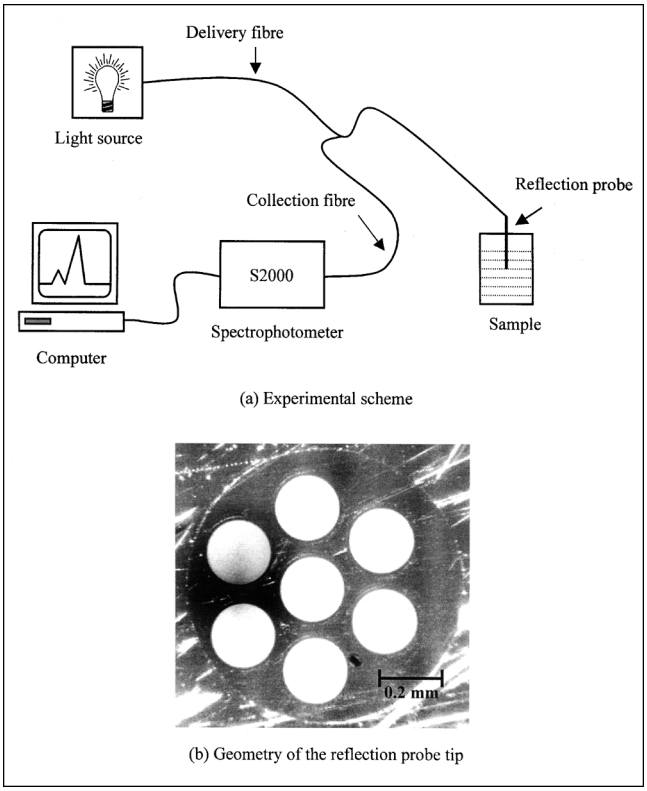

Figure 1. Experimental configuration of reflection measurements.

Experimental Details

Instrument description

Reflection measurements were carried out using a commercial spectrophotometer (Ocean Optics S2000). Figure 1a shows the scheme of the reflection measurement equipment. The reflection probe (Ocean Optics FCR-7UV) consisting of delivery fibers (six in our experiment) connected with a light source and collection fibers (one in our experiment) connected with the spectrophotometer is used to gain spectral information about the suspension. The geometry of fibers in the reflection probe is shown in the photomicrograph Figure 1b. The central circle is the collection fiber, which is surrounded by the six delivery fibers. All the fibers have the same specifications. The diameter of each fiber is 200 micron, and its numerical aperture is 0.22. The reflection probe is simply dipped into the sample to perform the measurements. Polychromatic light from the visible source (7W tungsten halogen bulb) is passed by the delivery fibers to the suspension. The reflected light then travels by the collection fiber to the S2000 fiber optic spectrophotometer. Light detection in the spectrophotometer is via a 2048 element CCD linear array, which splits the beam into its corresponding wavelengths. The information is then transferred to the spectrophotometer operating software package (Ocean Optics OOIBase32 Spectrometer Operating Software) where data on intensity vs. wavelength are recorded.

Experiments

Silica was chosen as the material used for the preparation of suspensions, because it is readily available commercially in a range of different size distributions. The silica used was a spherical powder with size distributions which were measured by laser diffraction (Malvern Mastersizer). Suspensions were prepared by measuring known masses of dry silica directly into volumetric flasks, and making up to a volume with deionized water, hence, giving samples with a known volumetric concentration. Care was taken to minimize exposure of the dry sample to the atmosphere to ensure that the mass of the sample taken was that of the particles only. Before any suspension was analyzed, it was placed in an ultrasound bath for approximately 20 min to break down any aggregates. The suspension of known concentration was then placed in an enclosed matt black painted beaker to eliminate the effect of ambient light on the results. A dark spectrum was collected with no light emitted from the probe to eliminate the electronic background signal. Reflected spectra were then collected from sample suspensions. The spectrophotometer recorded spectral intensity values over the range of wavelengths from 300 to 1,000 nm. In the experiments, the integration time of the spectrometer was set to be 7 milliseconds, maximizing the amount of light collected without exceeding the maximum limit of the spectrophotometer. The minimum

SNR (signal vs. noise ratio) of a single scan was typically 100. The operating software function "Samples to Dynamically Average" is a signal averaging function that averages several spectra after collection. In our experiments, the number of scanned spectra was 10 giving an SNR of approximately 300 for the averaged result.

The intensity of reflected light is a function not only of the particle concentration, but also of numerous factors such as size, shape, and refractive index of particles, geometrical configuration of optical fibers, optical path length through the suspension, and intensity of the light source. The optical path length in transmission measurements is the length of the direct line through the suspension between the source and detector. However, in reflectance measurements the optical path length is not easily defined. In our experiments, the maximum possible optical path length is defined as the length between the location of the probe and the bottom of the sample vessel. The maximum optical path length of the probe was found to influence the reflected intensities at different wavelengths. To determine a relationship between particle-size distribution and concentration of a suspension and the measured spectrum, it is necessary to eliminate the effects of the maximum possible optical path length and the intensity of the light source.

Several experiments were done to find the relationship between the reflected intensity and the maximum possible optical path length. For suspensions with different particle-size distributions and concentrations, Figure 2 shows the measured reflected intensities for different maximum possible path lengths over the range of wavelengths from 300 to 1,000 nm. It is seen that the reflected intensities at different wavelengths are independent of the maximum possible path length when it is larger than 5 mm. It means that the reflected intensity is determined only by the suspension's properties for path lengths over 5 mm. Thus, the distance from the sensor location to the bottom of the beaker was chosen to be larger than 5 mm in our experiments.

To eliminate the intrinsic intensity of the light source from the problem, the reflection measurement is expressed as a referenced measurement, the intensity relative to the reflected intensity from a standard reference substance

$$B(\lambda) = \frac{S(\lambda) - D(\lambda)}{R(\lambda) - D(\lambda)} \tag{1}$$

where $S(\lambda)$ is the reflected intensity measured at wavelength λ, $D(\lambda)$ is the dark intensity measured at wavelength λ, and $R(\lambda)$ is the reference reflected intensity measured at wavelength λ. Here, a suspension of 1% volume fraction silica particles with mean 0.38 micron and deviation 1.25 was chosen as the reference.

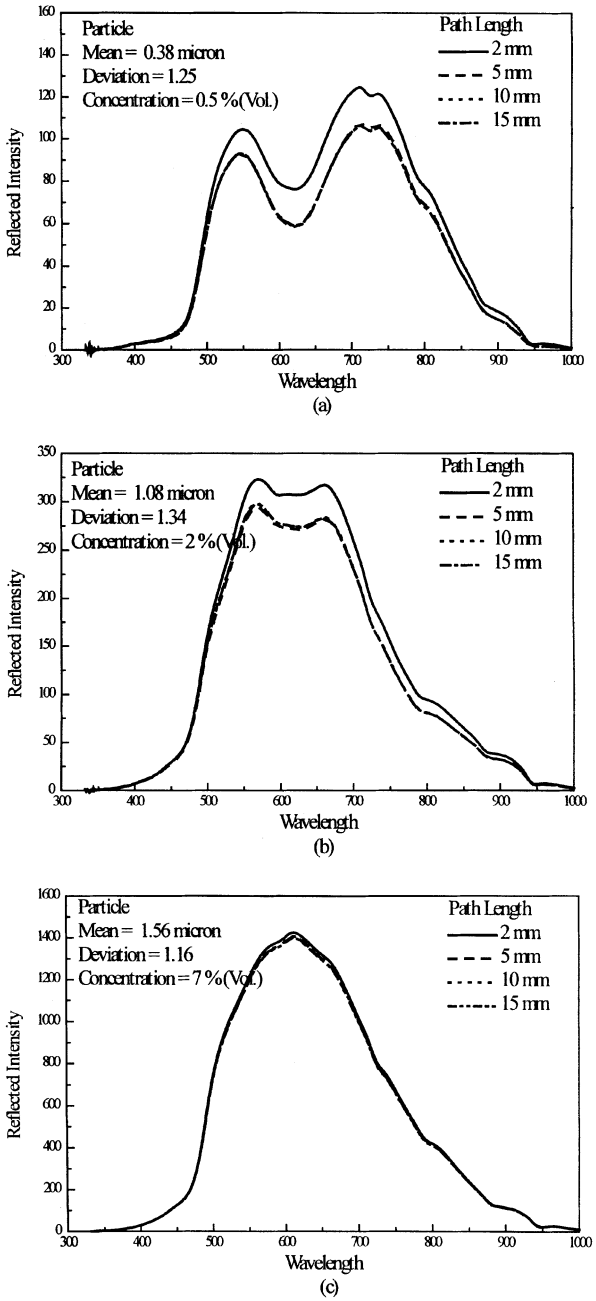

Figure 2. Reflected intensity spectra for different maximum possible path lengths.

Particle-Size Distribution and Concentration Determination Using Neural Networks

Neural networks

Neural networks have been attracting great interest as predictive models for complex nonlinear processes because of their outstanding ability in approximating an arbitrary nonlinear function. Success in using neural networks depends strongly on the choice of the neural network's structure, the available set of data, and training algorithms.

The most common neural network for modeling processes is the feedforward network. Figure 3 represents an n-input, m-output feedforward neural network with one hidden layer having n_h hidden units.

The output of the hidden units can be represented as

$$h_j = \rho(s_j) = \rho\left(\boldsymbol{V}_j^T \boldsymbol{I}\right), \qquad j = 1, 2, \cdots, n_h \tag{2}$$

where $\boldsymbol{I} = [1, x_1, \cdots, x_n]^T$ is the network input vector, $\boldsymbol{V}_j = [v_{j,0}, v_{j,1}, \cdots, v_{j,n}]^T$ is the weight vector connecting the net-

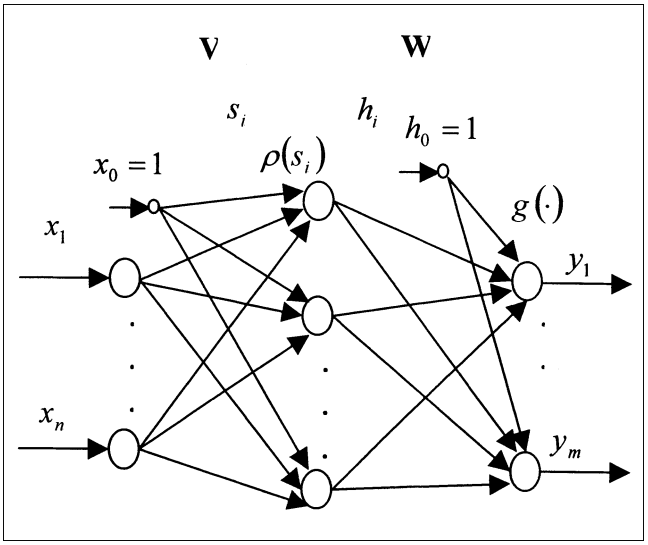

Figure 3. Structure of a feedforward neural network.

work inputs to the jth hidden unit and $\rho(x)$ is the sigmoid function. The output of the output layer neurons can be represented as

$$y_i = g(\boldsymbol{W}_i^T \boldsymbol{H}), \qquad i = 1, 2, \cdots, m \tag{3}$$

where $\boldsymbol{H} = [1, h_1, \cdots, h_{n_h}]^T$ is the network hidden layer output vector, $\boldsymbol{W}_i = [w_{i,0}, w_{i,1}, \cdots, w_{i,n_h}]^T$ is the weight vector connecting the network hidden layer units to the ith output neuron, and $g(x)$ may be either a line function or a sigmoid function depending on the problem. In this work, $g(x) = \rho(x) = 1/(1 + e^{-x})$, the sigmoid function, was selected.

The neural network defines a mapping $G : \boldsymbol{X} \rightarrow \boldsymbol{Y}$, where $\boldsymbol{X} \in \boldsymbol{R}^n$ is an input vector and $\boldsymbol{Y} \in \boldsymbol{R}^m$ is an output vector. Any nonlinear function can be approximated by the network through appropriately determined weight matrices $\boldsymbol{V} = [\boldsymbol{V}_1, \boldsymbol{V}_2, \cdots, \boldsymbol{V}_{n_h}]$ and $\boldsymbol{W} = [\boldsymbol{W}_1, \boldsymbol{W}_2, \cdots, \boldsymbol{W}_m]$.

The training of a neural network may be classified into either batch learning or pattern learning. With batch learning, the weights of the neural network are adjusted after a complete sweep of the entire training data, while, with pattern learning, the weights are updated during the course of the process using data gained online. Batch learning has greater mathematical validity as the gradient-descent method can be implemented exactly. Pattern learning, usually applied as batch learning approximations, can be used to modify network weights online so that a model can track the dynamics of a time-varying process. In this work, batch learning was selected as appropriate to train the network off-line.

Particle-size distribution and concentration determination

Eight different particle distributions were used to prepare suspensions. The suspension volume fractions varied from 0.1% to 10%. In total, 137 pairs of data were collected to train the neural network. Another 15 pairs of test data were used to test the neural network estimation results. Table 1 shows the parameters of the diluted distributions as measured by laser diffraction. Figure 4 shows reflected light intensities vs. solid concentrations for three different particle-size distributions at two different wavelengths. It is seen that the spectral intensity increases as the suspension concentration increases and the spectra are affected by particle size and measurement wavelength.

Table 1. Silica Particles*

Type	Mean (μm)	Dev.
1	0.38	1.25
2	1.08	1.34
3	1.56	1.16
4	11.3	1.46
5	7.25	1.70
6	38.6	1.75
7	2.91	2.00
8	1.46	2.05

*Nineteen concentration values in the range 0.1 (0.1) 0.9 vol. % and 1 (1) 10 vol. %.

Several items must be defined in order to use neural networks to determine particle-size distribution and concentration. These are the selection of the network inputs and outputs, the number of hidden units and the training algorithm. In this case, the network outputs are particle mean and deviation and volume fraction. The choice of the network inputs is a little more difficult, including the number of network in-

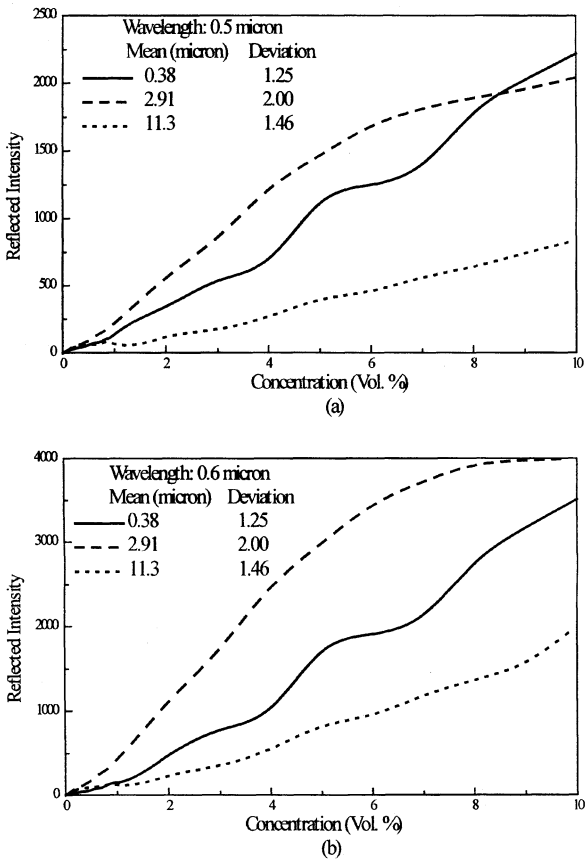

Figure 4. Effect of concentration on reflected intensity.

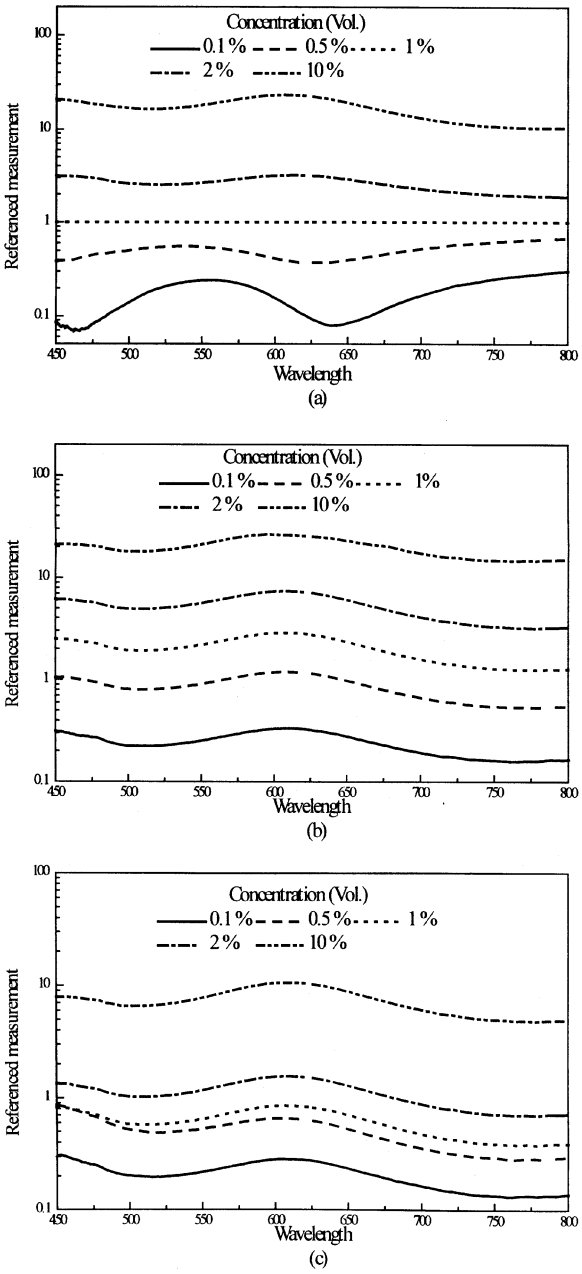

Figure 5. Referenced measurements for different particle-size distributions and volume fractions.

(a) Mean 0.38 μm, deviation 1.25; (b) mean 2.91 μm, deviation 2.00; (c) mean 11.3 μm, deviation 1.46.

puts (referenced measurements) and their corresponding wavelengths. From the experiments, it was found that the effective wavelength range is approximately only from 450 to 800 nm, although the instrument can measure wavelengths from 300 to 1,000 nm. This is the effective wavelength range of the light source and the spectra are less affected by noise. Figure 5 shows referenced measurements of measured reflected intensity relative to the reference spectrum (the suspension of 1% volume fraction and silica particles with 0.38 micron mean and 1.25 deviation) for three different particle-size distributions and at five volume fractions over the range of wavelengths from 450 to 800 nm. From Figures 4 and 5, it is clear that the referenced measurement is a nonlinear function of the suspension's concentration over the whole range from 0 to 10% volume fractions. When the suspension's concentration is less than 1% volume fraction, it is seen from the curves that the relationship between a referenced measurement and concentration is nearly linear, so that the single particle scattering theory is valid. The same result was also obtained by Shinde et al. (1999). With increasing concentration, multiple scattering occurs. For a fixed concentration, the referenced measurement is affected by particle size; it generally decreases with increasing particle mean size.

As the referenced measurement curves show significant variations for different distributions and concentrations, neural networks can be used to recover the particle-size distribution and concentration. From inspection of the shape of these curves, it was concluded that five spectra at wavelengths of 500, 550, 600, 650 and 750 nm are sufficient to represent the differences among the particle-size distributions and volume fractions. It is important to maximize the information content of data input to the neural network without requiring an excessive number of input nodes. Thus, the number of network inputs was set at five. The values of the wavelengths were selected following inspection of Figure 5 and other similar curves to achieve maximum information input with a limited number of input nodes.

Two important steps in the training procedure were the determination of an adequate number of neurons in the hidden layer and the selection of a training algorithm. Training experiments using from 15 to 21 hidden neurons were carried out in order to determine the appropriate number of hidden units according to a criterion of minimum mean square error (MSE) between the desired outputs and the calculated outputs from the network. There are several training algorithms, such as the Adaptive Backpropagation (ABP) algorithm (Weir, 1991), Conjugate Gradient (CG) algorithm (Xu et al., 1996), and Levenberg-Marquardt (LM) algorithm (Hagan and Menhaj, 1994). The LM algorithm was used to train the neural network, as it had been shown that the LM algorithm is the most effective training algorithm (Li and Wilkinson, 2001b). A maximum of 1,000 iterations were used in all runs. The training results are shown in Figure 6. It is clear that the neural network with 17 hidden neurons trained by the LM algorithm gave the best approximating results. The network training procedure was finished in 20 min using a 500 MHz Pentium processor. The recovered results obtained by the neural network are shown in Table 2 for different suspensions. It is seen that the recovered results generally agree well with the true values.

Discussion and Conclusion

In this article, a method to determine particle-size distributions and concentrations by reflectance spectroscopy using neural networks is proposed. Training data were obtained from reflectance measurements in suspensions of known particle-size distributions and concentrations. Five relative reflectance measurements at different wavelengths were chosen as the network inputs, and the network outputs were the particle-size distribution (mean and deviation) and concentration. A neural network with 17 hidden neurons was trained

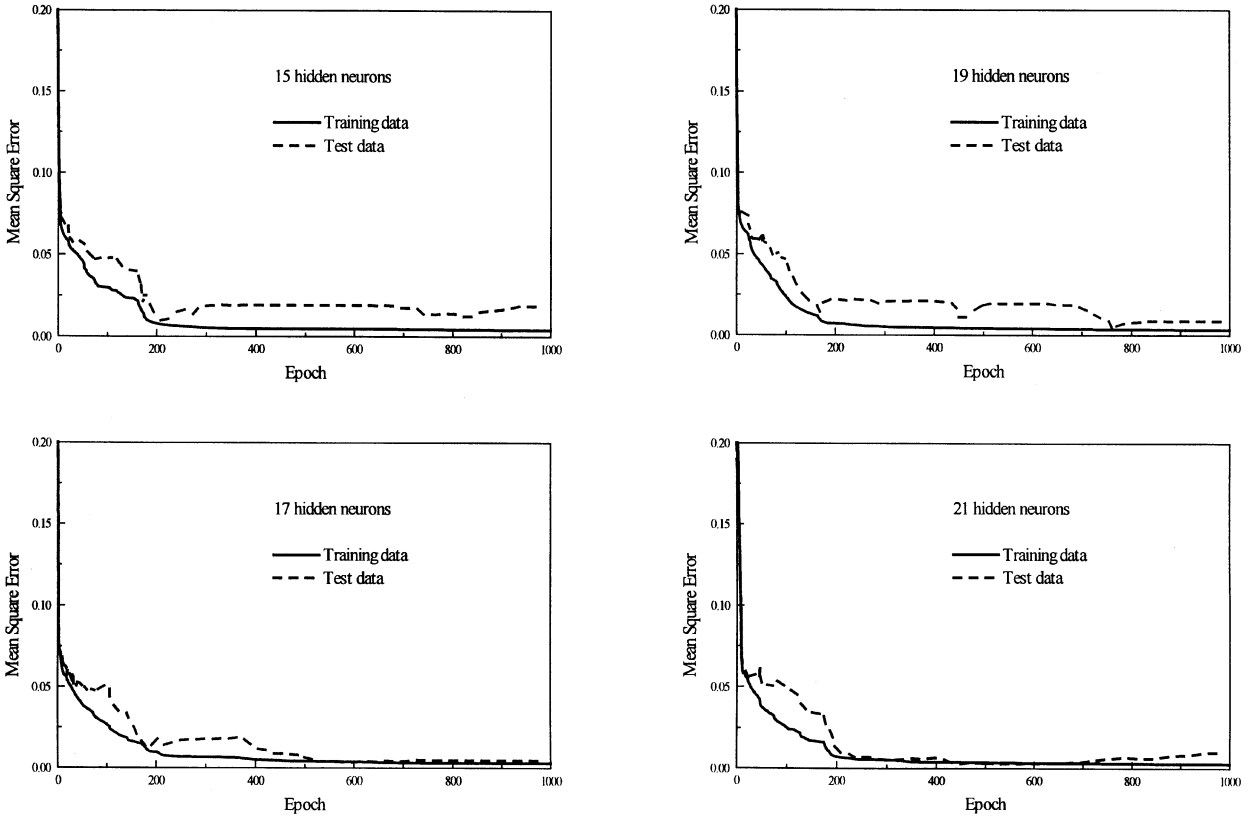

Figure 6. Neural network training results.

by the Levenberg-Marquardt algorithm. The test results confirmed that it is feasible to use neural networks to obtain the parameters of a particle-size distribution and concentration at high concentration suspensions by reflectance measurements. The method has several advantages over existing techniques including rapid data acquisition, simplicity of use, applicability to high concentration suspensions, and obtaining the particle-size distribution and concentration simultaneously.

Success in using neural networks depends greatly on the training data. The ability of neural networks to predict the unknown particle-size distribution and concentration of a suspension needs enough training data covering the entire ranges of all the parameters. It is difficult to obtain enough training data from experiments only. Thus, it is useful to develop a reflectance model to provide enough training data for the neural network. Work is ongoing to simulate the multiple scattering processes.

In this work reflectance was applied successfully in suspensions at up to 10% volume concentration, but there is no indication that this is a maximum limit of the method. The experimental material used in this work was silica powder,

Table 2. Results Recovered by Neural Network

	Mean (μ)		Deviation		Concentration (Vol. %)	
No.	True Value	NN Result	True Value	NN Result	True Value	NN Result
1	0.38	0.38	1.25	1.25	0.50	0.15
2	1.08	0.82	1.34	1.38	0.40	0.38
3	1.56	1.38	1.16	1.16	0.30	0.20
4	0.38	0.38	1.25	1.25	2.00	1.95
5	1.08	0.94	1.34	1.37	3.00	2.80
6	1.56	1.07	1.16	1.18	4.00	3.66
7	11.3	11.3	1.46	1.50	0.20	0.40
8	11.3	10.8	1.46	1.50	5.00	8.24
9	38.6	38.6	1.75	1.81	6.00	5.47
10	7.25	7.32	1.70	1.76	0.10	0.24
11	38.6	38.6	1.75	1.80	0.60	0.48
12	2.90	0.38	2.00	1.94	0.70	0.90
13	7.24	7.36	1.70	1.50	2.00	2.86
14	2.91	2.39	2.00	2.04	6.00	6.02
15	1.46	0.89	2.05	2.00	1.00	1.04

which has a small relative refractive index compared to water. Work will be carried out in the future on the application of reflectance measurements to efficient scatters as used in paints, pigments, and pharmaceuticals and at industrially relevant concentrations from about 30% to 50% by volume.

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