# Attaining a Controlled Graded Distribution of Particles in Polymerizing Fluid for Functionally Graded Materials

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**ABSTRACT:** The precise control on concentration profile of dispersion in functionally graded material (FGM) is essential for obtaining a desired material. A suitable simulation of parameters and an appropriate model that describes the motion of particles in the fluid can predict various aspects those are needed to produce FGM, by gravity sedimentation or centrifugation technique. Simulation was conducted to observe the changes in concentration profile, while using the following equations applicable to polymerizing fluid, and to determine the terminal velocities  $(V_m)$  of particles;  $V_m$  $= \{D^{2}(\rho_{s} - \rho_{l})g^{*}(1 - \phi_{s})^{4.65}\}/(18\mu_{0}e^{b_{c}}) \text{ for gravity sedimentation and } V_{m} = \{D^{2}(\rho_{s} - \rho_{l})r\omega^{2}(1 - \phi_{s})^{4.65}\}/(18\mu_{0}e^{b_{c}}) \text{ for cen-}$ trifugation, where *D* is the diameter of the spherical particle,  $\rho_s$ the density of solid particles,  $\rho_l$  the density of fluid,  $\mu$  the viscosity of fluid,  $g^*$  the acceleration due to gravity,  $\phi_s$  is the volume fraction of particles, and  $t_c$  is the elapsed time of curing of thermosetting resin. *b* is a constant, *r* is the radius, and  $\omega$  is

#### **INTRODUCTION**

To achieve the desired concentration profile that provides smooth transition in electrical and thermal stresses across the thickness, gravity sedimentation or centrifugal casting methods are being used while developing functionally graded materials (FGMs).<sup>1-6</sup> These are also considered the most economical and attractive processes for making FGMs.7-9 The performance of FGMs depends upon the concentration profile and the uniform dispersion in the matrix, and therefore, a few studies have been reported on measuring gradients, using X-ray attenuation, laser confocal microscopy, microstructure, dielectric strength, abrasive wear of material, z-axis laser light scattering device, etc.<sup>10–12</sup> The technology of making FGM still remains in the experimental stage, because of the lack of information on fundamental understanding of transport phenomenon, occurring in a dispersion under gravitational or centrifugal force, particularly in a matrix, which changes its characteristics with elapsed time; for example, polymerizing fluid. The motion of

the angular velocity. This simulation demonstrates that the time of centrifugation/sedimentation, particle size, distribution of particle size, and centrifugal/gravitational forces can be effectively utilized to attain a desired concentration profile in graded materials. Simulation also revealed that there exist the possibility of two graded profiles, namely low concentration profile and high concentration profile, in one sample of graded material, made either by centrifugation or sedimentation. Low concentration profile is more sensitive to particle size distribution as compared to high concentration profile. The present simulation method is also sensitive to concentration-measuring methods. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 99: 3009–3017, 2006

**Key words:** functionally graded material; epoxy resin; silicon carbide particles; centrifugation; concentration profile

particles that decides the concentration profile is influenced by several factors, such as concentration of particles, density difference in dispersion and dispersoid, extent of centrifugal force, packing density of particles, shape, size, and size distribution of particles, Reynolds number, viscosity of medium, interaction of particles, hindered settling effects, and so on. The movement of single solid particle in Newtonian fluid is fairly understood, even though the debate on scope and explanations of motion still continues, particularly in concentrated dispersions.<sup>13,14</sup> A few recent publications related to FGMs proposed the models for evaluating particle velocities under centrifugal force, which help in generating concentration profile of suspended particles. Gao and Wang<sup>7</sup> added one more dimension in the model by incorporating solidification process during the centrifugal casting of FGMs. This solidification was based on freezing front propagation and particle migration. As far as thermosetting type polymer-matrix-based FGMs are concerned, the viscosity of the matrix changes with the extent of polymerization. This change in viscosity of polymer is throughout the material, which is different from freezing front propagation, as was considered by Gao and Wang for solidification process. A few investigators<sup>15–17</sup> have advocated that movement of particles in

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nonNewtonian fluids can also be predicted with correction in Reynolds number, as follows:

$$R_e = R_{\text{egen}} = \frac{\rho_l V^{2-n} D^n}{K} \tag{1}$$

where  $R_e$  is the Reynolds number based on  $\rho Vd/\mu$ ,  $R_{\text{egen}}$  the generalized Reynolds number, V the terminal velocity of solid particles, D the diameter of the particle,  $\rho_l$  the liquid density, n the power law index, and K is the consistency index. Love<sup>18</sup> proposed a time-dependent viscosity,  $\mu(t)$ , of fluid that was assumed constant for some incubation period, followed by a linear increase with time after incubation period, as per the following relation:

$$\mu(t) = \mu_0 + f^* t \tag{2}$$

where  $\mu_0$  is the initial constant viscosity,  $f^*$  the transient viscosity factor, and t is the time following the initial constant viscosity. The viscosity parameter was proposed<sup>18</sup> for predicting the mud-line displacement of a simulated fluid dispersion using Stokes law, with hindered settling model of Richardson and Zaki.<sup>19</sup> Several models, including empirical, gelation, freevolume analysis-based, etc., have been proposed<sup>20,21</sup> to estimate the viscosity of the polymerizing fluid. Though these models provide a good approximate representation, the experimental data when plotted against time elapse showed a well-defined exponential curve of the following type, for polymerizing fluids<sup>20</sup>:

$$\mu(t) = \mu_0 \mathrm{e}^{bt} \tag{3}$$

where  $\mu_0$  is the coefficient corresponding to the viscosity of fluid at time t = 0 and b is the rate of change of viscosity with time. The values of constants  $\mu_0$  and *b* can be easily determined using experimental values. Hashmi and Dwivedi<sup>22</sup> proposed the model predicting the movement of particles in polymerizing fluid by incorporating the kinetic parameter, (eq. (3)) a time-dependent viscosity, along with Richardson Zaki crowd factor in Stokes law, at low Reynolds number. This model takes into account the change in viscosity of dispersion medium with time, as the crosslinking reaction proceeds, and is capable of explaining the progress of clear zone, sediment zone, and graded concentration zones of a particle-dispersed polymerizing fluid, under gravitational or centrifugal forces. According to this model, the maximum terminal velocity,  $V_m$  of the particle in a polymerizing fluid under gravitational force is estimated, as given later:

$$V_m = \{D^2(\rho_s - \rho_l)g^*(1 - \phi_s)^{4.65}\}/(18\mu_0 e^{bt_c})$$
(4)

In this equation, *D* is the diameter of the spherical particle,  $\rho_s$  the density of solid particles,  $\rho_l$  the density of the fluid,  $\mu$  the viscosity of the fluid,  $g^*$  the acceleration due to gravity, and  $\phi_s$  is the volume fraction of particles in the region under consideration. Time  $t_c$  is different than the time of sedimentation. It is the elapsed time of curing of thermosetting resin. *b* is a constant, which can be determined experimentally from cure-kinetic data of the polymerizing fluid.

For centrifugation, the acceleration due to gravity can be replaced with  $r\omega^2$ , where *r* is the radius and  $\omega$  is the angular velocity. The eq. (4) can be written as follows, for centrifugation:

$$V_m = \{D^2(\rho_s - \rho_l) r \omega^2 (1 - \phi_s)^{4.65}\} / (18\mu_0 \,\mathrm{e}^{bt_c})$$
 (5)

Both eqs. (4) and (5) describe the motion of ideal single particle diameter in the polymerizing fluid. In practice, there is always a size-range of particles used, in the preparation of FGM. Variation in particle diameter is in fact required, for making a graded material. In the present article, simulation is conducted, to observe the nature of the concentration profile that resulted from variation in time of centrifugation/sedimentation, volume fraction of particles, size distribution of particles, etc., which would help in attaining the desired concentration profile in functionally graded polymerbased composites.

## NUMERICAL AND EXPERIMENTAL PROCEDURE

The simulation was performed with the following assumptions:

- The flow of particles is entirely in the direction of centrifugal force or gravitational force (in case of sedimentation due to gravitational force).
- Gravitational force is negligible against the strong centrifugal force.
- Diameter of container is much larger than the particle diameter, and consequently wall effects are ignored.
- Interparticle forces and surface tensions are considered negligible.
- Compressive stresses are neglected.
- The motion of particles is slow, and reaches the terminal velocity instantaneously.
- The particles are rigid and do not react chemically with the matrix.
- Density of the matrix remains constant during the experiment, and volume change is negligible.

The concentration profile was worked out by evaluating the positions of particles from a reference distance, which was obtained by the information related to particle movement in the fluid. The distance covered by the particle in a short span of time was estimated from the velocity of the particle, given by either eq. (4) or (5) (whichever is applicable), assuming that clear front travels with the particle present at one end. The clear front is an imaginary plane that separates the particle-free-zone from the particle-dispersed medium. During each time interval, the velocity of particle and, hence, the distance covered by the particle was computed using the volume fraction of particles and the viscosity of the resin, estimated in the previous time interval. For computing sediment zone, a maximum of 0.40 volume fraction was fixed, and particle velocity was assumed to be zero at this volume fraction. The time interval for each computation was kept at 1 s, which can be increased or decreased, depending upon the total time of centrifugation/sedimentation and required data points. The initial uniform concentration was kept at 0.099, volume fraction of SiC particles, so that both low concentration as well as high concentration profiles can be seen easily. The initial concentration would definitely change the profile, but in the present simulation, it was kept fixed to avoid the number of figures. A volume fraction of 0.099 was also divided into three parts to study the effect of particle size distribution. The maximum possible concentration was assumed to be 0.40, volume fraction of SiC, and compaction of sedimentation was not included in the simulation. The particle shape was assumed to be spherical in all calculations. The minimum thickness of sedimentation layers was assumed to be 1.0 mm from the bottom. Once the 1.0-mm sediment layer attains 0.40-volume fraction of SiC, the additional particles were added to the next sediment layer, and the process was continued.

To study the effect of size distribution of particles on concentration profile at a particular time and place under centrifugal force, the volume fraction of multiparticle system ( $V_{mp r_i}$ ) at certain position,  $r_i$ , in the sample was estimated using the following relation:

$$V_{\rm mp\,ri} = \{\Phi_1 V_{\rm D1} \mathbf{r}_i + \Phi_2 V_{\rm D2} \mathbf{r}_i + \Phi_3 V_{\rm D3} \mathbf{r}_i \dots \dots + \Phi_n V_{\rm Dn} \mathbf{r}_i\} / V$$
(6)

$$V = \{\Phi_1 + \Phi_2 + \Phi_3 + \dots + \Phi_n\}$$
(7)

where *V* is the initial volume fraction of particles in the system at time t = 0,  $\phi_i$  the fraction of particles representing the *i*th particle fraction having a diameter  $D_i$  in *V* at time t = 0,  $V_{\text{Diri}}$  the volume fraction of particles at  $r_i$  position at a certain time and evaluated by assuming identical particles of diameter  $D_i$  and initial volume of particles as *V*, and *n* is the number of different diameters of particles present in the system.

Simulation was performed for SiC particles, having average diameter 20, 30 and 40  $\mu$ m, dispersed in polysulfide-modified epoxy resin, at different conditions.

A few experiments were conducted to compare this theoretical simulation. Room temperature-cure polysulfide-modified epoxy resin, obtained from M/s Choksey Chemicals, India, was used in this study. The density of epoxy resin cured at room temperature with hardener in the ratio of 2:1 was 1150 kg/m<sup>3</sup>. The nominal particle size of silicon carbide (SiC) was 20  $\mu$ m. The density of particles, estimated by relative density bottle, was 3200 kg/m<sup>3</sup>. Samples were prepared under centrifugal force and without centrifugal force at  $(30 \pm 2)^{\circ}$ C. Polysulfide-modified epoxy resin, with uniformly dispersed SiC particles, was filled in cylindrical molds of 30 mm height and 10 mm diameter, and were kept at room temperature for 24 h. The samples were then removed from the molds for observation. The simple arrangement was used for preparing centrifuged samples, as reported elsewhere.<sup>10</sup> Molds with samples were fixed at both the ends of a bar, which was fixed on a shaft of the motor by appropriate clamping arrangement. A tachometer recorded revolution per minute (rpm) of the assembly. Silicon carbide particles of 0.099 volume fractions were added to a mix of epoxy resin and hardener, and were thoroughly mixed. Equal amounts of mix were transferred to two identical molds. The molds were closed and fitted on the centrifugal arrangement. The rotational speed and distance between rotor axis and the end of sample was adjusted, to apply a centrifugal force equivalent to  $224 \times g^*$ . The time of centrifugation was 120 s. After centrifugation molds were removed from the rotating system and kept at room temperature for 24 h. Densities of the functionally gradient polymeric composites were determined, by cutting discs of 1.0 mm from the cylindrical pin. The densities of the specimen were measured by using a Mettler Toledo machine, using the Archimedes principle.

## **RESULTS AND DISCUSSION**

# Effect of time of centrifugation

Simulation was performed to study uniformly distributed SiC particles in polymerizing polysulfide epoxy resin, under centrifugation, to obtain FGM. Figure 1 is a plot between volume fractions of SiC particles (30  $\mu$ m) versus thickness of the sample. The initial volume fraction of particles was 0.099. The total length of sample was 30 mm and referred as zero length at top of the sample, which is near the center of revolution. The farthest end of sample would be referred as bottom of the sample. Symbols on curves are notions to differentiate one curve from the other. Volume fraction of SiC ( $\phi$ ) varied with time and length of the sample. After attaining nearly 40 vol % of SiC (assumed maximum packing density of SiC particles in epoxy resin), the increase in the volume fraction was stopped. Initially, a concentration profile was ob-



**Figure 1** Volume fraction of 30- $\mu$ m SiC particle along the length of centrifuged sample, at different time of centrifugation.

served, in which the concentration varied from 0 to 0.099, which could be referred as low concentration profile. Thereafter, the concentration of particles remained constant for a particular length of sample. At the end of the sample, another graded profile was observed, in which the concentration varied from 0.099 to a maximum possible concentration that was 0.40. This concentration profile would be referred hereafter as high concentration profile. After this gradient, a maximum packing zone was observed, which increased in thickness with time. The thickness of this zone was estimated using the following simple relationship:

$$t_{\rm sed} = t_{\rm cl} \{ \phi_i / (\phi_m^- \phi_i) \}$$
(8)

where  $t_{sed}$  is the sediment thickness,  $t_{cl}$  the length of clear zone,  $\phi_i$  the initial concentration, and  $\phi_m$  is the maximum concentration that can be achieved. As an ideal case for single diameter, noninteracting particle system, there should be a step increase in both places, that is, at low concentration gradient and high concentration gradient, as described earlier.

The present simulation was applied for 1-mm thick section of the sample, and the average concentration in this volume was shown here in these plots, and therefore, the profile was observed. This method was adopted to make simulation more realistic and sensitive to concentration measuring methods; for example in a system like SiC Epoxy, which is being discussed here, the concentration profile could be observed by cutting several discs from sample pin and using density measurement, a profile of volume fraction versus length of sample could be obtained. This method is based on averaging the concentration of SiC, within the thickness of sample. The thinner the section of sample, more accurate would be the results of repre-

sentative plots. This figure revealed that at time t = 0, the concentration profile was constant throughout the length of the sample. At 32 s, nearly 4 mm of top portion of 30-mm long sample was clear of the 30  $\mu$ m particles. Next 3 mm had a graded concentration. At 55th second, more than 7 mm length of sample was free from particles, and a steep gradient was observed at the bottom. The middle portion remained at 0.099 volume fraction. Increased time of centrifugation increased the clear front as well as the bottom thickness, as is evident from Figure 1. At t = 80 s, nearly 12 mm length of the sample became free from SiC particles. Next 3 mm was a graded material, followed by a high concentration profile, varying from 0.099 to 0.40 volume fraction SiC, followed by nearly 4 mm length of a material zone, having 0.4 volume fraction of SiC. At t = 145 s, a continuous gradient was observed starting from 20 to 25 mm length of sample, and thereafter, a zone of maximum packing was observed.

#### Effect of particle size

Figure 2 is a plot between volume fractions of SiC particles (40  $\mu$ m) versus thickness of the sample. The initial volume fraction of particles was 0.099. At 34 s, nearly 6 mm of top portion of 30 mm long sample was clear of the 40  $\mu$ m particles. Owing to the increased particle size as compared to the simulation shown in Figure 1, the speed of clear zone was faster. Other features of curve are similar, as those discussed for 30  $\mu$ m particles, and shown in Figure 1. Another Figure 3 is shown to further observe the effect of particle size, in which 20  $\mu$ m particles were shown to centrifuge under same conditions, as shown in Figures 1 and 2.

On comparing Figures 1, 2, and 3 the trend of concentration profile is almost the same, but on time scale, significant changes were observed. Particles with



**Figure 2** Volume fraction of  $40-\mu$ m SiC particle along the length of centrifuged sample, at different time of centrifugation.



**Figure 3** Volume fraction of  $20-\mu$ m SiC particle along the length of centrifuged sample, at different time of centrifugation.

20-mm size travel very slow as compared to 30 and 40 mm particles in polysulfide epoxy resin, and therefore, different profiles were observed. Length of clear zone is less and steep gradient was observed at the end. All three figures demonstrate that single size particles show normally four zones. The innermost side of sample shows a clear front that increases with time on centrifugation, and the speed of the clear front is controlled by particle size, under similar process conditions. Particles of large diameters move faster than smaller particles.

#### Effect of centrifugal force

Figure 4 is a plot between volume fractions of SiC particles (30  $\mu$ m) versus thickness of the sample, under different forces: gravitational as well as centrifugal



Figure 4 Effect of centrifugal forces on concentration profile.



**Figure 5** Variation in volume fraction of 20, 30, and 40  $\mu$ m size SiC particles (having equal volume fraction of each size) along the length of centrifuged sample, at different time of centrifugation, at high concentration region.

forces, after 80 s of centrifugation. It is evident that gravitational force  $(1 \times g^*)$  did not affect the concentration profile in a short time of 80 s, but centrifugal forces equivalent to  $50 \times g^*$ ,  $100 \times g^*$ , or  $150 \times g^*$  showed concentration profiles, in which the concentration varies from 0 to 0.40 volume fraction of SiC. An increase in centrifugal force increased the length of the clear zone as well as the thickness of the sediment zone. The effect of viscosity changes due to polymerizing reaction may not play a significant role in this case, because of the short span of time, and therefore, the increased centrifugal force would reduce the time of formation of a specified concentration profile.

## Effect of particle size distribution

It is evident from literature and simulated results presented here that different particles travel different distances, depending upon their sizes and crowding of other particles. In general, any sample of particle has a size-distribution pattern. This distribution pattern produces relatively different concentration profiles, corresponding to each particle size present in the sample. A set of particles having 20, 30, and 40  $\mu$ m size, with equal volume fraction of each size, is evenly distributed in poly sulfide epoxy resin and is centrifuged with a force equivalent to 100 times gravitational force. The overall initial volume fraction was taken as 0.099. A simulation of high concentration profile (as already defined in Results and Discussion) was obtained, as shown in Figure 5, for different times of centrifugation. In this figure, five curves belonging to different time of centrifugation are shown. Initial concentration profile across the thickness of sample is constant throughout the length, as shown, corresponding to t = 0. At t = 26 s, concentration profile



**Figure 6** Variation in volume fraction of 20, 30, and 40  $\mu$ m size SiC particles (having equal volume fraction of each size) along the length of centrifuged sample, at different time of centrifugation.

was observed between 27.5 and 30 mm. In this case, maximum packing zone, as described earlier, was not observed. Even at 56 s of centrifugation, the maximum packing zone was missing and a profile from 18 to 30 mm was observed, in which concentration varied from 0.033 to 0.40. At 142 s of centrifugation, another profile in which the concentration varies from 0.033 to 0.40 was shown in this figure. Centrifugation of 164 s produces a profile that started from nearly 22 mm and ended at 26 mm. Thereafter, maximum packing zone was observed.

Figure 6 shows both low and high concentration profiles, at different time of centrifugation. The low concentration profile illustrates differences and effects of size distribution of particles. At t = 26 s, a concentration profile from 0 to 0.033 within 2 mm of top side sample can be observed in the plot. After a constant 0.033  $\phi$  concentration of 2 mm length, the concentration increased to 0.099. Similarly, after 55 s, a similar profile with a different length was observed. In this profile, a small clear zone, a small concentration profile followed by a constant concentration and another gradient, was noticed. Since the range of particle size selected was 20, 30, and 40  $\mu$ m, and only three sizes of particles were used to simulate the conditions, and therefore, three step profiles were observed. Had it been more number of sizes, such as particles having diameter 21, 22, 23,..., 40 µm i.e., 21 numbers, a 21-step profile would have been obtained that could further smoothen the step height, as compared to that shown in Figure 6. Both low concentration profiles and high concentration profile would change the slope of the curve, depending upon the volumetric composition of various sizes of particles. Both Figures 5 and 6 show equal volumes of three different sizes of par-



**Figure 7** Variation in volume fraction of 20, 30, and 40  $\mu$ m size SiC particles along the length of centrifuged sample, at different time of centrifugation, at high concentration region.

ticles. On changing this proportion, the slope of the concentration profile would vary with composition and time, as shown in Figures 7 and 8, wherein the particle compositions were 0.022, 0.055, 0.022 and 0.011, 0.055, 0.033 volume fractions for 20, 30, and 40  $\mu$ m SiC particles, respectively. On comparing Figures 5, 7, and 8, the effect on concentration profile on particle composition within the sample can be observed easily. The variation in concentration that includes both low and high concentration profiles in the entire length of sample can be seen in Figures 9 and 10. The trends of variation in these two figures are similar to that shown in Figure 6, but the effect of variation of composition of particles can be seen clearly. A careful



**Figure 8** Variation in volume fraction of 20, 30, and 40  $\mu$ m size SiC particles (having different volume fraction of each size) along the length of centrifuged sample, at different time of centrifugation, at high concentration region.



**Figure 9** Variation in volume fraction of 20, 30, and 40  $\mu$ m size SiC particles along the length of centrifuged sample, at different time of centrifugation.

observation reveals that low concentration region is sensitive to size distribution of particles. It can be attributed to the relative terminal velocity of particles of different sizes in the low concentration region. In a high concentration region, crowding factor dominates particle movement and difference in relative velocities is less effective, and hence changes are less here.

### Concentration changes at different positions

So far, in this article, concentration changes with respect to sample length at a certain time of centrifugation were discussed. It is also important to know the concentration variation of SiC with time in polysulfide



**Figure 10** Variation in volume fraction of 20, 30, and  $40 \ \mu m$  size SiC particles (having different volume fraction of each size) along the length of centrifuged sample, at different time of centrifugation.



**Figure 11** Variation in volume fraction of 30- $\mu$ m SiC particles with time, at different positions, under gravity sedimentation.

epoxy resin, at various positions within the sample during sedimentation or centrifugation. Using same model, it was possible to get information on variation in volume fraction with time. Figure 11 is a plot between volume fractions of SiC particles (30  $\mu$ m) versus time in seconds. The initial volume fraction of particles was 0.099. The changes in volume fraction at different positions were evaluated under gravitational force and are shown in this figure. The total height of sample was 30 mm, and volume fraction of SiC was evaluated at three different heights using eq. (4). These heights were selected to represent the different regions; for example, 0.1 mm near bottom where particles would sediment with time forming a highest packing density of SiC, top 0.5 and 1.0 mm were selected to represent two different regions those become clear of SiC particle due to sedimentation. Volume fraction of SiC ( $\phi$ ) started increasing with time at 0.1 mm above the bottom, after filling up 0.1 mm depth at bottom. After attaining 40 vol % of SiC, the increase in volume fraction was not possible, and therefore after nearly 950 s, there was no change in value of volume fraction with time in a 0.1-mm thick layer. The volume fraction of SiC remained constant at two positions, i.e., 0.5 and 1.0 mm from top and parallel line to x-axis, show a constant value of  $\phi$ . At position 0.5 mm from top,  $\phi$  starts reducing after almost 256 s, and then, became free of SiC. Similarly, at 1.0 mm from the top, reduction in  $\phi$  was observed. The velocity of clear front slowly goes down, as is evident from the two values at 0.5 and 1.0 mm, where in had it been the same velocity of clear front, it must have reached to 1 mm in nearly 512 s, which is not true as shown in Figure 10. There appears the effect of hindrance of particles as well as extent of polymerization that reflected an increased viscosity of medium,



**Figure 12** Variation in volume fraction of 20- $\mu$ m SiC particles with time at different positions, under centrifugation equivalent to 10 times gravitational force.

and hence, reduced speed of clear front. Figure 11 is a similar plot to that shown in Figure 10, with a difference that particle size is smaller than 20  $\mu$ m instead of 30  $\mu$ m. On comparing Figures 10 and 11, decreased size of SiC increased the time to buildup sediment layer at 0.1 mm above the bottom. The region at 0.5 mm from the top was delayed to become clear, because of the lower speed of particles. The second top region at 1.0 mm remained as such, and no change was observed in the concentration of particles up to 1500 s, which is an outcome of slow speed of particle as well as the increased viscosity of polysulfide epoxy resin with elapsed time.

Figure 12 describes the changes in volume fraction of SiC particles with time, at various positions. The simulation was conducted for applying centrifugal force equivalent to 10 times that of gravitational force. The volume fraction of 20- $\mu$ m SiC particles increased from the initial concentration to maximum packing level in a very short period of 190 s, as compared to sedimentation under gravitational force, as shown in Figure 13. The top clear zones at 0.5 and 1.0 mm are also shown in Figure 12. The effect of change in viscosity of system (polymerizing polymer) may be negligible in the case, wherein the elapsed time is only few seconds at initial stages. Interestingly, at 1.0 mm height, after nearly 110 s, clear zone is observed that was missing in sedimentation even after 1500 s.

#### **Experimental validation**

Figure 14 compares the predicted concentration profile of SiC particles in graded material with measured volume percent of particles. Most of the points fall on either side of the predicted lines. Predicted results



**Figure 13** Variation in volume fraction of 20- $\mu$ m SiC particles with time at different positions, under gravity sedimentation.

were found well within the range of experimental variations. Uncentrifuged sample shows constant volume fraction throughout the length of sample, and both experimental and theoretical values are the same in this case.

The development of FGM using centrifugation or sedimentation techniques requires precise control on concentration level, at different positions of a graded material. The present simulation scheme provides a base for a controlled centrifugation process. This simulation is capable of predicting and explaining progress of clear zone, sediment zone, and graded concentration zones. The two, low and high .concentration profiles were also observed, which are important from the point of view of FGM-development activities. The length of the sample would be important



**Figure 14** Plot between volume fraction of SiC particle and length of sample.

to join both profiles into one to attain a desired concentration profile in FGM.

# CONCLUSIONS

This study concludes that parameters, such as particle size, distribution of particle size, time of centrifugation, centrifugal/gravitational forces, can be effectively utilized to attain a desired concentration profile in graded materials. Simulation reveals that there exist the possibility of two graded profiles, namely low concentration profile and high concentration profiles, in one sample of graded material made either by centrifugation or sedimentation. Size distribution of particle significantly affects the low concentration profile, as compared to high concentration profile.

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## References

1. Maciborski, J. D.; Dolez, P. I.; Love, B. J. Mater Sci Eng A 2003, 361, 392.

- 2. Ray, S. Mater Sci B 1995, 18, 693.
- 3. Asthana, R. J Mater Synth Process 1997, 5, 339.
- 4. Ray, S. J Mater Sci 1993, B28, 5397.
- 5. Hashim, J.; Looney, L.; Hashmi, M. S. J. J Mater Process Technol 2002, 123, 251.
- 6. Gao, J. W.; Wang, C. Y. J Heat Transfer 2001, 123, 368.
- 7. Gao, J. W.; Wang, C. Y. Mater Sci Eng A 2000, 292, 207.
- 8. Mortensen, A.; Suresh, S. Int Mater Rev 1995, 40, 239.
- 9. Ho, S.; Lavernia, E. J. Metal Mater Trans A 1996, 27, 3241.
- Hashmi, S. A. R.; Dwivedi, U. K.; Jain, D.; Naik, A.; Chand, N. J Appl Poly Sci 2005, 96, 550.
- 11. Lagasse, R. R.; Thompson, K. R. Polymer 2002, 43 ,803.
- 12. Hoffman, D. L.; Brooks, D. R.; Dolez, P. I.; Love, B. J. Rev Sci Instrum 2002, 73, 2479.
- 13. Brenner, M. P.; Mucha, P. J. Nature 2001, 409, 568.
- 14. Segre, P. N.; Liu, F.; Umbanhowar, P.; Weitz, D. A. Nature 2001, 409, 594.
- 15. Chhabra, R. P. Chem Eng Processing 1990, 28, 89.
- Chhabra, R. P.; Richardson, J. F. Non Newtonian Flow in the Process Industries; Butterworth-Heinemann: Oxford, 1999.
- 17. Kelessidis, V. C. Chem Eng Sci 2004, 59, 4437.
- 18. Love, B. J. Particulate Sci Technol 2004, 22, 285.
- 19. Richardson, J. F.; Zaki, W. M. Trans Inst Chem Eng 1965, 32, 35.
- Kiuna, N.; Lawrence, C. J.; Fontana, Q. P. W.; Lee, P. D.; Selerland, T.; Spelt, P. D. M. Compos A 2002, 33, 1497.
- 21. Srivastava, A.; Singh, P. J Polym Eng 2002, 22, 285.
- 22. Hashmi, S. A. R.; Dwivedi, U. K. J Mater Sci Lett, submitted.