DETERMINING THE EFFECT OF TURBULENT SHEAR ON CONTAINMENT AEROSOL DYNAMICS USING MICROGRAVITY EXPERIMENTS

by

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ABSTRACT

Determining the characteristics of large aerosol aggregates "clusters" under turbulent conditions is fundamental for predicting the behaviour of radioactive aerosols inside the reactor containment following a severe accident. Studying such rapidly settling clusters is extremely difficult in ground-based experiments due to the effect of the earth's gravity. In this study, the microgravity environment is exploited to investigate the effect of turbulent shear on the aggregation and breakage of clusters by examining their structure and measuring their strength parameters while suspended under weightlessness conditions. A parametric model is introduced to correlate the experimental results over into nuclear aerosol models. It was demonstrated that the cluster parameters depend mainly on the turbulent field intensity as well as initial powder conditions.

1 INTRODUCTION

Three processes are considered important in assessing the consequences of a radioactivity release to the environment following a postulated severe reactor accident. These processes include the release of the radioactivity materials from the damaged core or primary heat transport system; the removal of radioactivity from the atmosphere inside the reactor containment by gravitational settling and containment dousing system; and the transport phenomena and the radiological impact to the environment.

The process of sedimentation due to gravity contributes significantly to the removal of radioactive aerosols from the reactor containment atmosphere. The faster the rate of aggregation, the quicker the clusters settle to the containment floor. On the other hand turbulence tends to break up the aerosol aggregates thereby reducing the sedimentation efficiency. Therefore, there is a particle life cycle involving:

Aggregation → fragmentation → aggregation → sedimentation.

Understanding the life cycle and the physics of aerosols under the containment conditions is very crucial for predicting their behaviour. Therefore, experiments were performed worldwide to study the behaviour of nuclear aerosols in closed containers[1]. Since it is impractical to perform such experiments on a scale of real containment, the experimental findings need to be scaled to actual conditions. This is usually done through mathematical models which use data from the experimental results to simulate the behaviour of aerosol under various boundary conditions. Great efforts have been devoted in recent years to develop, improve and validate the aerosol models for both containment and reactor cooling system[2,3,4,5].

Aerosol aggregation kinetics is given by the Smoluchowski's rate equation[6]. The aggregation process is due to three independent mechanisms: Brownian motion, gravitational settling and turbulent flow. The Brownian coagulation is due to random motion of the particles as a result of collision with gas molecules. The gravitational coagulation results from collision between particles of different size as the larger particles overtake the smaller ones due to differential settling speed. Two independent mechanisms exist. For particles smaller than the turbulent eddies and thus are entrained coagulation is enhanced by turbulent diffusion. For particles larger than the turbulent eddies, coagulation is caused by differential motion between different sized particles. This is usually called turbulent inertial mechanism[6].

Atlantic Nuclear conducted a study to measure the aerosol aggregation parameters under microgravity conditions[7,8]. It was shown that the Brownian aggregation is a fractal phenomena and that the aggregation kernel can be derived in terms of the fractal dimension. The fractal dimension was successfully measured for tin powder. This is a typical aerosol material that is likely to be released in a severe nuclear accident. The microgravity laboratory has proven to be a viable tool for growing large aggregate clusters and studying their fractal characteristics under controlled flow regimes. It was recommended that the fractal dimension be used to replace the empirical shape factors normally used in the Smoluchowski's rate equation thus eliminating a major source of uncertainties in calculating particle interaction probabilities (i.e collision matrix).

In the current study, the particle fragmentation part of the cycle is investigated. Since the atmosphere inside the reactor containment is regarded as turbulent under nearly all conditions, the aerosol dynamics behaviour can not be decoupled from the thermohydraulics of the containment building[1,4]. The aggregate clusters may break up under the influence of turbulent shear forces. Depending on the size of the aerosols they may be suspended in the containment for some time and eventually released through leakage paths to the environment. The breakdown of the particle clusters under vigorous turbulent conditions depends on the short range forces which hold the particles inside the cluster.

Detailed computational models which account for aerosol behaviour inside the reactor containment under typical accident conditions are complicated and require extensive computational time. Additionally using such models requires the availability of reliable experimental database. There have been recent attempts to develop simplified models by correlating the results from detailed models[9]. In the following section we will introduce a parametric particle aggregation/breakage model under turbulent conditions. This model can be used to quantify the strength of clusters under various turbulent intensities.

2 AGGREGATION IN TURBULENT FIELDS

A model was developed and tested by Leu and Ghosh for predicting cluster size distribution in agitated fields[10]. The model was meant for applications in waste-water treatment by flocculation. In deriving the particle's kinetic equation, Leu and Ghosh, assumed that the size distribution of the parent clusters is characterised by a statistical mean. All clusters of such mean size may not have the same strength and on exposure to a specific shear force, a certain fraction will break up. The fraction that will break up was assumed to be some cumulative probability function of the Gaussian shape. The rate of change of the average cluster size ($\frac{dD_{av}}{dt}$) under the combined effect of aggregation and break up in an agitated field is given by the following parametric first order differential equation[10]:

$$\frac{dD_{av}}{dt} = (4/3\pi)(\alpha-\acute{\alpha})\Omega G D_{av} - \frac{G^2 (D_{av})^{1+r}}{3\pi c} \qquad (1)$$

where

D_{av} is the statistical average of the cluster size,

G is the average rate of turbulent shear gradient,

α and $\acute{\alpha}$ are the collision efficiencies leading to aggregation and break up, respectively,

Ω is the volume fraction of parent clusters ($\Omega=(\pi/6)nD^3$), where n is the number of primary particles, and r and c are parameters that represent the cluster strength.

In the case of mechanical agitators, the average rate of turbulent shear, G, is calculated as a function of the agitator speed and torque on the main shaft using the following equation:

$$G=\frac{Tg_c\omega}{V} \qquad (2)$$

and

T is the torque on the shaft of the mechanical agitator,
g_c is the gravitational acceleration,
ω is the angular velocity of agitation blades $\omega=2\pi N_r$,
N_r is the rotational speed of the shaft in revolution per second, and
V is the volume of the system where turbulent energy is deposited.

In order to predict the cluster size distribution using the above model, the parameters, $\alpha,\ \acute{\alpha},\ r\ and\ c$, must be determined experimentally. In the original work of Leu and Ghosh, these parameters were determined by observing the evolution of particle size in a flocculation experiment. The experiment was conducted in a small vessel filled with water and fitted with a mechanical mixer driven by variable speed motor. Material similar to those used in water flocculation were used for cluster formation and breakage. Two non-dimensional parameters $r,\ c$ were determined by fitting the experimental data on cluster size as a function of fluid shear gradient. The following functional form was used for data fitting:

$$d_{\max} = c\ G^{(-\frac{1}{r})} \qquad (3)$$

where

$d_{\max}$ is the maximum cluster size that can be found in a specific turbulent field intensity.
G is the effective fluid shear gradient calculated from Equation (2).

The parameters $r\ \ and\ \ c$ along with data on the average size of clusters as a function of agitation time were then used to determine the parameter $(\alpha-\acute{\alpha})$ in Equation (1). To that end, a non-linear least square iterative method was employed.

3 ROLE OF GRAVITATIONAL POTENTIAL AND MOTIVATIONS FOR MICROGRAVITY EXPERIMENTS

Although buoyancy has assisted in suspending the clusters up to a certain size limit, the flocculation experiments performed in the laboratory, did not faithfully depict the conditions in large flocculation units. This was attributable to the effect of gravitational settling of large flocs in the laboratory's apparatus. In the case of airborne aerosol particles, the effect of gravitational settling will be more pronounced and it will be almost impossible to emulate the conditions in large volume systems such as reactor containment.

In this work we have exploited the access to the microgravity environment to suspended the clusters while investigating them using, for example, imaging techniques. The following are the motivations for performing the experiments in a microgravity laboratory:

1- The gravitational settling will hinder any attempt to examine the airborne clusters "aerosols" and subsequently measure their parameters under terrestrial conditions. This is due to the fact that buoyancy effects will be almost negligible as compared to the case of water flocs.

2. In developing the parametric model of equation (1), an important assumption was made that the net volume/mass of particles at any experimental run remains unchanged; that is no settling of clusters occurs during the experiment[10]. This can be ideally achieved under microgravity conditions for both aerosol clusters and water flocs.

3- Under microgravity conditions data can be obtained for a wide range of cluster sizes thus avoiding the undesired data extrapolation in field applications.

4. The disturbance of clusters during sampling (i.e. interactions with walls, filter surfaces etc.) Will be significantly reduced under microgravity conditions especially if optical/imaging methods are used.

The main objectives of the current study is to investigate the effect of turbulent shear forces on the breakage of rapidly settling aggregate clusters by examining their structure under microgravity conditions. To that end, the cluster structure parameters defined in this section will be measured for aerosol substance similar to those that are likely to be released in the containment atmosphere following an accident. A secondary objective of the study is to use the parametric model, introduced in the previous section to correlate the experimental parameters such that the experimental results can be carried over into containment analysis codes.

In order to achieve the above goals an apparatus was designed and built to be used in studying the cluster structure and dynamics under microgravity conditions. The design of the apparatus was optimized using the readily available models to simulate cluster growth/breakage under turbulent conditions. The apparatus was bench tested on the ground to ensure its functionality. Two experimental campaigns were carried out using the DC-9 parabolic flights at NASA Lewis Research Center. Data on particle aggregation/breakage dynamics were collected in flight using imaging and other sampling techniques. Post-flight data analysis was carried out to quantify the cluster strength and dynamics parameters.

The DC-9 microgravity experiments are described in the following section. In section (5), methods of post-flight data analysis along with experimental results are presented.

4 MICROGRAVITY EXPERIMENTS

Two flight campaigns were conducted at NASA Lewis DC-9 microgravity facility during the week of August 26, 1996 and the week of January 13, 1997. During the DC-9 flight, a weightlessness environment is generated which lasts for 20 seconds followed by 50 seconds of 2g conditions. Each low gravity, "0g" and high gravity "2g" period represents a parabolic manoeuver. An average of 45 parabolic trajectories are usually provided for the researchers aboard the DC-9 every day. Our experiment consisted of eight flight days; four days for each flight campaign.

The experimental apparatus, described in details in reference (11), consists of a closed loop fabricated from stainless steel tubes of 64 mm diameter. The aerosol is circulated inside the loop through a pair of ducted fans. The fan speeds were computer controlled. In order to determine the turbulent shear gradient at various fan speeds (see Equation (2)) the torque on the fan shafts was measured using a torque metre and a relationship between the fans rpm and torque was derived. The loop was leak tight to prevent the leakage of aerosol during the experiment. An observation cell in the loop has three ports. One port of the observation cell was covered with glass and was used for optical imaging of the particles using a microscope and a CCD camera. The other two ports were used for drawing calibrated aerosol samples using polycarbonate filters and a cascade impactor with five impaction stages. The observation cell was illuminated using a strobe light to prevent image blurring due to particle motion. The opening of the CCD camera shutter was synchronized with the firing of the strobe light using a control signal from the camera and a phase shifter electronic circuitry. Image acquisition was achieved using a video recorder connected to the CCD camera. Additionally, a frame grabber and a dedicate image computer was used to obtain digital images. The operation of the apparatus was semi-automated using a computer control system.

During the flight experiment, the aerosols aggregate under a controlled turbulent flow field before the beginning of the parabolic manoeuver. During the manoeuver, the aggregation and break up takes place during the 2g periods of the parabolas. Sampling is performed during the 0g intervals while particles are suspended under weightlessness conditions. The operator determines the beginning and end of image sampling and changes the sample filters and cascade impactor substrates after samples are drawn. Aerosol samples were collected under two conditions, namely constant agitation intensity and variable agitation intensity.

Materials similar to those encountered in nuclear aerosol applications were used in the study. This includes silica powder to depict concrete aerosol and tin powder to represent the heavy fission products which are likely to become airborne in the containment atmosphere following a severe accident. Two silica powder cuts were used during the first flight campaign with average size values of 2.9 and 5.9 microns, respectively. A mixture of silica and tin powder was used to charge the system during the second flight. The initial tin particles had an average size of 20 microns.

Computer simulation was carried out in order to determine the initial powder concentration so that aggregation and breakage of the aerosol cluster can take place during the parabolic cycles. To that end, the kinetic equation for particle aggregation under turbulent conditions "Smoluchowski's equation" was used to predict cluster size distribution under various turbulent conditions. The differential balance equations were solved using the traditional sectionalized method[12]. The integration was performed over time intervals similar to those encountered under parabolic flight conditions. The simulation results showed that the parameters determining the extent of turbulent aggregation are; the initial particle size distribution, initial particle concentration, agitation speed and aggregation time.

For microgravity experiments, the time window is fixed, therefore, cluster size distribution can be controlled by changing the initial powder distribution and/or agitation speed. For the ease of operation of the experiment, the apparatus was only once charged with an aerosol, powder of known concentration and size distribution. Subsequently, the agitation speeds which lead to measurable change in particle size distribution over the parabolic flight time window were determined from the simulation results. The methods of post-flight data analysis along with results and discussion are given below.

5 DATA ANALYSIS AND RESULTS

Three types of samples were collected during the flights, namely cascade impactor samples, filter samples as well as digital and analog particle images. Such samples were collected during the 0g stages of the parabolic trajectories while aerosols were suspended in front of the microscope and CCD camera. To that end, the loop fans stops and the operator allows two seconds for the residual turbulence to die out before sampling. During the 2g intervals "pull up" and 1g "level flight", the turbulent energy deposition was controlled by adjusting the mixing intensity through the fan speed control system. Therefore, the change in size distribution between the parabolas is due to aggregation and break up under the influence of the turbulent field.

The cascade impactor and filter sample data were used to quantify the particle mass concentration, cluster volume ratio and particle size distribution (using the cut off size values of the impactor stages). Such data was used to quantify the particle concentration and volume ratio at various experimental conditions. The image data was analysed to determine the maximum and average size distribution of clusters under a specific turbulent field intensity. The results obtained from post-flight data analysis are presented below along with comments on the observed trends.

5.1 Cascade Impactor and Filter Sample Results

The amount of particle mass deposited on the cascade impactor substrate and polycarbonate filters was determined using a 1.0 10^{-5} accuracy precision balance. Data on sampling flow rate and sampling time, which were recorded during the flight was used to determine the mass concentration of particles. The average cluster volume ratio was found to be 1.3 10^{-6} for silica powder and 4.3 10^{-5} for silica-tin powder mixture. The concentration of particles effectively in circulation inside the loop was 1.2 10^{5} particles per cubic centimetre (base powder). Such a particle concentration was sufficient to produce large size aggregates under the agitation intensities which were achievable by the loop fans. This was also in accordance with the simulation results obtained during the design optimization stages.

Figures (1) and (2) show the mass deposition distribution on the five cascade impactor stages and backup filter for two powder types, namely silica powder and silica-tin mixture, respectively. The horizontal axes represents the diameters of spheres which represent the particles at each cascade impactor stage cutoff, Equivalent Aerodynamic Diameter. The vertical axis represents the percentage of powder mass deposited in corresponding impactor stage.

Figure (1) shows that more than 25 % of the silica particles/clusters have a size larger than 20 microns. The results in Figure (2) indicate that the ratio of large size clusters formed of silica-tin particles (typically larger than 20 microns) is about 80%. These results demonstrate that large size clusters were formed during the flight stages.

5.2 Results from Image Data

The image frame acquisition rate was five frames per second leading to the acquisition of over 2,500 frames per flight day. In order to determine the particle size distribution in the image frames digital and analog images were grabbed and analysed using the MATROX™ and Impact Professional™ image processing software packages.

The objective of the image analysis was to measure the maximum as well as the statistical average of the particle size distribution in each frame. This data was used to calculate the cluster parameters as explained in section (2). To achieve this goal, image enhancement was performed by segmentation using threshold operations in order to reduce the effect of background illumination. The threshold value for gray level display was adjusted to identify the edges of the blobs thus facilitating the process of their counting. Additionally, edge detection processes were performed using the familiar Sobel edge detection filters. The thresholding and edge detection processes were performed in an iterative fashion until the images were crisp enough to provide adequate counting statistics. Subsequently, a blob counting process started using the blob analysis feature of the image processing software. The minimum and maximum limits for the blob size to be counted were set to 20 and 100,000 pixel, respectively. Such a wide range of blob size was chosen to cover all possible particle and cluster sizes in the image frames. Image calibration was performed to convert the particle size from pixel units to millimetre units using information on the operating optical field of view.

5.2.1 Cluster Strength Parameters

Figures (3) and (4) show the relationship between the cluster size and the turbulent shear gradient, G for the silica and silica-tin powder, respectively. The horizontal axis represents the parameter G as defined in Equation (2). The vertical axis represents the maximum cluster size. The solid line represents a linear least square fit for the log-log relationship between the cluster size and turbulent shear gradient.

From the least square data fit, the silica cluster parameters, r and c in Equation (1) were found to be 0.639 and 2339, respectively. These parameters were 2.09 and 607 for the silica-tin powder mixture..
It should be noted that the parameter c is proportional to the cluster strength. It is also reported in the literature that the slope of the curves in Figures (3) and (4) (that is $\frac{1}{r}$) determines the mode of cluster breakage, i.e. erosion

and splitting[13]. A slope of 0.5 or larger indicates that the dominant break up mode is splitting.

The parameter c for silica only powder is larger than the silica-tin powder mixture. This implies that the clusters formed in the former case are stronger than those formed in the latter case. As expected this is attributable to the difference in the initial powder size used in the two flight campaigns. The parameter $\frac{1}{r}$ is larger in the case of silica powder mixture thus indicating that cluster breakage by splitting is more dominant than in the case of silica-tin mixture. On the other hand, this parameter is close to 0.5 for silica-powder mixture thus suggesting that erosion and splitting are equally responsible for the breakage of such clusters.

Another factor which may have contributed to the observed difference in cluster strength between the two powder types is the fact that the clusters were generally larger in size during the second experiment. Such large size clusters are easier to break, i.e. weaker, than small clusters due to the increased irregularity in their geometrical shape. On the other hand, the small clusters are somewhat rounded in shape and mechanically stronger. Obviously, the strongest cluster is the cluster which is formed of a single particle.

In explaining the difference between the cluster strength for the two powder types, one should not underestimate the effect of surface charge in determining the ultimate strength of the clusters. This is entirely dependent on the material type in this experiment. To that end, materials of metallic origin are characterized by their larger surface charge thus resulting in a stronger bonds between the particles.

5.2.2 Cluster Dynamics Parameters

Figure (5) and (6) show a comparison between model prediction and experimental results for average size distribution versus agitation time. The vertical axis represents the cluster size in units of microns and the horizontal axis represents the accumulated agitation time, i.e. multiples of 2g time intervals. These are the time intervals during which the loop fans were operational and aggregation/breakage of clusters took place under turbulent aggregation. Figures (5) and (6) demonstrate that the experimental and model prediction results are in good agreement.

The parameter $\alpha - \acute{\alpha}$ were calculated using nonlinear least square fit for the experimental data and the parametric model (Equation (1)). See reference (14) for details on deriving a generic computational model for non-linear least square fit. The values of these parameters were $\underline{0.028}$ and $\underline{0.168}$ for silica and silica-tin powder mixture respectively.

The minimum turbulent shear required to break the clusters was determined using the conditions at equilibrium. That is the condition under which the rate of change of the average cluster size ($\frac{dD_{av}}{dt}$) is very small. From Equation (1). , the following equation is deduced under equilibrium conditions:

$$(4/3\pi)(\alpha - \acute{\alpha})\Omega G D_{av} = \frac{G^2 (D_{av})^{1+r}}{3\pi C} \qquad (4)$$

Assuming that k is substituted for all the experimental parameters, Equation (4) becomes:

$$G D_{av}^{r} = k \qquad (5)$$

Figures (7) and (8) show the relationship between G and D_{av} using the experimental parameters for both types of powder.

For the sake of comparison, we calculated the minimum shear gradient required to break up the silica powder clusters and the silica-tin clusters. Such calculations were performed using Equation (4) for an initial cluster size of 8 microns. It was found that a minimum shear gradient of 819.6 s^{-1} is required to break up the silica-only clusters. In the case of silica-tin powder, the minimum turbulent shear for break up was 2733 s^{-1}. It can be concluded that if clusters can grow to a specific size, those which are formed of silica-tin particle mixture are stronger than the clusters formed of silica-only. Again this could be attributable to the fact that tin particles, being of metallic origin, are characterized by more surface charge than the silica powder.

It should be noted that data of the type presented in Figures (7) and (8) may be used as engineering guidelines for optimizing the turbulent conditions inside the reactor containment in such a way to minimize the breakage of clusters thus resulting in growing larger clusters by turbulent aggregation. These large clusters will quickly settle on the floor or other structural materials before they leak out to the environment through the deficient containment structure in case of a severe nuclear accident as explained in section (1).

Understanding the of aggregation - fragmentation -sedimentation cycle is also of great interest in many other scientific and technological applications. This includes polymers and powder technology, purification of drinking water, mitigation of air and soil pollution, prediction of the earth's climate as well as other medical applications.

6 CONCLUSIONS

The process of aggregation and fragmentation of aerosol clusters under turbulent conditions was investigated in a microgravity laboratory. Aerosol sampling under microgravity conditions has enabled the examination of large size clusters while they were suspended under the weightlessness conditions. Such rapidly settling clusters are extremely difficult to examine in ground-based experiments due to the effect of gravitational settling.

Two flight campaigns were conducted at NASA Lewis DC-9 microgravity facility. The experimental apparatus was robust and highly reliable during the two experimental campaigns.

The mechanical strength parameters of clusters formed with aerosol powder were measured using post-flight data analysis of the flight samples. Other parameters which determine the ratio of aggregation to breakage were also measured under various experimental conditions. Additionally, the minimum turbulent shear force required to break up the clusters were quantified for the two powder types. It was found that the magnitudes of such parameters depend mainly on the turbulent field intensity as well as the initial powder size, type and mass concentration.

The cluster dynamics parameters obtained in this study may represent an engineering database for mitigating the radioactive aerosols from the containment atmosphere by natural gravitational settling. Similar data could also be generated for process optimization in several other engineering/environmental applications which are based on particle aggregation physics.

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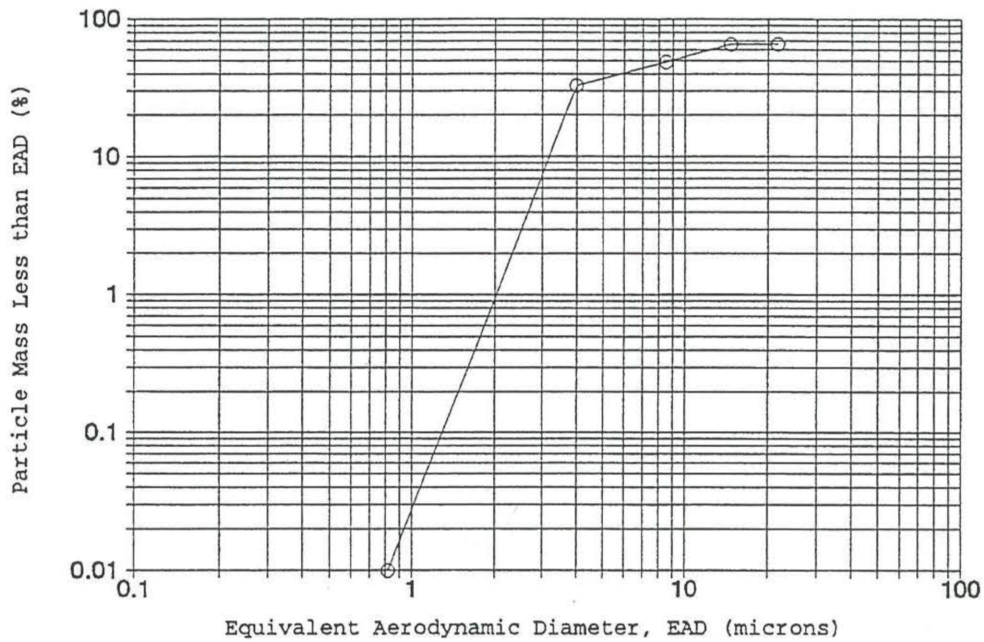

Figure (1): Mass deposition distribution, results for silica powder.

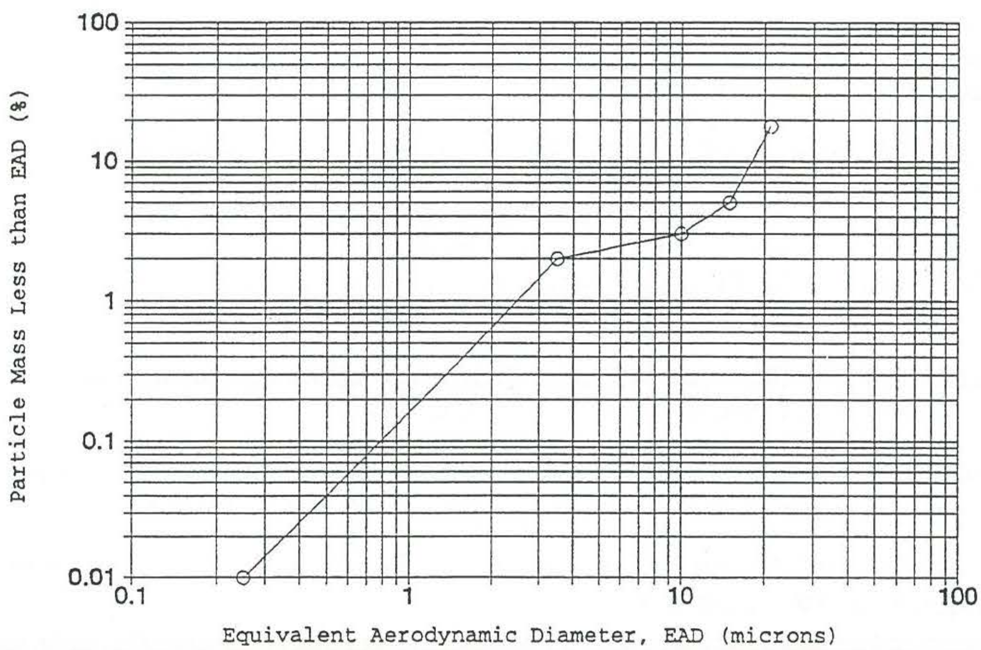

Figure (2): Mass deposition distribution, results for silica-tin powder mixture.

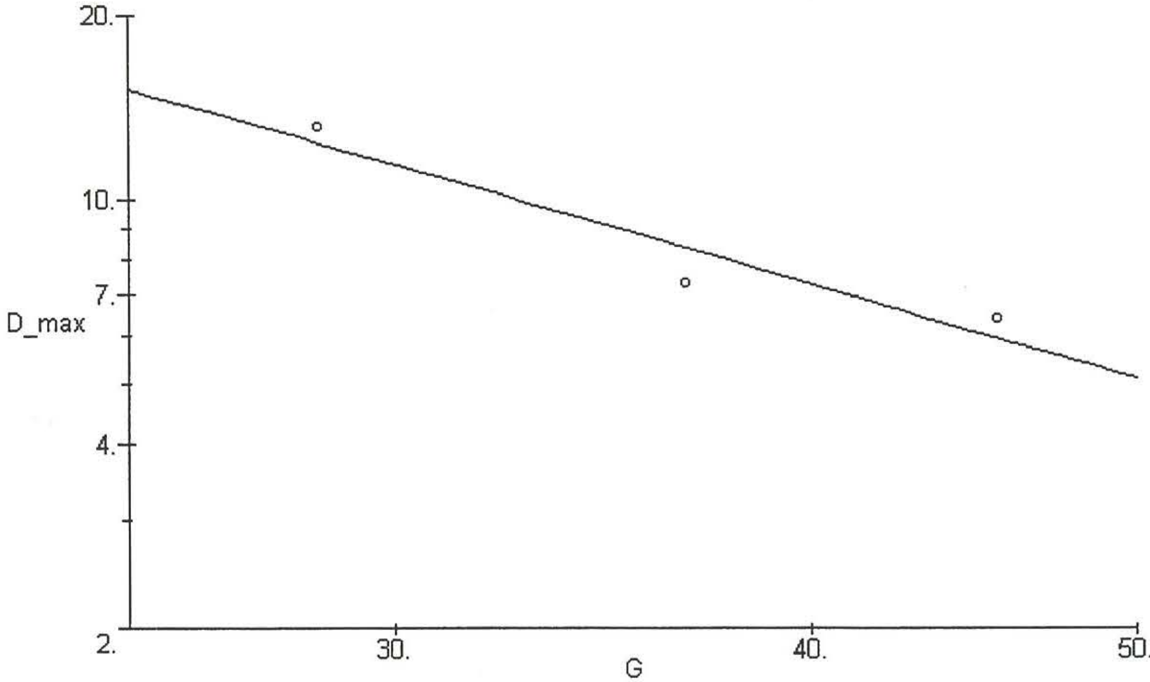

Figure (3): Maximum cluster size, microns, versus turbulent shear gradient, s^{-1}. Results for silica powder.

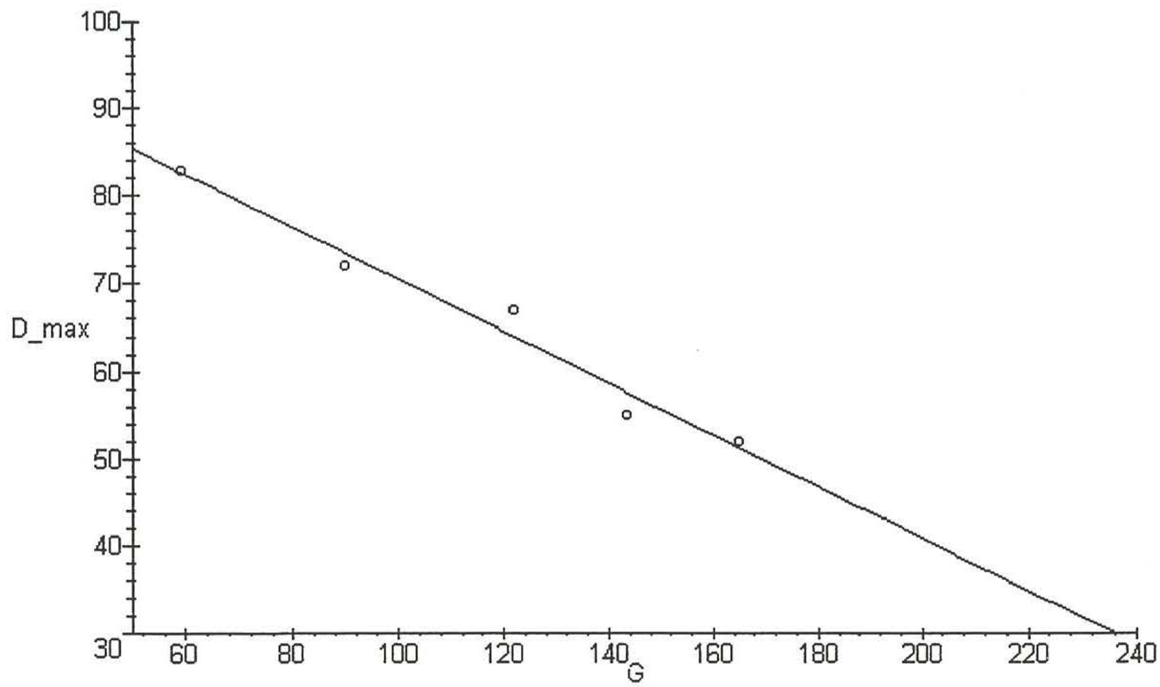

Figure (4): Maximum cluster size, microns, versus turbulent shear gradient, s^{-1}. Results for silica-tin powder mixture.

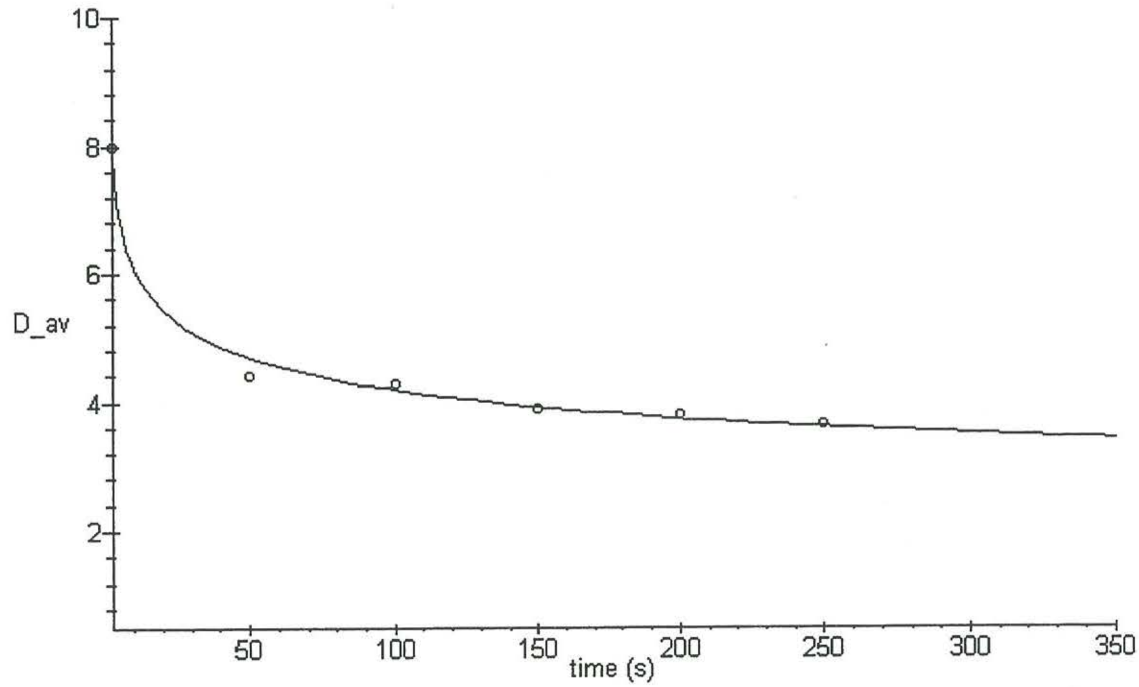

Figure (5): Average cluster size, microns, versus accumulated agitation time. Results for silica powder.

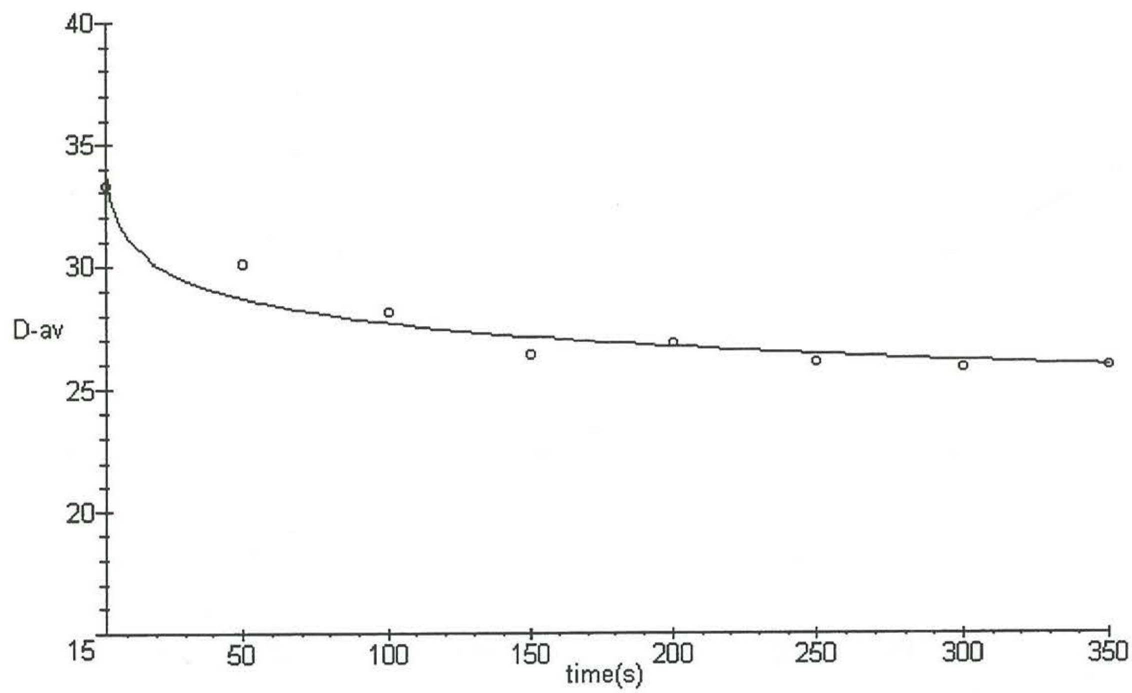

Figure (6): Average cluster size, microns, versus accumulated agitation time. Results for silica-tin powder mixture.

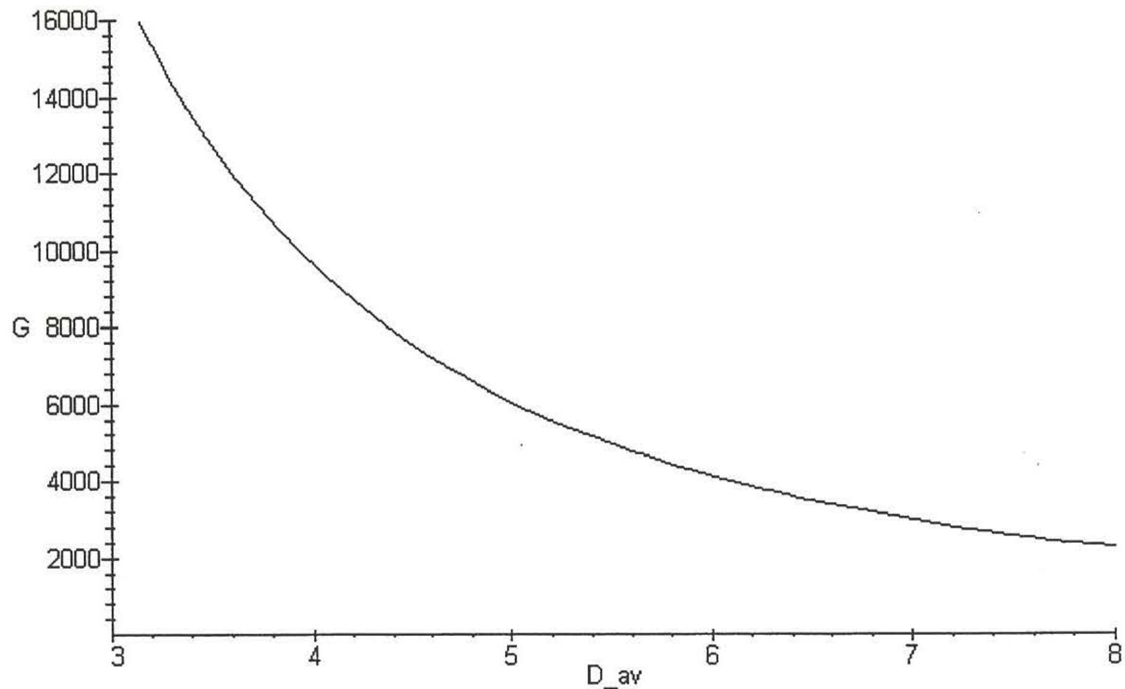

Figure (7): Minimum turbulent shear gradient required to break up the clusters. Base powder is silica.

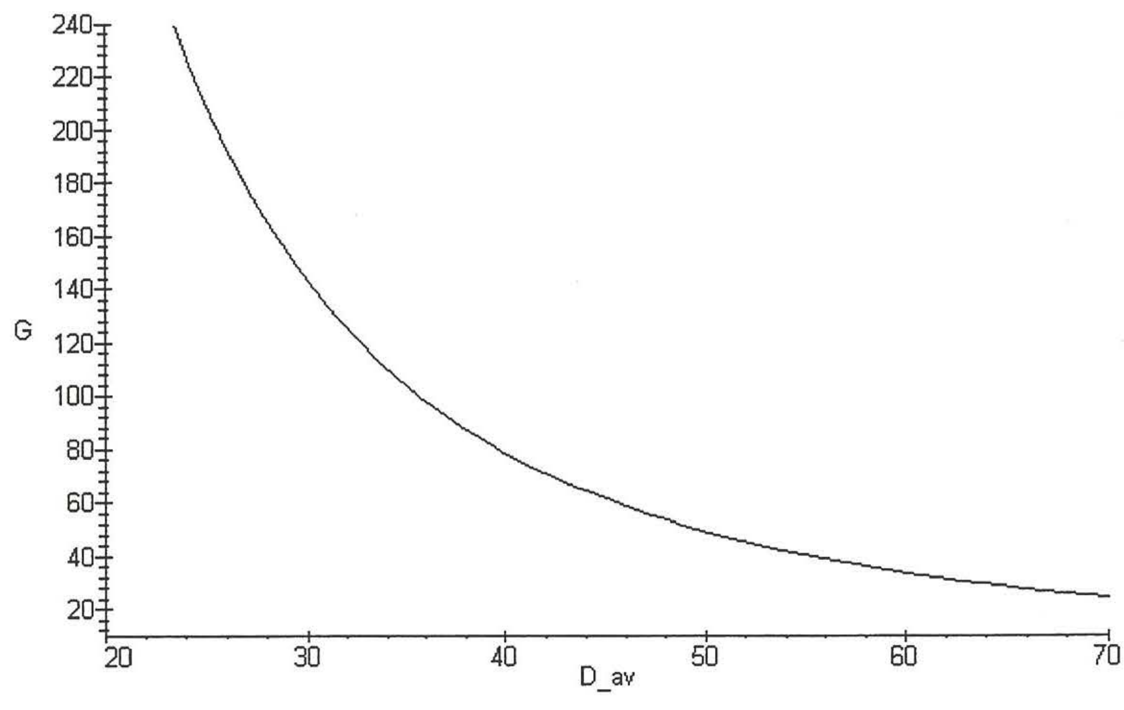

Figure (8): Minimum turbulent shear gradient required to break up the clusters. Base powder is silica-tin mixture.