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Communications

Functionally Graded Polymer Composites : Simulation of Fiber Distribution†

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Abstract : Centrifugation is a method to create functionally graded materials (FGM) with a thermosetting matrix. In this study the movement of short carbon fibers in an epoxy resin during the centrifugation process was modeled to determine the fiber distribution in the final product. For this purpose a form factor K was introduced to modify a set of equations that was previously shown to be valid for the motion of spheres. It was shown that the results of the simulation were in good agreement with the experimental data, when an empirical K factor of four was chosen.

Keywords : functionally graded polymer composites, computer simulation, fiber distribution.

Introduction

Functionally graded materials (FGM) are a new type of materials which exhibit a continuous compositional and/or micro-structural variation in one or more spatial directions. This results in special characteristics such as effective thermal stress relaxation, adhesive properties and so on. The new material concept has been initially studied in association with the development of super heat-resistant materials based on ceramics matrices.[1-3] Since then there have been a number of papers on polymer matrix FGMs with fibers or spherical fillers to deepen an understanding of the relationship between structure and properties of FGMs manufactured by the centrifugation technique.[4-9]

Recently Watanabe *et al.*[10] studied the motion of ceramic particles in a molten metal matrix under centrifugal force by numerical modeling. The objective was to confirm the formation process of composition gradients and to conclude that computer simulation was useful to predict the graded composition. Klingshirn *et al.*[11] tried to apply this computer simulation technique for the system of small glass beads in an epoxy resin matrix. They concluded that the results of the simulation were in reasonable agreement with the experimental ones.

In this study the experimental results of epoxy/short carbon fiber systems[12] were taken to test the validity of the simulation technique. Unlike spherical particles, carbon fibers do not have a definite diameter, which can be applied to the equation describing the motion of fillers in a viscous medium under centrifugal force. Therefore some modification of the controlling equation for the computer simulation was done, and the usefulness of this modification was discussed.

Analytical Procedure

It is known that the velocity of the motion of spherical particles under centrifugal force can be described by equation (1)

$$v = \frac{(\rho_f - \rho_m) G g D_c^2}{18\eta} \quad (1)$$

where v is the velocity of particles, ρ_f and ρ_m are the density of the particles and the matrix, respectively, G is the ratio of centrifugal force to gravity, and g is the gravitational constant. In addition, D_c represents the particle diameter, and η is the local viscosity of the filler modified matrix, which, in turn, is a function of the local filler content. This equation is valid for spherical particles. For the fillers with an aspect ratio other than one a form factor had to be introduced. In this study $D_c' = KD_c$ was considered, where K is a form factor which may change with the aspect ratio of the fibers. Accordingly D_c should be replaced in equation (1) by D_c'. The Brinkman equation[13] (2) was considered to estimate the viscosity change of the medium as a function of the fiber content:

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$$\eta = \frac{\eta_0}{(1 - V/V_{max})^{2.5}} \quad (2)$$

where η_0 is the viscosity of the polymer matrix without fibers, V is the fiber volume fraction, and V_{max} is the maximum packing fraction of the fibers. V_{max} was determined as the maximum filler content of a sample centrifuged under very severe conditions.

The simulation was based on these two equations, i. e., equation (1) modified by D_c' and equation (2), in order to estimate the one-dimensional motion of the fibers parallel to the direction of the centrifugal force. The volume of the samples was divided evenly into very thin layers, and then the travel of fibers from one layer to the next was calculated for very short periods of time simultaneously for all layers. With the new distribution of fibers the process was repeated by using newly calculated local viscosities (with reference to equation(2)), until the sum of time intervals reached the centrifugation time.

Results and Discussion

Two sets of graded carbon fiber/epoxy resin samples from an earlier study[12] were considered for the comparison with the results of the simulation. These groups were classified according to the holding time, i. e., the time interval from the beginning of the mixing of the epoxy resin with the curing agent to the start of the centrifugation (Table I). Experiment groups (a) and (b) had a holding time of 30 min. and 60 min., respectively. The deviation of the viscosity value in the same group should be attributed to the fluctuations of temperature during the measurements, which resulted in different degrees of chemical reaction of the room-temperature-curing epoxy resin system.

Table I. Two Groups of Epoxy/Carbon Fiber Products with Different Holding Time

Product No.	Turning Speed ω [rpm]	η_0 [Pa • s] (a) holding time of 30 min.	η_0 [Pa • s] (b) holding time of 60 min.
1	1,000	1.81	7.00
2	2,000	1.45	4.24
3	3,000	1.65	6.77
4	4,000	1.60	5.63

Figures 1 and 2 show the results of the computer simulation

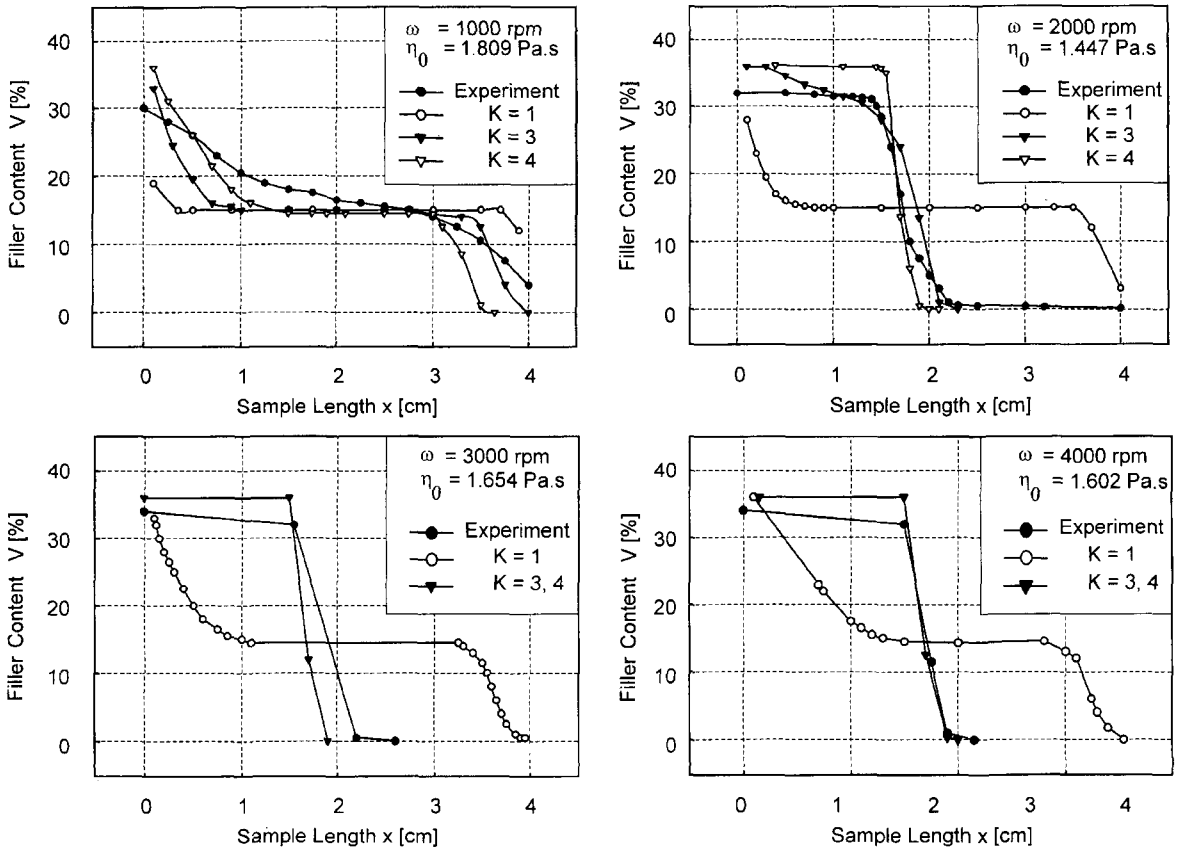

Figure 1. Comparison of the fiber distributions from the experimetal group (a) with the corresponding simulation.

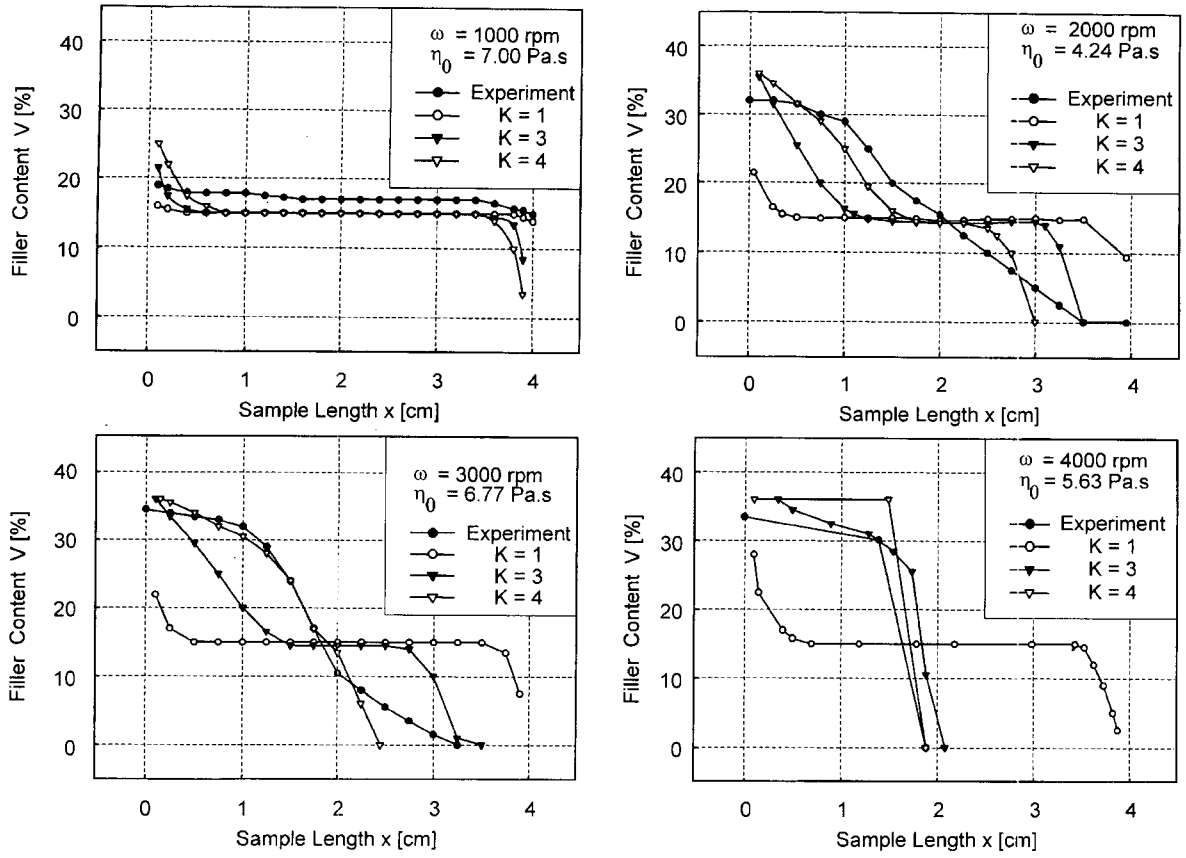

Figure 2. Comparison of the fiber distributions from the experimetal group (b) with the corresponding simulation.

for the experimental groups (a) and (b), respectively, depending on the turning speed in comparison with the experimental ones. In this calculation the initial volume fraction of fibers was 15% and the maximum packing fraction of fibers, V_{max} was determined to be 36% by volume (according to the fiber fraction after centrifugation of a sample at 5,000 rpm over a period of 20 min). The aspect ratio of the carbon fibers used (M-101S, Kureha Chemical Industry) amounted to $L/d = 9$ (L = average fiber length, d = average fiber diameter).

The figures indicate irrespective of holding time, i.e., initial viscosity that the experimentally determined gradients in filler volume fraction V over the specimen distance x (starting with 0 at the outer side of the sample) come more and more close to the calculated values (according to the modified equation (1)) with the change of K-value from 1 to 4. Around the K-value of 4 the computer simulation represents quite well the experimental results over the whole experimental range of turning speeds and resin viscosities. This implies that fibers with an aspect ratio of around 9 perform in a similar way as spheres with an apparent diameter, which is 4 times larger than spheres (index sp) with a diameter being of the same size as the actual fiber (index f) diameter. On the other hand, the K-value should be connected to the dimensional parameters of fibers. The ratio of the actual volume reaches a value of 13.5 ($V_f = 9/4\ \pi d^3$ and $V_{sp} = 1/6\ \pi d^3$) and the ratio of the actual surface area is 9.5 ($A_f = 9.5\ \pi d^2$ and $A_{sp} = \pi d^2$) between fibers and equivalent spherical particles, but this simple consideration in relation to the aspect ratio does not give any hint to the relationship of these parameters with the K-value. More intensive studies should be carried out for a wider range of fibers with different aspect ratios in order to deduce a physical meaning for the K-value.

Conclusions

A form factor K was introduced to the equation, modeling the motion of spherical particles in a viscous medium under centrifugal force, in order to describe the movement of fibers under similar conditions. This modified equation was used for the simulation of the movement of fibers during the centrifugation process. The fiber distributions resulting from the calculations were compared with experimental data. For a K-value of 4 the results of the simulation were in good agreement with the experimental ones, implying that the equivalent diameter of fibers with an aspect ratio of 9 is four times larger than the actual diameter of the fibers. It

was concluded, however, that more intensive studies should be carried out for the fiber systems with different aspect ratios to produce a systematic and physically meaningful value for the form factor K.

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