

Settling Behavior of Fillers in Thermosetting Epoxy Casting Resins During Cure

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Synopsis

Using a theory based on a modified Stokes' law equation, the settling behavior of fillers was investigated (glass beads and silica powder) in thermosetting epoxy casting resins during cure. In this study a suspension (a thermosetting epoxy casting resin) containing a large amount of filler particles is assumed to be a homogeneous fluid, and the settling phenomena are treated such that one or more particles of the filler settle into the homogeneous fluid under gravity without interference from other particles. The viscosity increase of the fluid during cure is taken into consideration. The experimental results agree well with the theoretical predictions when the settling distances are small and the particle size distribution is narrow. When these conditions are not satisfied, various effects are observed such as convection, filtering, combination, bottom effect, and a broad particle size distribution. In addition, the formation of a compression zone and of a compaction zone are clearly observed.

INTRODUCTION

At present epoxy casting resins are widely used in the electrical industry as insulating materials. They also have inherent advantages of good processability, design flexibility, dimensional stability, and good electrical and mechanical properties.¹⁻³ In these epoxy casting resins, a large amount of fillers (such as glass beads, silica powder, powdered kaolin, corundum, calcite, etc.) are incorporated as an important ingredient to improve mechanical properties, reduce shrinkage, impart flame retardancy, lower cost, and so on. An epoxy casting resin can be regarded as a suspension before it reaches the gel stage. Its filler particles tend to settle easily when stored or during the curing process due to their greater densities.

The sedimentation of fillers causes heterogeneities in cured casting resins. Although fillers are necessary to provide improved properties, they are ultimately undesirable for optimum properties in the finished product. Hence, it is important to understand the settling behavior of filler particles in a thermosetting epoxy casting resin even if the most desirable filler with respect to its particle size or size distribution, which hardly settles during cure for a given epoxy casting resin composition, is used. To date, however, there have been few reports on this subject.⁴ In the field of paint technology, there are a number of testing methods⁵ for assessing the degree of the settling of pigment in paint during storage. However, these are qualitative and do not take into consideration the effect of the viscosity increase of the fluid during cure. Therefore, they cannot be applicable to the particular case involving thermosetting epoxy casting resins. This paper explains first the relationship between the curing process and the filler sedimentation of a typical thermosetting epoxy casting resin. It also describes the quantitative treatment of settling behavior of spherical and granular filler

particles in a thermosetting epoxy casting resin, using a modified Stokes' law equation. The theoretical predictions are then tested by the experimental results for typical thermosetting epoxy casting resin compositions which are filled with glass beads and silica powder.

Relationship Between Curing Process and Filler Sedimentation

Figure 1 shows an example of the curing process of a typical thermosetting epoxy casting resin,⁶ as represented by density changes. Process A-B is mainly a thermal expansion process of formulated liquid epoxy casting resin; process B-D is a shrinkable process of formulated resin due to the curing reaction; and process D-F is a thermally shrinkable process of a fully cured epoxy casting resin. Point C is the gelation point, and point E is the glass transition temperature, respectively. During the process from point A to point C, the epoxy casting resin can be regarded as a suspension, but it nearly loses its liquidity before point C is reached because of the viscosity increase of the fluid during curing. If point A is allowed to approach point B, the settling of filler particles must occur during process B-C. Therefore, only the sedimentation during process B-C is reported herein.

THEORETICAL CONSIDERATIONS

An epoxy casting resin prior to gelation is a suspension which contains filler particles; as each particle settles, it interferes with other particles. As the concentration of particles increases, the settling velocity decreases. This phenomenon occurs because the density of the suspension becomes larger than that of the fluid, which augments the buoyancy of a particle.⁷ The apparent viscosity

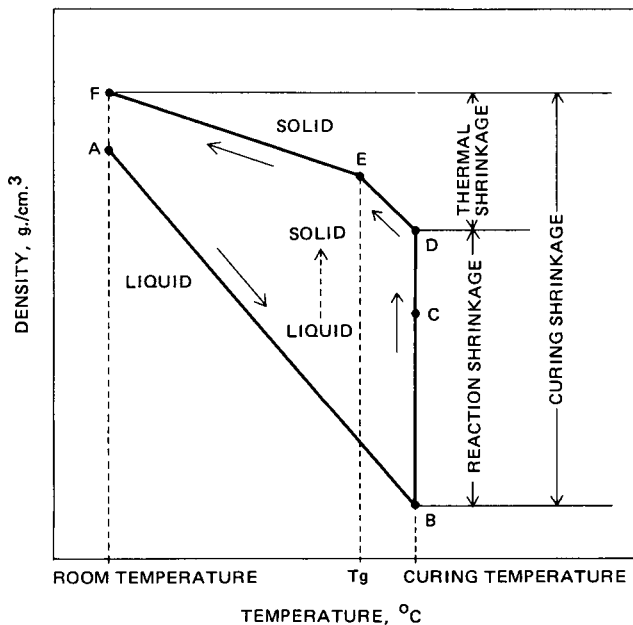


Fig. 1. Density changes of a typical epoxy casting resin during the curing process (schematic diagram.)

of the suspension increases due to the increase in the velocity gradient in the fluid, which corresponds to the increase of viscous resistance to a particle.⁷ A convection of the fluid resulting from the displacement of the fluid by settling particles influences the sedimentation of a particle.

Furthermore, in thermosetting epoxy casting resins it must be considered that the viscosity increase of the fluid during cure greatly affects the sedimentation of filler particles, and, conversely, there may be some cases where particles themselves or contaminants on particles have a catalytic effect on the cure reaction. Therefore, during process B-C (Fig. 1), the following factors influence the sedimentation of filler particles: (a) hindered settling due to the high concentration of filler particles, (b) increase in the viscosity and the density of the fluid as the cure reaction proceeds, and (c) catalytic effect of particles or contaminants on the cure reaction.

Concerning the hindered settling of filler particles, if spherical or granular filler particles are of uniform size and density and are well distributed throughout the fluid, the particles obey Stokes' law. The rate of sedimentation can be represented by the Stokes velocity multiplied by a term which is a function of the concentration of filler particles.⁷ In considering (b) and (c) above, the suspension (epoxy casting resin) should be considered as one component and not as two parts (particles and the fluid).

Accordingly, the suspension itself is regarded as a homogeneous fluid,^{8,9} and then the settling phenomena are dealt with so that one or more particles, which are essentially identical to the particles contained in the suspension, settle into the homogeneous suspension under gravity. In this case, the Stokes equation is applicable to the settling behavior of filler particles in the epoxy casting resin, and therefore we have

$$H = \frac{g(d_p - d_s)D_p^2 T}{18\eta_s} \quad (1)$$

where H is the settling distance of the filler particle, g is the acceleration due to gravity, d_p is the density of sediment particle (or the filler particle), d_s is the density of the suspension (or the epoxy casting resin), D_p is the diameter of the filler particle, T is the settling time (it may be approximately the gelation time of the epoxy casting resin), and η_s is the viscosity of the suspension. In eq. (1), g and d_p are constant and, as will be shown later, d_s can be regarded as constant. Although D_p of each particle is not identical, it is possible to make them approximately the same as in our experiment.

Since η_s varies significantly as the cure reaction proceeds, H is not calculated directly from eq. (1). However, H can be calculated using equation (2) since in this case η_s in a short time Δt_k is considered to be constant (Fig. 2):

$$H = \frac{g(d_p - d_s)D_p^2}{18\eta_s} \sum_{k=1}^n \frac{\Delta t_k}{\eta_s(\alpha_k)} \quad (2)$$

where $\Delta t_k = t_k - t_{k-1}$ ($t_0 = 0$ and $t_n = T$), α_k is the middle point in Δt_k , and $\eta_s(\alpha_k)$ is the viscosity at α_k .

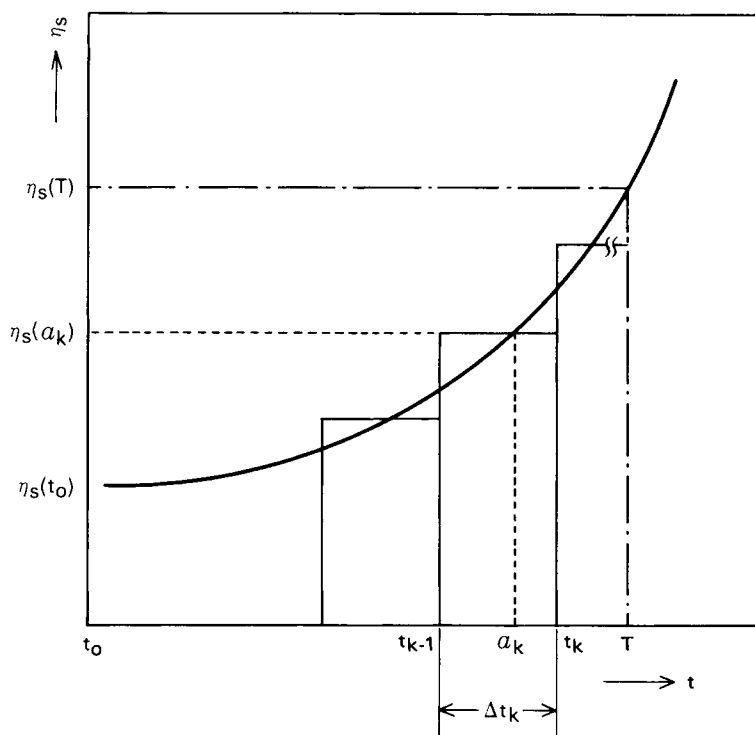


Fig. 2. Diagrammatic representation of calculating method for settling distances using eq. (2).

EXPERIMENTAL

Reagents and Compositions

The epoxy resin was Epikote 1001 (solid diglycidyl ether of bisphenol A resin) of Shell Chemical Co., Holland, epoxy equivalent = 450–500. The acid anhydride of the hardner was hexahydrophthalic anhydride (HHPA) of New Japan Chemical Co. Ltd., Japan. The melting temperature was 34°–36°C. The spherical and granular fillers were glass beads and silica powder, respectively. Glass beads were Glass Bead MS-L of Shinetsu Chem. Industrial Co., Japan, with a diameter range of 44–74 microns. They were made of conventional sodalime glass (SiO_2 72.8%, Al_2O_3 1.3%, CaO 8.0%, MgO 3.9%, Na_2O 13.3%, and K_2O 0.3%). The reagents, except for the silica powder, were used as received. The silica powder was prepared from Silica KW-150 of Nagase & Co. Ltd., Japan. The purity of Silica KW-150 was 99.5%–99.8%. Silica KW-150 was wet sieved using 105-micron and 149-micron sieves until the filtrate of tap water became almost clear to give a batch of narrow-distribution particles of the required particle size range (149 to 105 microns). The above batch was then dried for more than 16 hr at 120°–130°C just prior to use. Epoxy casting resin compositions and the theoretical filler loadings (the feed concentrations) studied are shown in Tables I and II.

The mixtures for the measurements were prepared as follows: the epoxy resin,

TABLE I
Composition and Filler Loading for Glass-Bead Filled Systems

System	Composition, parts by weight					
	Sample G-1	G-2	G-3	G-4	G-5	G-6
Epikote 1001	100	100	100	100	100	100
HHPA	32.5	32.5	32.5	32.5	32.5	32.5
Glass beads	7.0	14.7	33.2	56.8	88.3	132.5
Filler loading, wt-%	5	10	20	30	40	50

TABLE II
Composition and Filler Loading for Silica Powder-Filled Systems

System	Composition, parts by weight			
	Sample S-1	S-2	S-3	S-4
Epikote 1001	100	100	100	100
HHPA	32.5	32.5	32.5	32.5
Silica powder	88.3	132.5	198.6	309.2
Filler loading, wt-%	40	50	60	70

HHPA, and each filler were preheated separately at 125°–130°C and then mixed and degassed for about 5 min, using an Ishikawa mixing and grinding machine of Ishikawa Mfg. Co. Ltd., Japan. The cure of each composition was performed at 120°C.

Measurements

The particle size distribution and the densities of both fillers were measured at about 25°C with a Sedimentograph Type SA-3 of Shimadzu Seisakusho Ltd., Japan and a helium–air pycnometer Model 1302 of Micromeritics Instrument Corp., U.S.A.

The viscosity changes of the mixtures during cure were measured by the B-type viscometer (rotational viscometer) of Tokyo Keiki Co. Ltd., Japan. During measurements, a viscosity measuring vessel containing a mixture was immersed in a silicone oil bath maintained at 120° ± 1°C. The measurements of the viscosities of lower filler content mixtures were carried out immediately after mixing the given mixture quickly using an immersed rotor of the viscometer to disperse filler particles which settled during the successive measurements.

The initial density of the fluid at 120°C was measured with a common pycnometer. The density changes of the fluid during cure were measured using a modified Gay-Lussac-type pycnometer.¹⁰ It is composed of a vessel of about 25 ml in volume and a cap fitting to the vessel. To the top of the cap is attached a long, thin glass tube with an inner diameter of approximately 2.5 mm and a length of approximately 350 mm. The glass tube is graduated to read to 0.01 ml over the range of 0–1.0 ml. A fluid of about 10 g was poured into the pycnometer and weighed precisely. After the available space in the pycnometer was filled with silicone oil, which is insoluble in the resinous fluid, the pycnometer was submerged into the oil bath at 120° ± 1°C up to the depth of the meniscus in the glass tube.

The volume decrease of the fluid due the cure reaction was measured continuously, and the density increase of the fluid during cure was calculated.

Specimens which were employed to evaluate both the settling distance of the filler particle and the filler content change due to the sedimentation during cure were prepared by pouring each mixture into a glass settling cylinder (approximately 30 mm in diameter and 195 mm in height), allowing it to cure by holding it vertically in an electric oven at $120^{\circ} \pm 1^{\circ}\text{C}$, and then removing the cured mixture from the cylinder. The settling distance of the filler particle was determined by measuring the depth of the cured, clear liquor phase formed at the top zone of the specimen while the temperature of the specimen was maintained at 120°C to exclude the effect of thermal shrinkage.

The filler content change at the specified section in the cured specimen was determined from the ash content of the section. The cured specimen was equally divided into six sections, and in each section the portion adjacent to the wall of the settling cylinder was cut off and discarded in order to exclude the wall effect.¹¹ The ash content was obtained by the following procedure: each sample in block form was put into a small crucible, and the organic portion was burned in the electric furnace at approximately 500°C for glass beads and 800°C for the silica powder; the fillers were not allowed to melt until a constant weight was obtained.

RESULTS AND DISCUSSION

Physical Properties of Reagents and Compositions

The density of the glass beads and the silica powder was 2.30 g/cm^3 and 2.57 g/cm^3 , respectively. Their particle size distributions are shown in Figure 3. The distribution of the sedimentation diameter of the beads is in good agreement with the diameter range, but that of the silica powder is wider than the two sieve-opening sizes. This suggests that particles larger than 149 microns (or larger in long-axis diameter of projected diameters but smaller in short-axis di-

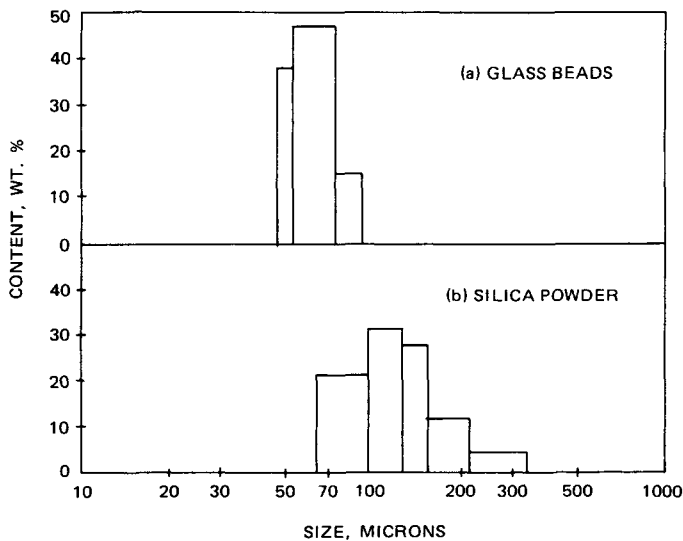


Fig. 3. Particle size distribution of glass beads and silica powder.

ameter of projected diameter) pass through the sieve, and particles smaller than 105 microns remain on the sieve. Larger particles will easily pass through as the sieve is deformed by wet sieving. However, the median size (50% size) is within the range. The above densities and diameter ranges of two fillers are nearly equal to those at 120°C because their thermal expansions are negligible.

Figures 4 and 5 show the viscosity changes of unfilled and filled mixtures during cure at 120°C. If the reactions of unfilled and filled mixtures obey first-order kinetics, the apparent rate constants of the cures can be determined from the slopes of the plots of $\log \eta$ (where η is viscosity) versus t .¹² The silica powder greatly retards the cure reaction of the fluid, whereas glass beads slightly accelerate it as the filler loading increases (Figs. 4 and 5). In general, fillers change the reaction rate, and this is due to their specific heat, thermal conductivity, and surface chemical and physical properties, etc. The fillers do not retard

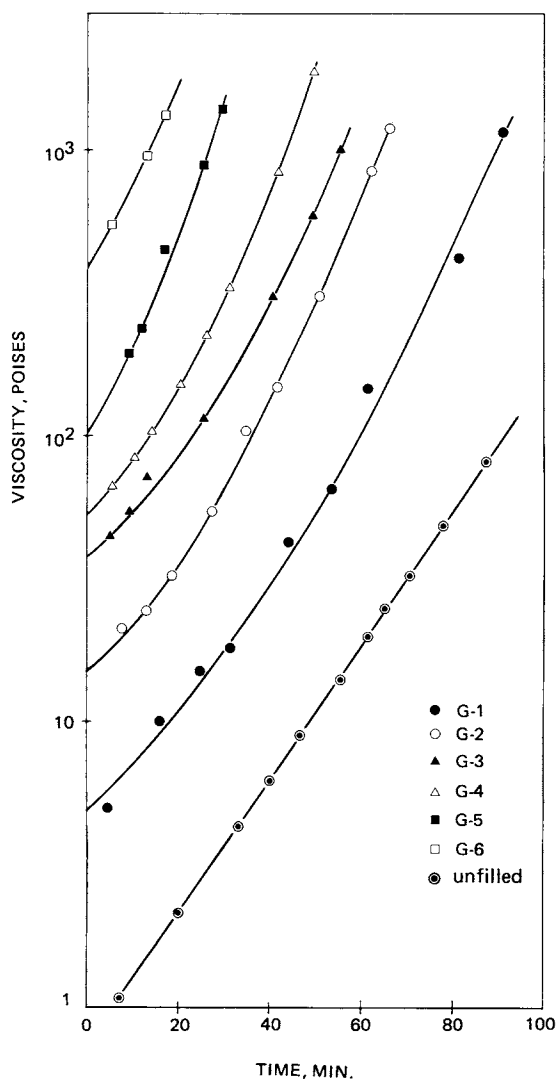


Fig. 4. Viscosity increases with advance of curing reaction, at 120°C.

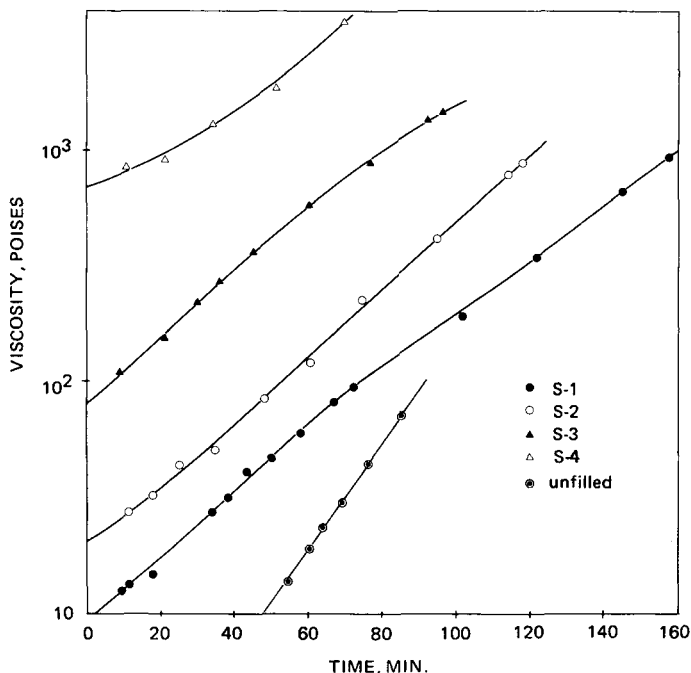


Fig. 5. Viscosity increases with advance of curing reaction, at 120°C.

the reaction rate in all cases.⁴ But in glass beads, the following results were obtained: When glass beads were added with shaking to an 0.1% solution of bromothymol blue in 50% aqueous ethyl alcohol (pH indicator range 6.0–7.6) in a quartz beaker, the color of the solution changed remarkably from yellow (acid) to blue (alkaline). However, when beads were added to a dilute solution of bromothymol blue in benzene in a quartz beaker, the color change of the solution was not observed, but slight color changes of the beads were noted. On the other hand, in the case of the silica powder, color changes were not seen. Hence, it is evident that (1) the surface of the bead is a weak base and (2) there are some alkaline components in the bead or alkaline contaminants mingling with beads which are easily soluble in ethyl alcohol or water. Such alkaline materials behave as accelerators for the reaction of the epoxy resin with the acid anhydride hardner.¹³

Composition S-1 shows the longest cure reaction time to reach 1000 poises. During this reaction time, the density increase of the fluid was only 0.015 g/cm³, from 1.140 g/cm³ immediately after mixing to 1.155 g/cm³ 160 min after mixing. Hence, the density of the filled compositions at 120°C can be regarded as constant during cure. Densities were calculated by applying the additive law where the value of 1.145 g/cm³ was used for the density of the fluid at 120°C.

Calculation Method

Assuming that all the particles in a composition have an equal diameter, the settling distances of the particles can be considered to be the same and are calculated from eq. (2) and the previously described physical values. For D_p in eq. (2), a median value of 57 microns of size range described by the supplier was used

for the beads, and a median value of 127 microns of two sieve-opening sizes was used for the silica powder. When calculating the settling distance of each specimen using eq. (2), it was integrated from $\eta_s = \eta_s(t_0)$ (where $\eta_s(t_0)$ is viscosity at t_0) to $\eta_s = \eta_s(T) = 1000$ poises; T is not equal to the gelation time, because it was concluded that the sedimentation of the filler particles in the mixture could be neglected over $\eta_s = 1000$ poises.

The theoretical filler content in the specified section of the specimen after settling was determined from the above settling distance. That is, if the settling distance is very small (consequently the height of each section is much greater than the settling distance), the filler contents in the sections, except for the top and the bottom sections, do not vary from place to place. The weight of a settling-in particle is equal to that of a settling-out particle in the same section. In these cases, the filler content is coincident with the feed concentration.

In the top sections the filler content decreases to $(h - H)/h$ of the initial value, where h is the height of section and H is the settling distance. The flow-in fluid, whose volume is equal to that of the settling-out particles, takes part in forming the clear liquid phase.

In bottom sections, the particles equaling those in the top sections settle into the bottom section, and the fluid is displaced by the settling-in particles. The displaced fluid, having the same volume as that of the settling-in particles, ascend higher.

From the above considerations, the theoretical filler contents of the top and the bottom sections after settling can be determined. Noticing the unit area in the horizontal section (the particle distributions in the horizontal directions are considered to be homogeneous), the theoretical filler contents (the weight fractions of the filler) in the top section, ϕ_t , and the bottom section, ϕ_b , can be calculated from eqs. (3) and (4), respectively:

$$\phi_t = \frac{f(h - H)}{h - fH(1 - d/d_p)} \quad (3)$$

$$\phi_b = \frac{f(h + H)}{h + fH(1 - d/d_p)} \quad (4)$$

where f is the feed concentration of the filler, h is the height of the section, H is the settling distance, d is the density of the suspending medium, and d_p is the density of the filler particle. In the cases where the settling distances are large, on the other hand, settling behavior is complicated, and it is discussed later.

Comparison Between Theoretical and Experimental Results

Figure 6 shows the relationship between the settling distance and the feed concentration. Figure 7 shows the relationship between the filler content change caused by sedimentation and the feed concentration in each section. Also shown in Figures 6 and 7 are the theoretically calculated results for the settling distances and the filler content changes. In Figure 7, the ordinate represents the height of specimens, and six equally divided sections of each specimen are indicated by the horizontal broken lines. The abscissa represents the filler content of each section, the vertical solid lines are the theoretical results, and the dotted lines are the experimental ones.

Figures 6 and 7 support the earlier considerations in which the settling distance

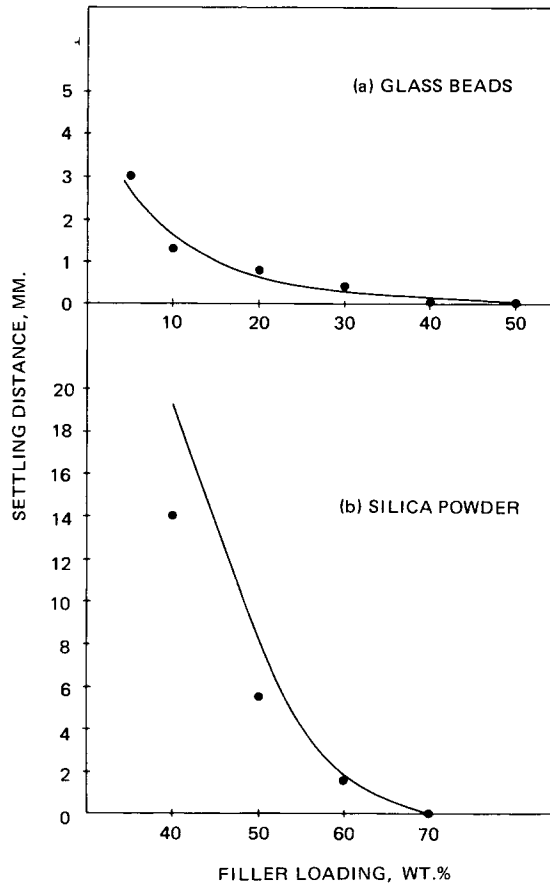


Fig. 6. Relationship between settling distance and feed concentration; (—) theoretical; (O) experimental.

was considered to be small. Especially in the cases of glass-bead filled compositions, experimental values agree well with the theoretical. In silica specimens S-3 and S-4 excellent agreement between the theoretical and the experimental results are also found.

Hence, in the compositions filled with narrow-distribution glass beads or the highly concentrated ones filled with the silica powder where the settling distances are small, the settling behavior of filler particles can be described quantitatively. In these cases the sedimentation should be regarded as the movement of the particle group in the fluid. The main reason for hindered settling of particles is the buoyancy and the increase in viscous resistance from the relative motion of the particles and the fluid.

But when the settling distances are large, the situations are complicated. That is, specimens S-1 and S-2 in Figure 6(b) show that the experimental values are always smaller than the theoretical, and the difference between them becomes greater with the decrease in the feed concentration. Similar phenomena are also observed for the filler content changes in the bottom sections of specimens S-1 and S-2 [Fig. 7(b)]. However, in the top sections the experimental values always tend to be larger than the theoretical, and the difference between them becomes greater with decrease in the feed concentration.

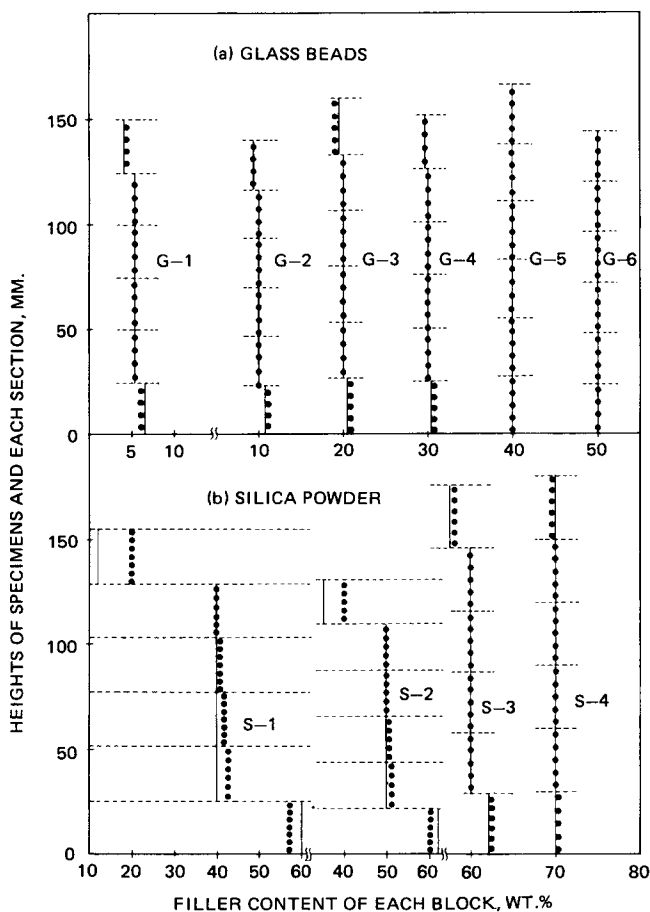


Fig. 7. Filler content change in specimens due to sedimentation during the curing process: (—) theoretical; (· · ·) experimental.

In addition, in three sections of specimen S-1, from the third to the fifth from the top, the experimental values are always larger than the theoretical, and the difference in the two values for the higher section is smaller than that for the next lower section. Similar phenomena are also noted in specimen S-2. Unclear boundaries between the clear liquid phase and the filled one were found in specimens S-1, S-2, and G-1. A semiclear liquid phase, where many small particles were distributed almost homogeneously, was observed only for specimen S-1.

Moreover, at the bottom of specimens S-1 and S-2, so-called compaction zones¹⁴ whose depths are approximately equal to the uppermost liquid phase were also observed clearly.

Incidentally, convection of the fluid (an ascending flow of the fluid is assumed), which results from the displacement of the fluid by settling particles as well as the settling velocity of a particle, increases as the feed concentration decreases.¹⁵ Apparently, this convective effect prevents the particles from settling because of the increasing viscous resistance. Then, the filtering action of the larger particles becomes weaker as the feed concentration decreases.¹⁴ Each particle, therefore, cannot substantially retain its position with respect to all its neigh-

boring particles, so that larger particles and smaller particles will settle separately and establish a concentration gradient in the suspension. Hence, it is easily inferred that the sedimentation of particles, in particular, relatively small particles, will be more influenced by the convective effect as the filtering effect becomes weaker (for reference, we call this phenomenon "a combination effect"). It is reasonable to assume that the above-described effects are enhanced in the case of the silica powder because of its broader particle size distribution [Fig. 3(b)].

The main reason for the differences between the experimental and the theoretical values in both the settling distance and the filler content change, as to each top section of specimens S-1 and S-2, can be attributed to the combination effect. Namely, the experimental value should deviate to the larger value because of the suppressed settling of the particles by the increasing convective effect with the decrease of the filtering effect. In these cases, medium and small particles will be greatly influenced, since the convective effect is strong and the filtering effect is weak. Moreover, it is expected that these effects are more noticeable for the silica powder since the particle size distribution is broad.

The unclear boundaries found in specimens S-1, S-2, and G-1 are due to the heterogeneity of the filler particle size and the weakened filtering effect with the decrease in feed concentration.

The semiclear liquid phase observed in specimen S-1 is the extended unclear boundary which results from the strongest combination effect. That is, here much smaller particles in the silica powder are not only settled separately but are also prevented from settling by the strongly convective fluid. This situation prevails until the reaction of the fluid develops and the sedimentation is finished, since the filtering effect is the weakest and the convection of the fluid is the strongest of all compositions investigated here. The discrepancies between the theoretical and the experimental results for both the settling distance and the filler content change *increase* as the feed concentration *decreases*. The semiclear liquid phase is observed only in specimen S-1. The above effects become noticeable as the feed concentration decreases and the particle size distribution broadens.

On the other hand, in the bottom sections of specimens S-1 and S-2, the difference is attributable mainly to the strong ascending flow of the fluid resulting from the displacement of the fluid by the settling particles at the bottom (for reference, we call this phenomenon "a bottom effect"), because the formation of a compaction zone was found in each specimen. The fact that the difference increases as the feed concentration decreases demonstrates that the ascending flow is also enhanced with decrease in feed concentration.

Moreover, it should be noted that this ascending flow becomes weaker and finally becomes constant as the position in height in the specimen is further from the bottom. The difference between the ash content and the feed concentration of a higher section among those from the second to the fifth sections in specimen S-1 becomes smaller and eventually tends toward zero. That is, the weaker the ascending flow as the position in the specimen becomes higher, the less will be the difference between the settling-in particles and the settling-out particles in the same section. As a result of more settling-in particles, the experimental value is always larger than the theoretical. In the second section, the difference disappears because of the constant convective effect. The sections where the

above-mentioned differences were found correspond to the so-called compression zones.¹⁵ Clearly, the main reason for the establishment of the compression zone should be the bottom effect, although the ascending bottom, caused by the formation of the compaction zone which gradually increases, might be partly responsible.

CONCLUSIONS

The settling behavior of fillers such as glass beads and silica powder in thermosetting epoxy casting resins was investigated quantitatively in connection with the curing process, using a modified Stokes' law equation.

The modified Stokes' law equation was derived based on the assumptions that a suspension (a thermosetting epoxy casting resin) is regarded as a homogeneous fluid, and then treating the settling phenomena such that one or a few particles, which are essentially identical to the particles contained in the suspension, settle into the homogeneous suspension under gravity. The equation considers the effect of viscosity increase of the fluid during cure by the use of an integral form.

Excellent agreement was found between the experimental results and the theoretically calculated ones with respect to the compositions filled with narrow-distribution glass beads and compositions highly filled with silica powder where the settling distances were small. In these cases the sedimentation should be regarded as the movement of the particle group in the fluid, and the main reason for hindered settling of particles is buoyancy and increase in viscous resistance.

When silica powder is involved and the feed concentrations are low, that is, when the particle size distribution is broad and the settling distances are large, such as in specimens S-1 and S-2, the settling behavior is understandable by considering various effects such as convective, filtering, combination, and bottom effects, and the effect of the broad particle size distribution of the silica powder. These effects tend to prevent particles (in particular, smaller particles) from settling as the feed concentration decreases. The bottom effect, which causes the formation of a compression zone, becomes weaker as the position in height in the specimen is farther from the bottom. In addition, a compaction zone was clearly found in specimens S-1 and S-2.

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