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AN EVALUATION OF THE MALVERN OPTICAL PARTICLE MONITOR

By
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July 1983

U. S. Department of Energy
Morgantown Energy Technology Center
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INTRODUCTION

The Malvern 2200/3300 Particle Sizer is a laser-based optical particle sizing device which utilizes the principle of Fraunhofer Diffraction as the means of particle size measurement. The instrument is designed to analyze particle sizes in the range of 1 to 1800 microns diameter through a selection of lenses for the receiving optics. It is not a single-particle counter but rather an ensemble averager over the distribution of particles present in the measuring volume. Through appropriate measurement techniques, the instrument can measure the volumetric size distribution of:

- o Solids in gas or liquid suspension;
- o Liquid droplets in gas or other immiscible liquids; and,
- o Gas bubbles in liquid. (Malvern Handbook, Version 1.5)

This report details a limited laboratory evaluation of the Malvern system to determine its operational characteristics, limitations, and accuracy. This investigation focused on relatively small particles in the range of 5 to 150 microns. Primarily, well characterized particles of coal in a coal and water mixture were utilized, but a selection of naturally occurring, industrially generated, and standard samples (i.e., glass beads) were also tested. The characteristic size parameter from the Malvern system for each of these samples was compared with the results of a Coulter particle counter (Model TA II) analysis to determine the size measurement accuracy. Most of the particulate samples were suspended in a liquid media (water or isoton, plus a dispersant) for the size characterization.

Specifically, the investigations contained in this report fall into four categories: (a) Sample-to-lense distance and sample concentration studies, (b) studies testing the applicability to aerosols, (c) tests of the manufacturer supplied software, and (d) size measurement comparisons with the results of Coulter analysis. Sample-to-lense distance and sample concentration effects on the characteristic size and width of the distribution measured by the Malvern system were carried out for both the Malvern flow sample cell and stirred cell using 63-, 100-, and 300-mm lenses. Capabilities for the system to measure particles entrained in gas streams were examined through the design of a simple aerosol cell to determine the limitations placed on the measured result by window fouling. In addition to mechanical tests, characteristics of the software data-fitting routine were also examined. Criteria to select which of the three available distribution functions (Normal, Rosin-Rammler, and Log-Normal) best fits the data were determined and sensitivity to abnormal data was tested. Also, the use of model independent software, in cases of data known to or not to possess one of the three available size distributions, was examined.

EQUIPMENT

The Malvern 2200/3300 Particle Sizer tested consisted of five basic components (see Figure 1):

1. Optical measurement unit;
2. Commodore Model No. 4032-32N computer;
3. Commodore Model No. 4022 bidirectional printer;
4. Malvern software package; and,
5. sample cells and lense selection.

The transmitting end of the optical unit contained a 1mW He-Ne laser which was spatially filtered through a 35 μ m pinhole and collimated to produce a parallel monochromatic beam of light. This light was arranged to illuminate a volume of particles whose Fraunhofer Diffraction was received and analyzed by the instrument. The resulting diffraction pattern was, in theory, stationary and independent of the position or movement of the particles (Swithenbank, et al., 1976). As the particles entered and left the illuminated area, the pattern at any instant reflected the size distribution present. A representative bulk analysis of the particles was obtained by integration of the scattered light signal for a specific period of time.

A Fourier transforming lense was used to focus the diffraction pattern of near-forward scattered light onto a multi-element, photoelectric detector placed at the focal plane of the lense. The detector consisted of 32 semicircular photosensitive rings surrounding a circular hole at the center behind which a separate photodiode was located. In a very simple sense, each ring was sensitive to the diffraction from a particular size particle, so the output was an analog signal proportional to the received light intensity and characteristic of the particles present. The center hole was used to monitor transmission of the incident beam for determinations of intensity extinction and optical alignment. The entire detector array was interfaced directly to the Commodore minicomputer which sampled the instantaneous diffraction pattern and performed the necessary signal integration digitally.

Once a predetermined number of instantaneous patterns had been collected and averaged to produce a pattern characteristic of the bulk sample, the computer used the method of nonlinear least squares analysis to find the appropriate constants for a preselected distribution function that produces a computed diffraction pattern most similar to that experimentally observed. The distribution used in the analysis was chosen by the operator from one of the three available two-parameter models: Rosin-Rammler, Log-Normal, Normal. A model independent approach was also available which matched the weight in 15 size bands to the data, thus producing an illustration of the shape of the size distribution -- but not a measure of the mean size or spread. The model independent analysis was generally used when the other distribution functions provided a poor fit as measured by the log error. For example, the two-parameter models were not suitable for multimodal samples. The choice of

FIGURE 1. BASIC OPTO-ELECTRONIC SYSTEM OF THE MALVERN OPTICAL PARTICLE MONITOR

selecting one of the two-parameter models or the model independent method in data analysis was left to user discretion.

If one of the two parameter models was used in the analysis, a size parameter ($\bar{X}$) and a distribution width parameter (N) were generated that were characteristic of the bulk distribution. These parameters were used in conjunction with the internal models to generate the relative weight (mass) of the particulate sample per preselected size interval (particle diameter). The cumulative weight below or above a given size was also generated. The size intervals on which the analysis was based were determined by the receiving lense and the distribution of the photodiode array sensors. However, the size intervals used in the generated distribution were determined by the user to examine specific areas or extrapolate results to sizes not physically measured. If the model independent method was used, then only weight in the fixed size bands was available and no extrapolations from a model were possible.

Results of analysis and numerical listings of input data were available via the line printer while graphical presentations were available on the CRT screen of the computer or printable as hard copy.

GENERAL OPERATION

Alignment Procedure

Ease of optical alignment and stability of the alignment are major advantages of an ensemble scattering system over single particle scatters. Optical alignment was significantly less complicated for the Malvern than for comparably sophisticated single particle scattering devices.

Prior to initiation of the evaluation of the Malvern system, the optics of the instrument were carefully aligned according to manufacturer alignment procedures. The alignment was divided into two phases, the transmitter section and the receiving section. Housed in the transmitter were the He-Ne laser, its power supply, a spatial filtering lense and pinhole, plus a collimating lense. The recommended procedures to align the transmitter were straightforward. The manufacturer's suggestion to make target screens to assist optical alignment was found to be very useful. One exception to the handbook alignment procedure was observed. Without the collimating lense, a much expanded diverging laser beam was generated which was to be properly aligned by maximizing the intensity received at the alignment meter diode. Although the manufacturer indicated that the receiving lense should be removed, it was necessary for the receiving lense to be in place to achieve any reading on the alignment meter.

The alignment procedure for the receiver section was also adequately detailed in the operator's manual. This section contained the receiving lense and its mount, the photodiode array, and a printed circuit board interfacing with the computer. Only minor difficulties were encountered in achieving the Malvern specification for background counts of 10 or less on all elements of the photodiode array for the 63-mm and 100-mm lenses. In one case, a light cover over the photodiode did not fit properly due to a lack of clearance with the housing and adjusting screws on lenses and mounts. The 300-mm lense also suffered from stray light scattering onto the ring nearest the center hole. The stray light resulted in a background higher than specified in the manual.

Once properly aligned, the instrument alignment was only sensitive to large changes in room temperature and stresses on the frame or supporting surface. These two conditions produced a minor misalignment which usually appeared as a drop of 0-30 percent in the transmitted intensity as measured on the alignment meter. External mechanical controls for horizontal and vertical alignment of the photodiode array were sufficient to realign the system from such effects. These external controls also provided for simple day-to-day maximization of alignment or alignment correction from one run to another.

Computer Control and Software

Particle sizer functions, data analysis, and output of information were all controlled from the operating system contained in software on the integral Commodore computer. This program was recorded on cassette tape as a line interpretive BASIC written operating system and a data reduction and control program written in 6502 assembler language. The BASIC program, which executed printouts and command line recognition, was accessible to the user for desired modifications. However, a listing of the proprietary assembler language program controlling sizer functions and performing data reduction was available only through Malvern Instruments.

Control of sizer functions, through a series of two-letter command symbols, was straightforward and simple. The instrument could be controlled manually, one command line at a time, to critically examine process results while the "GO" command was quite useful to repeat a block of instructions for batch processing. However, no decision making or branching instructions were available within the "GO" command loop, limiting its usefulness to well-defined tasks.

Standard program and data storage for the computer was through the use of cassette tapes and a recorder. However, an optional floppy disk based system was available. The cassette system not only retrieved and stored data slowly, but tape positioning was a user responsibility. Only sequential access was available and the computer began a search or started writing wherever the tape was positioned. No internal checks were performed to determine if the tape head was in the middle of existing files or to find the first vacant file. The computer did not recognize the end of tape condition and continued reading or writing operations off the end of the tape. Termination of this error with a run/stop command when outputting data resulted in improperly closed files.

Sample Cells

The mechanical operation of the sizer was not managed by the software. The user selected which sample cell or measurement technique to use and was responsible for proper optical alignment. In addition to normal optical alignment procedures, additional alignment was required to compensate for the effects of windows in sample cells. All of the results discussed below were obtained with the aid of a sample cell; one of the two standard liquid cells available from Malvern (flow or stirred cells), or a specially designed aerosol cell.

All sample cells required at least two windows; one for the laser beam to enter and one for scattered light to exit. These windows introduced refraction of the transmitted beam which was compensated through the vertical and horizontal alignment of the receiver. Any scratch, imperfection, or deposit on the window surfaces, diffracted light and were interpreted as particles unless corrected for as background through the software. The current version of the Malvern flow and stirred cell design caused scratches at the inside shoulder of cell windows. The resulting scratches did not affect the transmitted beam of the stirred cell since they occurred only at the edge of the windows but could affect the beam from the flow cell which has a large oblong shoulder (see Figures 2 and 3). Care was taken to return the window to the original orientation in the flow cell and to not switch windows between cells. A simple cleaning procedure of single passes of a methanol-soaked lense tissue was satisfactory to remove most deposits. Loose dust particles were removed with "dry air" or a lense-cleaning bulb.

Some effort was extended to maintain optical alignment when the sample cells were removed and replaced. Fortunately, the cell mounts on the 2200/3300 model were sufficiently rigid that removal and replacement of the sample cells resulted in an optical misalignment that could be corrected by the external aligning screws. Changes in optical alignment were usually small when the windows were replaced in their original positions. However, reversal of the cell resulted in a significant degradation of the optical alignment. In general, even severe misalignment could be compensated with the available external adjustment.

FIGURE 2. STIRRED SAMPLE CELL

FIGURE 3. FLOW SAMPLE CELL

Flow Cell -- The flow sample cell illustrated in Figure 3 was used with solid or gas particles suspended in a fluid. This factory-supplied cell had a 5-mm beam path for particle analysis. The bulk sample was held in an external, stirred ultrasonic tank. A centrifugal pump was used to continuously circulate some of the bulk sample through the cell. A valving system supplied with the Model 2200/3300 sizer allowed the bulk tank to circulate samples to the cell to be drained, or to be filled with a fluid from an external source. An in-line filter was necessary to maintain acceptable background levels when using tap water as the suspension fluid. Adequate filtration was obtained with an Aqua Guard water filter equipped with taste and odor filters to remove particles larger than 5 μm diameter.

An unregulated water level in the bulk tank provided an annoying problem during filling, draining, and flushing. Occasional fluctuations from a constant input/output rate resulted in the tank either overflowing onto the instrument and the floor or draining completely (in which case the pump had to be reprimed). The factory-supplied valving system also contained plastic tygon tubing that was slipped over smooth connections. The potential existed for these unclamped connections could separate inside the instrument case.

During operation, the suspension liquid within the bulk tank was continuously circulated through the flow cell. Background counts due to the suspension fluid and artifacts created by the lenses or windows were determined prior to the introduction of sample to the liquid. Following the addition of the sample (see Sample Preparation), a set of size determinations was performed and, subsequently, corrected for the predetermined background count in the liquid.

Two primary problems were observed with the circulation of particles through the flow cell: adherence of particles to tubing walls and particle sedimentation. Adherence of particles and particle residues to tubing walls led to the formation of a layer of deposits on the inside surface of the tubing. Eventually, this film became concentrated enough that it began to flake off the tube walls and cause increases in the background counts. The Malvern handbook did not address at what point this buildup would affect measurements nor criteria for the cleaning or replacement of the tubing. This problem was managed by occasional cleaning of the tubing.

Particles of sufficient size tended to quickly settle out of the suspension. In order to keep the larger particles suspended, pump and mixer speeds were necessarily increased to levels which, in some cases, caused cavitation in the suspension fluid. Particle sedimentation was enhanced by the presence of a dead zone at the center of the bulk tank where little or no mixing occurred. This settling was dependent on the relative densities of the suspension fluid and the sample. The use of an alternate fluid such as glycerin would minimize settling due to the reduced differences in density, but the pump's ability to circulate such a fluid was not determined. (In routine analysis, the use of fluids other than something like water or isoton is probably not practical.)

For materials used in this study with water as the suspension fluid, results indicated that particles smaller than 50 μm diameter could be accurately characterized using the flow cell. Analysis of particles larger than 50 μm diameter tended to be in error due to sedimentation of the larger particles.

In addition, removal of particles from the tank, cell, and lines subsequent to analysis required significantly more effort for particles larger than 50 μm diameter.

Stirred Cell -- An illustration of the factory-supplied, stirred sample cell is presented in Figure 2. This cell was also used with solid particles suspended in a liquid. It had an illuminated length of 14.3 mm and a magnetic stirrer in the bottom of the cell. The cell mounted over a magnetic driver which turned a stirring element, keeping the particles suspended and well stirred. This was in contrast to a continuous flow of sample-laden fluid in the flow cell.

This stirred cell required a much smaller sample volume than the flow cell (about 100 ml versus 2 l, respectively). All particles ($< 150 \mu\text{m}$ diameter) were found to stay well mixed even with the magnetic stirrer at its lowest speed. However, large particles did occasionally stop the stirring element. The cell was easily cleaned since it could be physically removed from the instrument and cleaned by hand between runs. Since the two sides of the cell were not optically equivalent, reversal of the cell orientation resulted in a significant misalignment condition. This misalignment was easily observed on the alignment meter but could, in error, be attributed to a sample in applications. Failure to realign the instrument resulted in erroneous measurements.

The stirred cell was tested with the full range of particle sizes utilized in this study and its results agreed well with those of the flow cell and Coulter counter. The mean particle size was observed to decrease in size with the length of time in the cell for only one sample (18-inch fluidized-bed material). This size decrease was attributed to grinding of the particles by the magnetic stirrer.

Aerosol Cell -- A simple aerosol cell was designed for studying aerosol particles in the laboratory (see Figure 4). The cell design was based on criteria for minimum sample concentration and necessary illuminated path length for an assumed 1 gm/m³ source concentration. Experiments with this cell indicated that the design criteria was adequate to manufacture a working cell, but that a more sophisticated design is required to prevent particle deposition on the cell windows. These tests identified important design considerations and gave an indication of the Malvern's sensitivity to window fouling.

Repeatable, dry aerosols were generated by suspending commercially manufactured glass beads (Potter Industries) with a fluidized-bed aerosol generator (TSI Model 3400). Glass beads in the stated size range of 5-20 μm diameter (MMAD of 14.2 μm as determined by the Coulter counter) were employed as the known sample. Initially, aerosol particles were fed into the cell through a port near one window and extracted from a second port near the opposite window. This introduction technique resulted in the input window becoming covered with particles and unusable for sizing with the Malvern in a very short time.

To minimize aerosol deposition on the cell window near the inlet, a simple flat baffle was introduced to deflect incoming particles. Although the baffle resulted in improved data collection intervals, it also created a recirculating air pattern that caused rapid buildup of particles on sections of the

FIGURE 4 . AEROSOL CELL

windows. A curved baffle was finally used to reduce the vorticity behind the baffle. The resultant window deposition was reduced to rates that allowed the suspended glass beads to be characterized, but for short intervals. The attempt to transit relatively large size aerosols over a significant horizontal distance was necessary in order to continue to utilize the available optical configuration of the Malvern. A vertical flow configuration, where particle deposition and sedimentation would be minimized, is preferred and recommended.

Sample Preparation

The solid particles used in this study were available in either bulk quantities of dry dust or slurries (suspended particles in a fluid). When characterizing dry dust, the particles were dispersed in a liquid by a stirring element and occasionally also by sonication. Since the Malvern particle sizer results were to be compared with those of the Coulter counter, identical sample preparation techniques were used.

The procedure for sample preparation consisted of: (1) collection of a small volume of particles from random locations in a well-mixed sample (the amount depended on sample volume requirements and particle concentration but was usually less than 5 mg); (2) dispersion of the small volume of particles in a 30-ml sample bottle containing isoton only or an isoton plus a dispersant (Tamol); and, (3) sonication of the entire sample bottle for 30 seconds while the contents were stirred continuously. The sample was then transferred with a medicine dropper to the desired sample cell for analysis. The remainder of the sample was stirred in the sample bottle frequently to insure a uniform sample dispersion.

DATA AND RESULTS

This study was limited to particle sizes in the range of 5-150 μm , although the instrument is designed to measure this size as well as much larger particles. The selection of this size range was based on the availability of well characterized ground coal and standard samples. The particulate materials tested in this study are listed in Table 1. In addition to a broad size range, the particles also included a range of dielectric constants and shapes. The instrument did not consider the refractive index of the sample or the background medium. However, the manufacturer was attempting to incorporate these two parameters in software for the system. A systematic error due to refractive index was not observed.

Comparison With Coulter Particle Counter

A Coulter counter was employed in this study as a reference for comparison or confirmation of the Malvern analysis. The Coulter counter was chosen as the reference because it is commonly used as a laboratory service tool to size particles. Samples for analysis were prepared with identical or nearly identical methods for the Malvern and the Coulter. With the exception of the use of the aerosol cell on the Malvern, all samples were suspended in a fluid for analysis. Separate operators ran the Malvern and the Coulter analysis. A comparison of the Malvern characteristic size parameter, $\bar{X}$, with the median size parameter from the Coulter counter is presented in Figure 5 for a number of samples. In general, the agreement of results was quite good. For samples from the same large batch, but not necessarily the same test sample, the measured size values for the two units are usually within $\pm 2 \mu\text{m}$ of each other for particles in the smaller end of the size range. For larger size particles (approximately 150 μm), the difference was generally within 5-10 μm . Therefore, the relative error due to both sampling technique and instrument differences over this range of particle sizes was usually 25 percent or less. When identically the same test samples were run on both instruments, the agreement was better. The inherent error of the analytic technique has been reported to be 2 percent (Swithenbank, Beer, 1976). Error analysis of the instrument itself, from studies utilizing a photo mask manufactured to be a known Rosin-Rammler distribution, have reported a 10 percent accuracy for the system (Hirleman, 1982).

Distribution shape analysis was not attempted since most samples fell primarily into the smaller size ranges for each lense. However, two results dependent on the shape of the distribution were observed. First, a very narrow size distribution could not be properly fit with software models. Attempts to analyze samples of monodisperse particles (i.e., polystyrene latex spheres) produced erroneous and erratic results. Also, when presented with a bimodel particle size distribution, the three available built-in distribution functions (Normal, Log-Normal, Rosin-Rammler) provided poor fits to the experimental data (as indicated by large log error values). Bimodel and other multimodel distributions were analyzed on the Malvern system using a model independent approach. This model independent fit provided a weight of the distribution in each size band but did not give a characteristic size. Using either the model independent or one of the three internal distribution functions, the Malvern system was not sensitive to a small number of large particles. This was inconsistent with the expected behavior of a weight averaging system.

COMPARISON MALVERN SYSTEM TO COULTER

FIGURE 5. COMPARISON OF MEASURED PARTICLE SIZES BETWEEN MALVERN AND COULTER COUNTER.

TABLE 1

List of Particulate Materials Tested

Sample Identification	Comments	Probable Constituents
82-135	Coal Water Mixture (Waltermire) Grinding Test	Water Coal
82-138		
82-139		
82-140		
82-141	18-Inch Fluidized	Limestone
82-142	Bed Material	Coal Ash
82-143		
82-144		
82-145		
82-146		
82-147 (CH3342)		
82-155 (CH3361)	Loop Fluid Bed (Mei)	Sand Coal Ash
82-163 (CH3373)		
82-216	Coal Water	Water
82-217	Mixtures	Coal
82-218	(Carpenter)	
Potters Glass Beads 3-10 μm , 5-20 μm 10-40 μm	Potter's Industries	Type of Glass (Unknown)
Kaolinite Paper Mulberry Pollen Fort Martin Fly Ash		

Sample-To-Lense Distance Effects

Two types of controlled experiments were conducted to investigate the effects of sample-to-lense distance and sample concentration effects. Both the flow and stirred cells were employed to study the Malvern system response using the 63-, 100-, and 300-mm lenses. Measured particle sizes were not dependent upon the type of sample cell; therefore, the cells have been grouped together in this section to present the results of distance and concentration effects.

For tests of sample-to-lense distance effects, the sample cell was mounted on the optical bench at a minimum distance from the receiving lense and the full measurement procedure followed. A background sample was collected with a clean fluid, then a sample was added and experimental data collected. The cell was then moved to a new location on the optical bench further away from the receiving lense and the entire measurement procedure repeated. In this step-wise fashion, a series of independent measurements on the same test sample were collected which covered the range of travel available on the optical track.

The results of these tests for a stirred cell test utilizing the 63-mm lense are presented in Figure 6. Similar results for a flow cell test using the 100-mm lense are presented in Figure 7. For both lenses, with either the flow or stirred cell, a pronounced drop in the measured particle size was observed at specific locations along the optical track for the same particulate sample. No correlation to a reflection from the sample cell windows was observed.

The observed drop-off in measured particle size occurred at different positions for the two lenses and was correlated to a property of the receiving lense. No drop-off occurred in measurements made with the 300-mm lense over the accessible portion of the optical track. Figure 8 is a normalized graph of the $\bar{X}$ values, measured versus position, demonstrating these results for the stirred cell tests.

Since parallel light was emitted from the transmitter section of the Malvern, in theory, the distance of the sample from the receiving lense should not influence particle sizing measurements (Swithenbank, Beer, 1976). In addition, the focal length of the Fourier transforming lense should be important only in determining the spatial position of the Fraunhofer Diffraction pattern cast on the photodiode. However, these conditions must be modified by the predictions of scattering theory. For a distance from the receiving lense, given by

$$Z_c = \frac{f\pi d}{7.66 F\lambda}$$

there will be a significant amount of energy diffracted by particles of diameter (d) truncated by the transform lense (Hirleman, 1982). In this equation, F is the f-number of the transform lense and is given by the focal length (f) divided by the lense diameter. A sketch of this truncation process is presented in Figure 9. Physically this means, at the position of the receiving lense, the forward scattering lobe from the particle is larger than the lense opening, so a fraction of the scattering information was not received.

SAMPLE TO LENS DISTANCE TEST

FIGURE 6. OBSERVED EFFECTS OF DISTANCE FROM RECEIVING LENSE ON DATA (63 MM LENSE).

100MM LENS--SAMPLE DISTANCE TEST

FIGURE 7. OBSERVED EFFECTS OF DISTANCE FROM RECEIVING LENSE ON DATA (100 MM LENSE).

COMPARISON OF DROP-OFF LOCATIONS

FIGURE 8. OBSERVED EFFECTS OF DISTANCE FROM RECEIVING LENSE ON MEASURED PARTICLE SIZE FOR VARIOUS RECEIVING LENSES.

FIGURE 9. ILLUSTRATION OF LOSS OF SCATTERING INFORMATION
RESULTING FROM EXCESSIVE DISTANCE FROM RECEIVING LENSE

For the approximately 35-mm diameter, 63-mm lense, the equation above predicted significant losses at 9.3 cm while it predicted losses at 23 cm for the roughly 55-mm diameter, 100-mm lense if $6.5\text{ }\mu\text{m}$ (coal slurry) particles were considered. The falloff occurred within these distances for both cases above, although the signal from the smallest particles in the distribution (below $6.5\text{ }\mu\text{m}$) was affected before the predicted critical point was reached. Further, the Malvern result appeared, at first, to contradict this expected loss of small-particle signal by dropping to smaller sizes with increasing distance.

The drop in size was not caused by a loss of scattered light from large particles. Analysis of the light energy measured and the signal collected during the tests indicated that the loss of scattering information from small particles caused the observed behavior. It appears that the abrupt drop was associated with the nonlinear least squares data fitting routine acting upon an incomplete light scattering data set.

The results of a series of sample-to-lense distance experiments, which traverse the region of the abrupt drop, are presented in Figure 10. The actual light intensity measured on the diode array is shown on the left and the corresponding model-independent analysis which determined the weight observed in each size band is shown on the right. In this figure, the sample distance from the lense is increasing from bottom to top. Since the scattering angle is inversely proportional to particle size, the light intensity plots indicate particle size decreasing left to right. However, the model-independent plots on the right are corrected to give size increasing left to right. Note that the light intensity plots are all very similar and each shows that the size distribution of the particulate sample corresponded to the smaller sizes observable with the receiving lense. Consideration of the rightmost bars (smallest particles) in the intensity series indicated the loss of signal with increasing distance as expected. The intensity received from the smallest particles was reduced first, then the next larger size began to be affected, while the smallest was still further reduced. The abrupt drop in measured particle size occurred between plots numbered 5 and 6 in the series (Figure 10). From the series of model-independent analyses, it was observed that the percent weight/size in the predicted distribution representing larger particles was increased with distance from the lense corresponding to the small particle signal loss.

At the start of signal loss, the least squares routine would still achieve the best fit by retaining the distribution peak at approximately the correct particle size. When the signal corresponding to the light scattered by smaller particle was reduced past a certain critical point, the fitting routine abandoned the attempt to match distribution peaks. It achieved a better fit by moving the distribution peak to a very small size value in such a way that the actual data were fit by one decreasing side of the predicted size distribution. As the signal from additional small particles was lost, it is not difficult to visualize from Figure 10 that an apparent distribution peak was clearly generated and the least squares routine responded by increasing the predicted size to match this peak and achieve the lowest error. This behavior is apparent in Figure 6 and Figure 7 where, after the critical point, the measured size began to increase again toward the correct value.

(7)

(6)

(5)

(4)

(3)

(2)

(1)

DISTANCE FROM LENSE INCREASING

FIGURE 10. LIGHT INTENSITY MEASURED ON DIODE ARRAY (LEFT) AND RESULTS OF MODEL INDEPENDENT ANALYSIS (RIGHT) AS A FUNCTION OF DISTANCE FROM RECEIVING LENSE.

The above explanation of observed behavior was corroborated by experiments with larger particles and the model dependent tendencies. For particles possessing a diameter roughly three times that of the coal slurry particles, there was no critical behavior. Although signal loss from small particles could be clearly observed, its affects on the predicted size were not significant. In this case, the distribution was centered on the 9th of the 15 size bands from the smallest rather than centered close to the size region where signal loss was occurring. The data fitting routine then had no difficulty achieving the best fit by matching distribution peaks and did return a correct size over the entire range of travel.

The process of signal loss from small particles with increasing distance is fundamental. Applications of the sizer to situations in which small particles are likely to be far from the receiving lense should be avoided. Where "small" in this situation is defined as corresponding to the smaller size bands as determined by the receiving lense.

The log error criteria was useful in identifying conditions involving loss of small particle scattering information. With the proper model distribution, an initial log error value could be achieved which indicated an exceptionally good fit (<5). This value was maintained as the distance of the cell from the receiver was increased until the transition point was reached. At this point, the log error value abruptly jumped to higher values. Normally, the new log error value at transition was sufficiently high to indicate a poor fit and continued to indicate a poor fit well past the transition. There was a tendency for the log error value to indicate an improving fit at distances well past the transition point, but this was not significant. Occasionally log error values were still in the handbook's acceptable fit range even at or beyond the transition point. As a result, it is advisable to maintain the sample cell position close to the receiving lense and develop independent standards of acceptable data fits.

Sample Concentration Effects

The experiments on the effects of sample concentration were more routine. The aim of this investigation was two-fold: To relate effects on the measured parameters to sample concentration; and to establish an estimate for the maximum and minimum concentrations which may be analyzed. Effects on the measured parameters were examined for all cells and lenses by analyzing a sequence of controlled sample concentration tests. Sample concentrations producing from 5 to 100 percent attenuation of the laser intensity were studied. The tests began with a clean background fluid in the cell, then sufficient sample was added to decrease the transmitted intensity in steps of 5 percent. The alignment meter was adequate in determining the intensity steps. At each concentration, experimental data was collected and analyzed using the Rosin-Rammler distribution. Using this procedure, particles from previous measurements were present to help maintain sample consistency. The initial background measurement was used for each entire series of data.

The results for all cells and lenses were similar to those shown in Figure 11. These results are from flow cell measurements utilizing the 100-mm lens. In the recommended range (0 to 50 percent attenuation), the measured size parameter, $\bar{X}$, remained fairly constant as did the distribution width parameter, N (not shown). Although the log error remained essentially constant over this range, there was a tendency for both parameters to decrease slightly. Outside

100MM LENS--SAMPLE CONCENTRATION TEST

FIGURE 11. OBSERVED EFFECTS OF LASER BEAM ATTENUATION ON DATA.

the recommended range (between 50 and 100 percent attenuation), there was a very steady drop-off in both the size and distribution width parameters. However, the log error values remained constant until extreme concentrations caused them to increase sharply. It was common to observe a 50 to 100 percent difference in size measurements between the dilute and concentrated samples which did not affect the log error value. In fact, log errors indicating exceptionally good fits could be obtained at all but the highest attenuation positions even though there was a substantial change in the measured size.

This behavior was attributed to the sample concentration reaching the point where multiple scattering effects became important. The relatively uniform distribution of scattered light produced at the photodiode array by such a process was equivalent to that produced by a sample of small particles. The sizer was unable to distinguish the two conditions and proceeded to compute small-size parameters. Apparently, the sizer's log error fitting criteria was insensitive to multiple scattering. Restrict the range of concentrations to avoid this effect.

As a result of the inability to detect erroneous data at high-particle concentrations, potential measurement errors may be avoided by restricting the range of concentrations. The recommended 0 to 50 percent attenuation range appears to be adequate.

A second goal of these tests was to estimate the minimum and maximum concentration requirements of the Malvern system. The tests were conducted in the flow cell using 5 to 20 μm diameter glass beads. Sufficient quantities of the pre-sized glass beads were added to the sample cell to produce a 50 percent attenuation of the transmitted beam. A second data set was obtained at the minimum attenuation which would still yield acceptable size results. The mass of particles contained in the cell was obtained gravimetrically from filter samples of a known volume of sample. A density determination on the glass beads, conducted with a Micromeritics Auto Pycnometer 1320 combined with the known mean size and measured total particle mass, permitted a number density to be determined.

At 50 percent attenuation, the instrument recorded a sample concentration by volume of 0.0556 percent. From the mean size, this corresponded to approximately 3.7×10^5 particles per cubic centimeter. The experimentally determined concentration was 1.7×10^5 particles per cm^3 . A near minimum concentration, which still yielded accurate results, was obtained at 0.0002 percent concentration by volume, corresponding to a recorded obscuration of 0.00. This corresponded to 1.3×10^3 particles per cm^3 while the experimental value was 2.7×10^3 per cm^3 .

The maximum and minimum concentrations necessary for Malvern 2200/3300 analysis may be estimated through the equations (Malvern Scientific Corporation No. 2):

$$\pi d^2 N_m L_p = 0.5 \quad (\text{Maximum Concentration})$$

$$\pi d^2 N_m L_p = 0.005 \quad (\text{Minimum Concentration}).$$

In these equations, N_m is the particle concentration (particles/ cm^3), d is an estimated average diameter, and L_p is the path length along the laser beam over which particles are present (5 mm for the flow cell). For 14.2 diameter μm

glass beads, these equations yield a maximum concentration of 1.58×10^5 particles per cm^3 and a minimum of 1.58×10^3 particles per cm^3 . These results agree well with those determined experimentally.

Aerosol Cell

In addition to the characterization of particles suspended in liquids, tests were performed to evaluate the Malvern for the characterization of aerosols. Initially, the output of a TSI fluidized-bed aerosol generator (Model 3400) was sprayed from an open air jet into the sensing volume of the Malvern. Under these conditions, the total number of particles in the sensing volume was insufficient for an accurate size characterization. An aerosol cell was constructed which increased the total particle concentration in the sensing volume by increasing the path length. The simple design of the cell (see Figure 4) was based on criteria provided in technical Bulletin No's. 4 and 5 from Malvern Instruments (Malvern Scientific Corporation No. 2). These criteria were found to be adequate for construction of a working cell.

However, the Malvern was very sensitive to any small deposits on the windows of this cell. The lack of a window cleaning device resulted in window fouling being a major problem of this simple cell design. The rate at which window fouling occurred could be slowed, but not eliminated. Deposition on the windows was recorded by the instrument as a decrease in the transmitted beam intensity.

Figure 12 is a plot of measured size versus time of operation for the aerosol cell. Initially, size measurements of 5 to 20 μm glass beads in the aerosol cell produced a result within ± 2 microns of the Coulter result. This comparison was similar to those using the flow and stirred cells, indicating that the liquid suspension data may be extrapolated to aerosols. The increase in measured size of the particles with time was attributed to the aerosol cell. The increase in size was probably due to the overlap of particles on the windows.

The Rosin-Rammler and Log-Normal distributions fit the aerosol data about equally well, according to log error criteria. The results of both distributions are shown in Figure 12. Although there was a considerable size difference between the results of the two models, both distributions indicated a strong correlation between an apparent increase in size and the running time. Although not shown, the width parameter, N , of both functions could be interpreted as indicating a broadening of the distribution.

The instrument's sensitivity to window deposits will require the use of purge gas to minimize window contamination in applications to aerosol studies. Current techniques of applying purge gas to optical access ports are probably inadequate to allow long-term operation. The development of innovative designs of optical access ports will require additional research.

Software Tests

Software supplied with the Malvern contained three model dependent distributions and a program utilizing a model independent approach. The three model dependent distributions (two-parameter functions) were the Normal, Log-Normal, and Rosin-Rammler functions. A model independent approach determines the relative sample weight in each of 15 size bands and can be used to detect

AEROSOL CELL-FIRST TIME TEST

FIGURE 12. OBSERVED EFFECTS ON DATA OF DEPOSITION OF PARTICLES ON AEROSOL CELL WINDOWS.

mono- or multi-modal size distributions. The model independent software does not provide calculated size information. In general, the decision to use one of the two-parameter functions was based on the smallest respective log error value.

Applications of all three two-parameter functions to identical data sets are presented in Figures 13 and 14. For the coal slurry characterized in Figure 13, the Rosin-Rammler distribution provided the best fit as compared to Coulter data. In spite of the small log error value associated with the Log-Normal distribution. The error value of both distributions was below the recommended threshold of 5. The Log-Normal distribution provided the best fit to the data for glass beads presented in Figure 14. Table 2 lists the three two-parameter distributions with a brief description of their likely area of applicability to various particle types and some experimentally observed tendencies.

As a general rule, the model giving the lowest error corresponded to the most accurate size result. As a matter of procedure, all three distributions were used to fit data and the model producing the lowest log error value in the analysis was used. In some situations, two models produced similar log errors but substantially different size determinations. Additional information is required in these circumstances to select the correct answer. Also, the log error value was not sensitive to sample concentrations. That is, for high concentrations producing an attenuation of greater than 50 percent, the determined size might actually be in error while the log error value indicated the fit to be accurate.

The ability of the software to use a specific distribution function to fit a particular data set was observed to be dependent upon the initial "guess" used in the program. In practice, the use of the computer parameters (CP) command, which relied on a computer-generated "guess" to initiate the fit, did not produce the same results as the input parameter (IP) command which required an operator "guess." Sufficiently incorrect inputs to the IP command could cause the fit to converge in the wrong direction. Figure 15 is a comparison plot showing the result of independently applying both CP and IP commands to sample distance data. In this figure there is a difference in $\bar{X}$ values of 0-2 percent and in width parameters of 0-3 percent. The log error values change from 2-20 percent. The percentage differences observed in $\bar{X}$ and N was indicative of the error associated with the stopping criteria of the nonlinear least squares analysis. Apparently, the software could be modified to reduce such differences.

TABLE 2

Available Model Distribution Functions and Observed Applicability

Distribution	Brief Description and General Suggestions for Particle Applicability	Experimentally Seemed to Provide Best Fit to
<u>Rosin-Rammler</u> $R = \exp(-d/x)^N$ $R = \text{Normalized Weight Below Size } d.$	<p>The Rosin-Rammler distribution was originally devised for modeling the size distribution of broken coal. Generally useful for materials such as sands, ores, clays; however, its most useful area of applicability for the Malvern system is probably in modeling the distribution of droplet sizes in various types of spray atomizers.</p>	<p>Coal Slurry Samples Coal-Water Mixtures 18-Inch Fluidized-Bed Particulates LFB</p>
<u>Log-Normal</u> $l_d = \frac{1}{\ln(N)\sqrt{2\pi}} \exp \frac{[(\ln(d) - \ln(X))]^2}{2 \ln(N)^2}$ $l_d = \text{Weight Frequency at Size } d.$	<p>The Log-Normal distribution is useful to model the size distribution of samples obtained by processes of comminution such as grinding, milling, etc. It has wide application to particulates from industrial processes. In addition, may be transformed to obtain area size distribution and number distribution.</p>	<p>Glass Bead Standard Samples Paper Mulberry Pollen</p>
<u>Normal</u> $n_d = \frac{1}{N\sqrt{2\pi}} \exp \frac{(d-X)^2}{2N^2}$ $n_d = \text{Indicates the Relative Frequency of Size } d.$	<p>The Normal is primarily used in modeling distributions by number rather than weight. However, it may be useful in some situations, particularly for narrow distributions.</p>	

COMPARISON ROS RAM-NORM-LOG NORM

FIGURE 13. EXAMPLES OF DECONVOLUTED DATA RESULTING FROM APPLICATION OF AVAILABLE MODELS TO IDENTICAL RAW DATA SET.

COMPARISON ROS RAM-NORM-LOG NORM

FIGURE 14. EXAMPLES OF DECONVOLUTED DATA RESULTING FROM APPLICATION OF AVAILABLE MODELS TO IDENTICAL RAW DATA SET.

IP-CP COMPARISON-LOG NORMAL-DIS. TEST

FIGURE 15. ILLUSTRATION OF DEPENDENCE OF DECONVOLUTED DATA ON INITIAL "GUESS" FOR ITERATION.

SUMMARY

In terms of general operation, the optical alignment of the Malvern was convenient for practical use. The alignment procedure was straightforward and misalignment conditions which occurred in normal use could be identified and easily corrected using external adjustments. Computer control of the instrument was satisfactory although the standard cassette-based system was slow and placed most of the magnetic storage responsibility on the user.

Both manufacturer-supplied sample cells examined in this study were observed to yield good size measurements (as compared to Coulter analysis); however, both possessed restrictions or limitations on their applicability. The flow cell was practical only for small particles where gravitational settling was not significant. The cell also suffered from fouling of the transport tubing. Particles up to the 150- μm diameter limit of this study could be examined utilizing the stirred cell, although particles this large occasionally stopped the magnetic stirrer. It was necessary to be consistent with the positioning of the stirred cell in the illuminating beam since its two sides were not optically equivalent.

The manufacturer's criteria for minimum concentrations and illuminated path length were found to be adequate to design and construct a working aerosol cell. However, the instrument was found to be very sensitive to window fouling. As a result, the simple test cell fabricated for the subject tests was impractical to use for extended measurements.

Comparisons of the measured size of various particulate samples indicated good agreement between the Malvern and a Coulter counter. In the smaller particle size ranges, the two systems usually agreed to within 2 μm (~15 μm diameter), increasing to 5-10 μm for the larger particle ranges of this study (~150 μm diameter).

In the sample-to-lense distance tests, a pronounced drop in the measured size value at a critical distance was observed for both sample cells with either the 63-mm or 100-mm lense. This fall off did not occur within the available travel of the optical bench for the 300-mm lense. The potential error from this effect can be eliminated by controlling the sample-to-lense distance during sample characterization.

In general, to prevent concentration effects, sample concentrations should be controlled to provide a maximum of 50 percent attenuation of the incident beam. For a path length of 0.5 cm (flow cell) and 14.2 μm diameter particles, the minimum concentration was a few thousand particles per cm^3 and the maximum concentration was several hundred thousand per cm^3 .

Software supplied with the unit contained three model-dependent distributions and a model-independent routine. In general, the distribution function which best fits available data could be determined by the minimum log error value associated with the fit of each of the three functions. The model-independent routine was particularly useful for identifying mono- or multi-modal size distributions which the model-dependent functions could not adequately handle.

This investigation necessarily exposed the weaknesses of the instrument. All particle sizing techniques and instruments have limitations or restrictions on their applicability. The Malvern system has demonstrated good agreement with the Coulter counter across the particle size range of this investigation (5-150 μm diameter). The versatility and simplicity of the instrument are strong points.

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