Study on the Interfacial Properties of Two-Dimensionally Arranged Glass Fiber/Epoxy Resin Model Composites

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ABSTRACT: The effect of interfiber distance on the interfacial properties in two dimensional multi-E-glass fiber/epoxy resin composites has been investigated using fragmentation test. In addition, the effect of the fiber surface treatment on the interfacial properties has been studied. We found that the interfacial shear strength decreased with the decreasing interfiber distance at the range of <50 μm and the extent of the decreasing was more serious as the increasing of the number of adjacent fiber. This is probably that the interface between the fiber and the resin was damaged by the breaking of adjacent fibers and the damage increased with minimizing the interfiber spacing and the number of adjacent fibers. We can guess that interfacial shear strength in real composites is much smaller than that of multifiber fragmentation sample with touched fiber. When the interfiber distance was >50 μm, the interfacial shear strengths were saturated regardless of fiber surface treatment and were in close agreement with those of the single fiber fragmentation test. Finally, the interfacial shear strength evaluated using two dimensional fragmentation tests are shown as real values in-site regardless of fiber surface treatment, interfiber distance, and existing matrix cracks. J Appl Polym Sci 99: 1541–1551, 2006

Key words: interfiber distance; interfacial properties; two-dimensional fragmentation test; matrix crack; damage

INTRODUCTION

It is well known that external stresses were transferred from resin to fiber through the interface in fibrous reinforced composites and most of the external load is carried by the fibers. So the interface between the fiber and the matrix resin is the heart of the fibrous composites. And the exhibition of original fiber performance depends largely on the interfacial properties of the composites.

The interface in fibrous reinforced composites plays a very important role in determining the final performance of a composite. Especially, the interfacial shear strength is one of the most fundamental factors in evaluating the mechanical properties and durability of the fiber-reinforced composites in the specific environment.[1]

If interfacial shear strength in fibrous composites is too low, it is hard to expect that the performance of reinforcing fiber is reflected in composites, even when high performance fiber is used.[2] On the other hand, if the interfacial shear strength is too high, there is a possibility of a decrease in fracture toughness of composites because of the poor resistance against crack propagation.[3] Therefore, it is necessary that the interfacial shear strength of the fiber-reinforced composites is controlled in accordance with the expected performance of material.

In principle, the interfacial shear strength can be controlled by the suitable combination of fiber, matrix resin and fiber surface modification, etc. It is very important to evaluate exactly the interfacial shear strength thus controlled. In the last few decades, several techniques such as the pull-out,[4–7] microbond,[8–12] fragmentation,[13–20] and indentation[21] test methods have been developed to measure the interfacial shear strength correctly.

Among them, one popular technique is single fiber fragmentation test. The merits of this method are that a lot of data are generated from one sample test, and the preparation of the sample is easy comparing the other techniques. But it takes about 5 h to test a single sample. To overcome this problem, new approaches have been developed. Many works[13,22–27]have been done exploring the effects of testing multiple fibers in a fragmentation test.

Most of the current multifiber research have focused on using laser Raman spectroscopy (LRS) as a detection tool for directly measuring the strain in broken and unbroken fibers.[28–40] In particular, the magnitude and location of the overstress region in the fibers adjacent to the broken fibers has been cited as a critical fiber–fiber interaction effect that controls the initial composite failure process. However, this technique is

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restricted to aromatic fibers that are Raman active. Hence, this detection technique is not applicable to glass fibers.

We have investigated the effect of interfiber distance on the interfacial properties in E-glass fiber/epoxy resin composites using single- and two-fiber fragmentation test. And we found that when the interfiber distance was more than 50 μm, interfacial shear strength was the same as that measured by the single-fiber fragmentation test and when the interfiber distance was less than 50 μm, the interfacial shear strength decreased with the decreasing interfiber distance.

The objective of this paper is to study whether the results obtained at previous work might be applied in two-dimensional fragmentation test.

In this paper, multifiber fragmentation test specimens (two–dimensional) were fabricated. We have investigated the effect of interfiber distance on the interfacial properties multi-E-glass fiber/epoxy resin composites arranged in two-dimension. In addition, the effect of surface treatment on the interfacial properties has been studied.

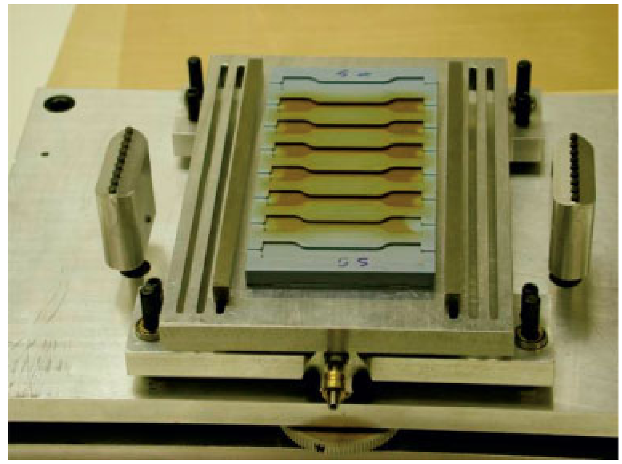


Figure 1 Apparatus used for arranging fiber in the preparation of multifiber fragmentation test specimen. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

EXPERIMENTAL

Materials

The materials used in this study are as follows. The fibers used were sized and desized E-glass fiber(Owens–Corning). Sized fiber was coated with 3-aminopropyltriethoxysilane (A-1100) and desized fiber meant the sized fiber washed by distilled water at 50°C for 24 h. And the average diameter of glass fiber was 13 ± 1.23 μm. The matrix resin used was epoxy-(diglycidl ether of bisphenol A, DGEBA, Epon 828, shell co.). The hardener used was *meta*-phenylene diamine (*m*-PDA, 14.5 phr, Fluka Chemical Co., Japan).

Preparation of fragmentation test sample

The sample preparation for single and multifiber testing was similar to that described by Drzal et al.[14] Figure 1 shows the special tool used to arrange interfiber distance of multifiber fragmentation test sample in this study.

Fragmentation test

Samples were marked a standard gauge length of about 10 mm to measure real strain by permanent pen (blue color). When the sample was installed, we adjusted that the grip was not much tight. The sample was loaded in tension by the sequential application of step strain. The total number of strain step was 28 and the total strain was 2.4 mm. The strain rate was 85 μm/s and the average deformation at each step was 85.7 μm through the whole sample length between the sample holders at both sides. The delay time between the application of successive step strains was 10 min. After 28 steps, the sample was unloaded and every fiber fragment length within gauge length was measured.

Microscope observation

Optical microscope with polarized transmitted light was used to observe interfacial properties between fiber and resin in the fragmentation test. The cover glass and silicone oil with refraction index of approximately 1.6 were used to improve image clarity. Refraction index of the oil was almost same as that of the matrix resin used in this study. The oil should flow to fill all contact area between the specimen and the cover glass.

Interfacial shear strength calculation

We calculated the interfacial shear strength using the following eq. (1).[14] The distribution of fiber fragment lengths have been satisfactorily described by a two-parameter Weibull analysis, and the expression for the interfacial shear strength τ is determined as,

$$\tau = \sigma\Gamma(1 - 1/\alpha)/2\beta \quad (1)$$

Where α and β are the shape and scale parameters, respectively.

Γ represents the Gamma function. In eq. (1), σ is the average fiber tensile strength at the critical fiber length needed to calculate interfacial shear strength. However, in this equation, we used the single fiber tensile strength at 20 mm of gauge length. One goal of this

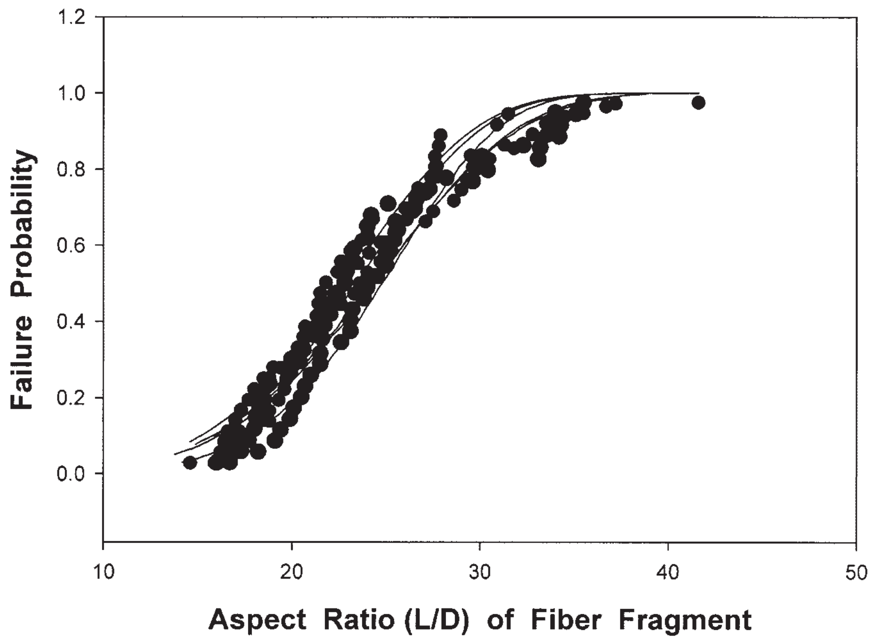


Figure 2 Failure probability versus aspect ratio of fiber fragment in the sized single fiber fragmentation test.

study is to discuss the effect of interfiber distance on the interfacial properties in fragmentation test.

RESULTS AND DISCUSSION

Single fiber fragmentation test

The tensile strengths of sized and desized E-glass single fibers were 2.10 ± 0.57 and 1.63 ± 0.44 GPa, respectively. The tensile strength of sized fiber is shown to be stronger than that of desized fiber. This is probably due to the fact that desized fiber can easily get damaged when they rub against each other during processing operations. As described in the Experimental section, desized fiber was subjected to one more process compared with sized fiber. This is also one of the factors to reduce the tensile strength of desized fiber. Generally it is known that the most important factors determining the tensile strength of the glass fiber is the damage of fiber surface and processing history.

Figure 2 represents the plot of a failure probability as a function of aspect ratio of the fiber fragment in the sized single fiber fragmentation test. Solid lines are plotted by the Weibull distribution equation with two parameters. We can see that experimental values and Weibull distribution equation with two parameters are in good accordance. It is shown that the range of aspect ratio is 15–37.

Figure 3 shows the plot of a failure probability as a function of aspect ratio of the fiber fragment in the desized single fiber fragmentation test. Solid lines are plotted by the Weibull distribution equation with two parameters. We can see that the experimental values and the Weibull distribution equation with two parameters are in good accordance. It is shown that the range of aspect ratio is 12–33. The comparison of the aspect ratios of different kinds of fibers used in fragmentation test is not much important because the aspect ratios depend on various parameters such as the fiber tensile strength, diameter, and interfacial shear strength.

Figure 4 represents the normalized number of fiber breaks as a function of applied strain in sized and desized single E-glass fiber/epoxy resin fragmentation test. The normalized number of fiber breaks is obtained by dividing the number of fiber breaks at each strain by the number of the total fiber breaks at saturation within gauge length of about 10 mm. We can see that after the number of desized fiber breaks was saturated, the number of sized fiber breaks was also saturated. Also we can see that the extent of sized fiber strain from initial fiber break to saturation was smaller than that of desized fiber. And the strains of initial fiber break were about 2 and 3% of desized and sized fiber, respectively.

As a result, the interfacial shear strengths of sized and desized single fiber fragmentation were 46.70 ± 2.59 and 40.69 ± 2.60 MPa, respectively.

Multifiber fragmentation test

Figure 5 represents the plot of interfacial shear strength versus interfiber distance in the multi-E-glass fiber/epoxy resin fragmentation test. We employed only good samples with equal interfiber distance. It

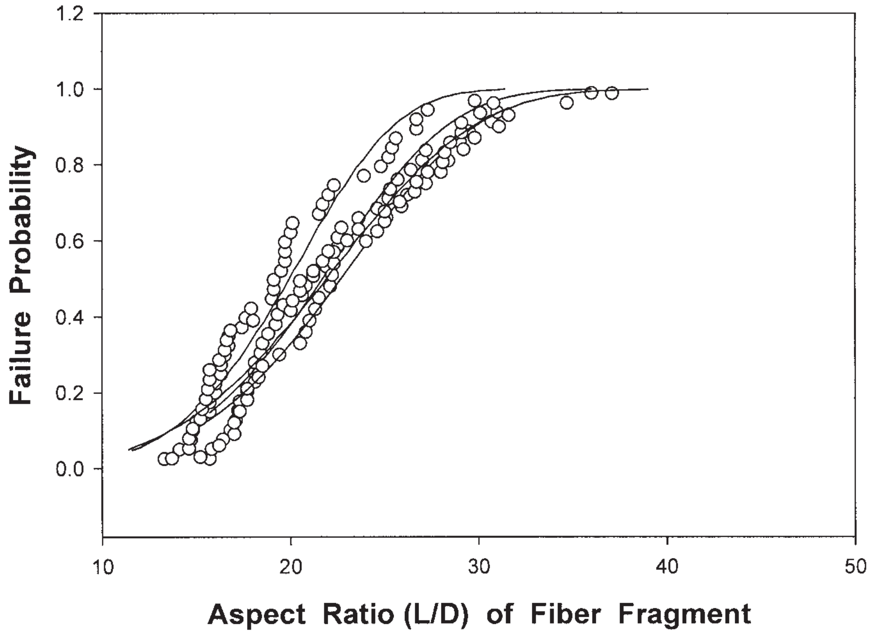


Figure 3 Failure probability versus aspect ratio of fiber fragment in the desized single fiber fragmentation test.

was shown that the interfacial shear strength increased until interfiber distance became about 50 μm, and then was saturated regardless of sizing.

In other words, in case the interfiber distance was less than 50 μm, the interfacial shear strength decreased with the decreasing interfiber distance, and the extent of the decreasing was more serious as the increasing of the number of adjacent fibers. This is probably due to the fact that the interface between the fiber and the resin was damaged by the breaking of adjacent fiber, and the damage increased with the closing of interfiber spacing and the number of adjacent fiber. We considered interface was damaged and become weak by breaking of the adjacent fiber, and then fiber was broken within damaged interface sequentially. We can guess interfacial shear strength in real composites is much smaller than that of multifiber fragmentation sample with touched fiber.

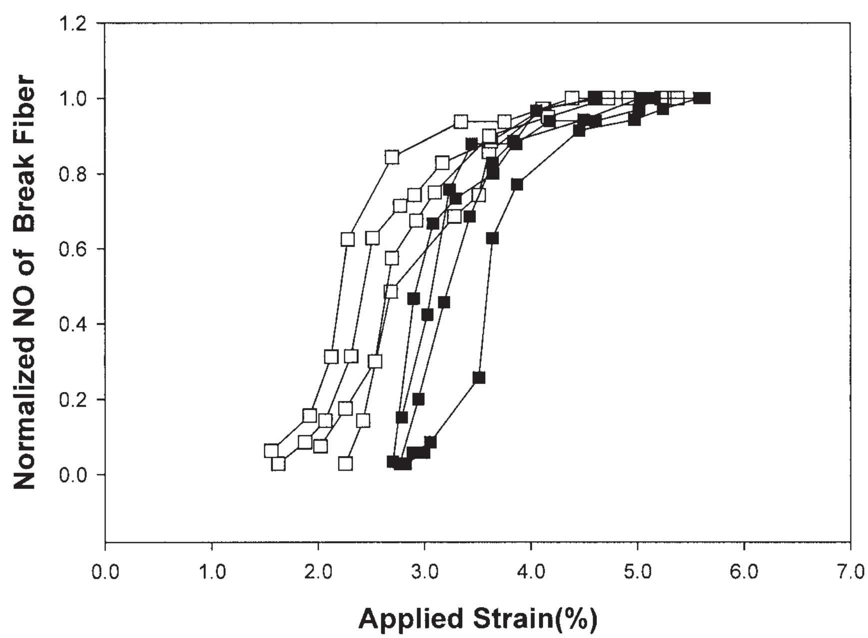


Figure 4 Plot of normalized number of fiber break as a function of applied strain in fragmentation test of single E-glass fiber/epoxy resin (solid, sized; open, desized).

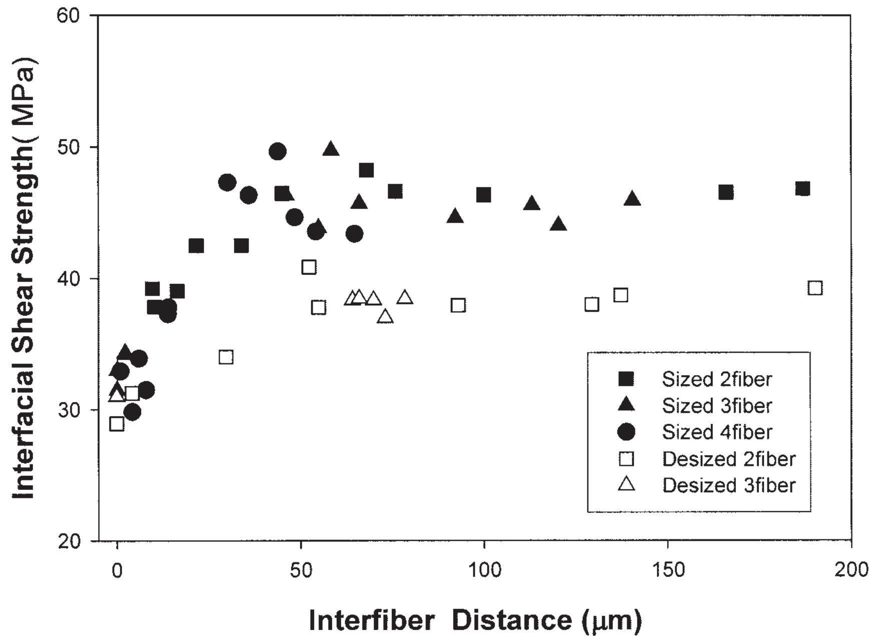


Figure 5 Interfacial shear strength versus interfiber distance in the fragmentation test of the different fiber number.

It was seen that the interfacial shear strengths saturated when the interfiber distance was over 50 μm, regardless of fiber surface treatment and were in close agreement with those obtained by the single fiber fragmentation test.

The damaging factors considered because of the breaking if fibers are strain energy release, stress transfer, stress concentration, etc. The extent of stress concentration depends mainly on existing matrix crack in case of sized fiber sample. Therefore, it was shown that when the interfiber distance is small, the decreasing of interfacial shear strength in sized fiber fragmentation was more serious than the decreasing of that in the desized fiber fragmentation test.

Figure 6 shows the plot of the failure probability as a function of aspect ratio of the fiber fragment in the

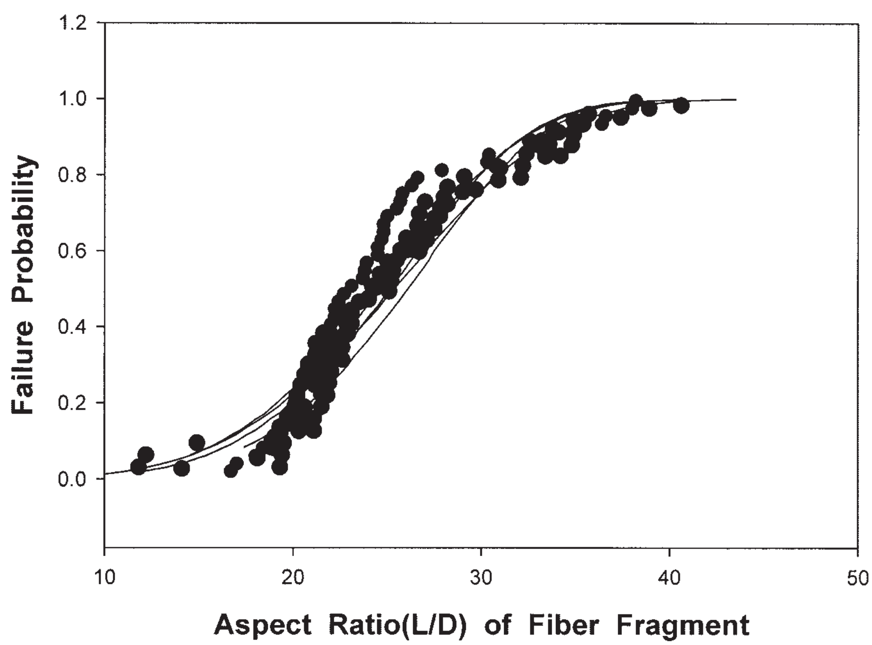


Figure 6 Plot of failure probability versus aspect ratio of fiber fragment in the fragmentation test of two-dimensionally arranged sized multi-E-glass fiber/epoxy resin.

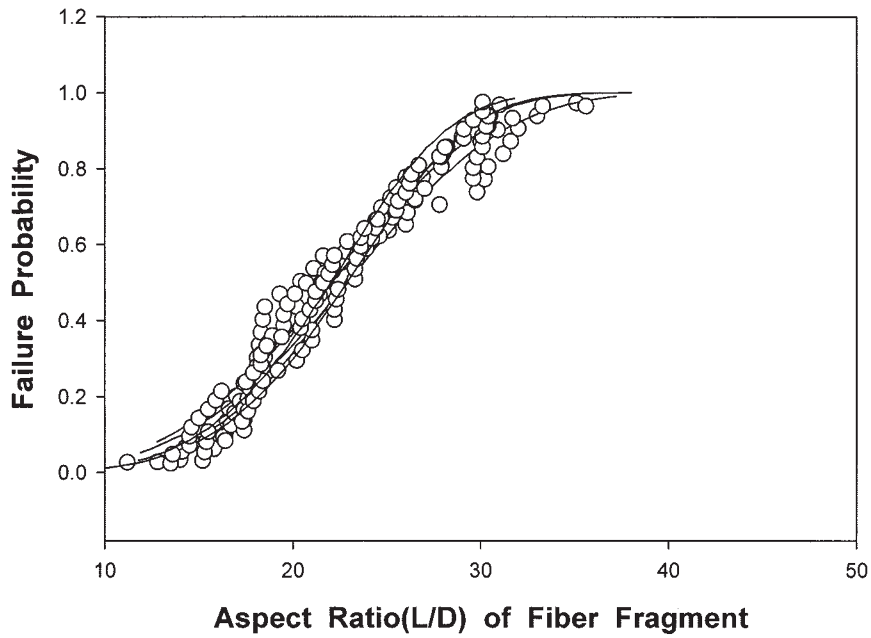


Figure 7 Plot of failure probability vs. aspect ratio of fiber fragment in the fragmentation test of two-dimensionally arranged desized multi-E-glass fiber/epoxy resin.

fragmentation test of sized multi-E-glass fiber/epoxy resin (two-dimensionally arranged). We used only proper samples with over 50 μm of interfiber distance (Fig. 12). Solid lines are plotted by the Weibull distribution equation with two parameters. We can see that experimental values and two parameters Weibull distribution equation are in good accordance. It is shown that the trend is very similar with the result of sized single fiber fragmentation as shown in Figure 2.

Figure 7 shows the plot of a failure probability as a function of aspect ratio of the fiber fragment in the fragmentation test of desized multi-E-glass fiber arranged two dimension/epoxy resin. We used only proper samples with over 50 μm of interfiber distance (Fig. 13). Solid lines are plotted by the Weibull distribution equation with two parameters. We can see that experimental values and two parameters Weibull distribution equation are in good accordance. It is shown that the trend is very similar with the result of desized single fiber fragmentation as shown in Figure 3.

Figure 8 reveals the normalized number of fiber breaks as a function of applied strain in fragmentation test of sized multi E-glass fiber arranged in two-dimension/epoxy resin. We can see that the initial fiber crack began to occur at strain of about 3%, the number of fiber breaks was saturated at strain of 6%. Also we can see that the results are very similar with that of sized single fiber fragmentation test as shown in Figure 4.

Figure 9 represents the normalized number of fiber breaks as a function of applied strain in fragmentation test of desized multi E-glass fiber arranged in two-dimension/epoxy resin. We can see that the initial fiber crack began to occur at strain of about 2%, the number of fiber breaks was saturated at strain of 6%. Also we can see that the results are very similar with that of desized single fiber fragmentation test as shown in Figure 4.

From above-mentioned, we can conclude that when interfiber distance is >50 μm in the two-dimensional multifiber fragmentation test, the trends of plots of failure probability and aspect ratio of fiber fragment, normalized number of fiber crack, and applied strain are same as the results of single fiber fragmentation test.

Figure 10 reveals the polarized transmitted light micrographs of desized multi-E-glass fiber/epoxy resin fragmentation test at saturation. We can see that interfiber spacing was not uniform. In fact, the preparation of good sample with equal interfiber distance is extremely difficult. We could arrange the fiber with equal interfiber spacing using a special tool as shown in Figure 1. However, there were many factors to disturb the arranged fiber during processing such as putting down the arranged fibers on the long rectangular stick, gluing, pouring the resin, and curing.

In Figure 10, we can see that when the interfiber distance is small, the stress distribution pattern is shown like that of a fiber with bigger diameter and when the interfiber distance is large, the stress distribution pattern is independent of between-fiber breaks.

And from the Figure 5, we can easily see that when the interfiber distance was >50 μm the interfacial shear strength became independent value. Therefore

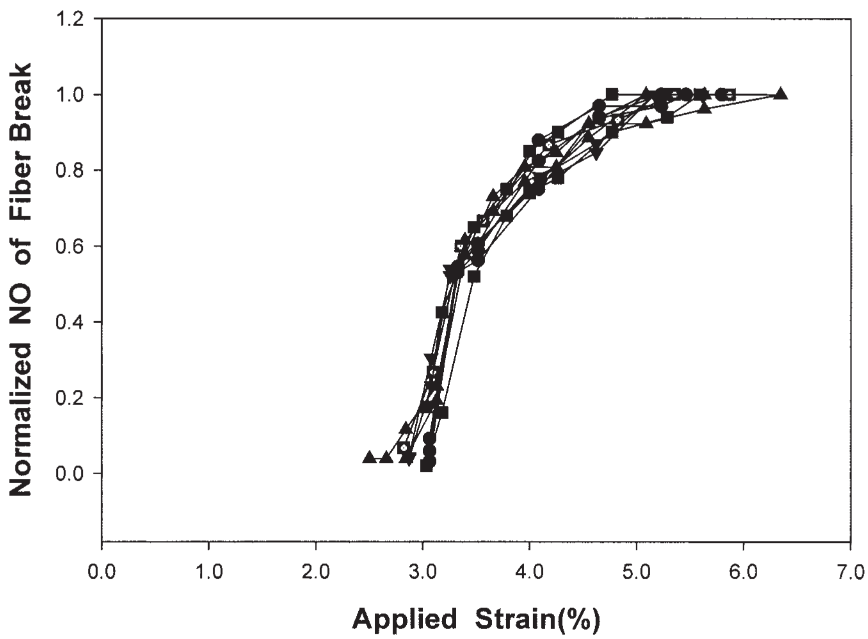


Figure 8 Plot of normalized number of fiber break as a function of applied strain in fragmentation test of two-dimensionally arranged sized multi-E-glass fiber/epoxy resin.

when we analyzed the results of two dimension fragmentation test, if the interfiber distance was at least over 50 μm, we considered nothing the effect of interfiber distance on the interfacial shear strength. For example, in Figure 10, we used the interfiber distances of 0, 17, and 154.9 μm from the upper part of the figure respectively. Unless otherwise noted, interfiber distance means the same as that mentioned previously.

Figure 11 represents the plot of interfacial shear strength versus interfiber distance in the two-dimensional multi-E-glass fiber/epoxy resin fragmentation test. It was shown that the interfacial shear strength increased until the interfiber distance became about 50 μm, and then were saturated regardless of sizing. In the whole region, we can see the interfacial shear strength of sized fiber was bigger than that of desized

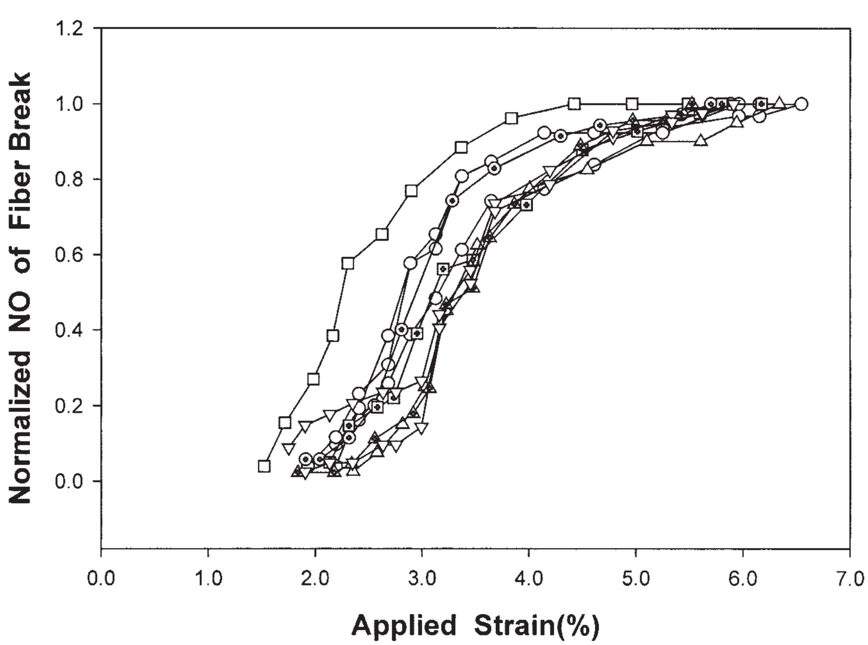


Figure 9 Plot of normalized number of fiber break as a function of applied strain in fragmentation test of two-dimensionally arranged desized multi-E-glass fiber/epoxy resin.

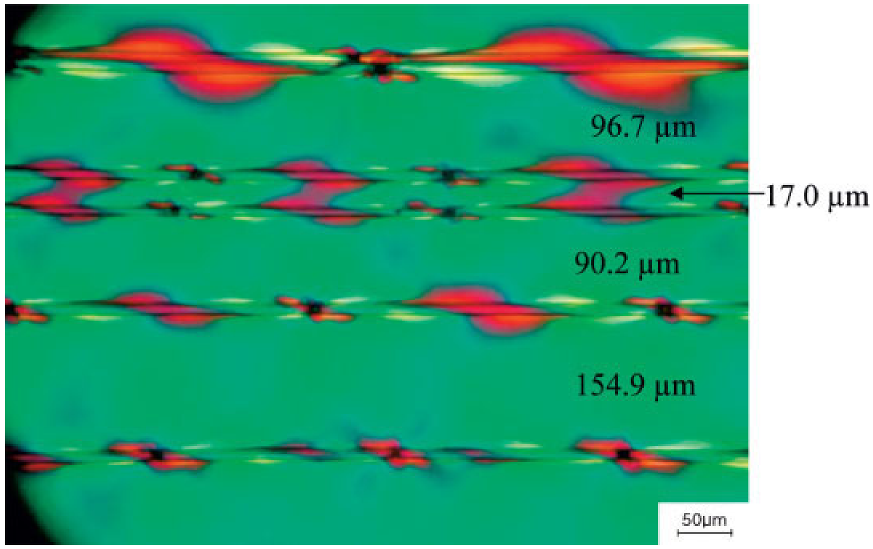


Figure 10 Polarized transmitted light micrograph of the desized E-glass fiber/epoxy resin fragmentation test at saturation. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

fiber. But in case of the interfiber distance <50 μm, we can see that some of the interfacial shear strengths of sized fiber were smaller than those of desized fiber. This is probably the effect of matrix crack as previously mentioned damaged mechanism. We described there were matrix cracks in the sized fiber fragmentation test specimen. In fact, it is hard to decide the interfiber distance in the fragmentation test specimen having matrix crack correctly.

Figure 12 shows the polarized and nonpolarized transmitted micrographs of fiber fragment in the fragmentation test of sized multi-E-glass fiber arranged in two dimension/epoxy resin. Figure 12(a) shows the polarized photo and 12(b) is enlarged photo of (a).

From this figure, we can see that all fiber was arranged very well and the stress distribution patterns and the locations of fiber break were also independent. And we can easily see that some of sized fiber had fiber breaks with matrix cracks like bat shape and the fiber surface with matrix cracks was not smooth(2nd fiber of Fig. 12(b)). We considered that rough surface of fiber could become one of the reasons to make matrix cracks.

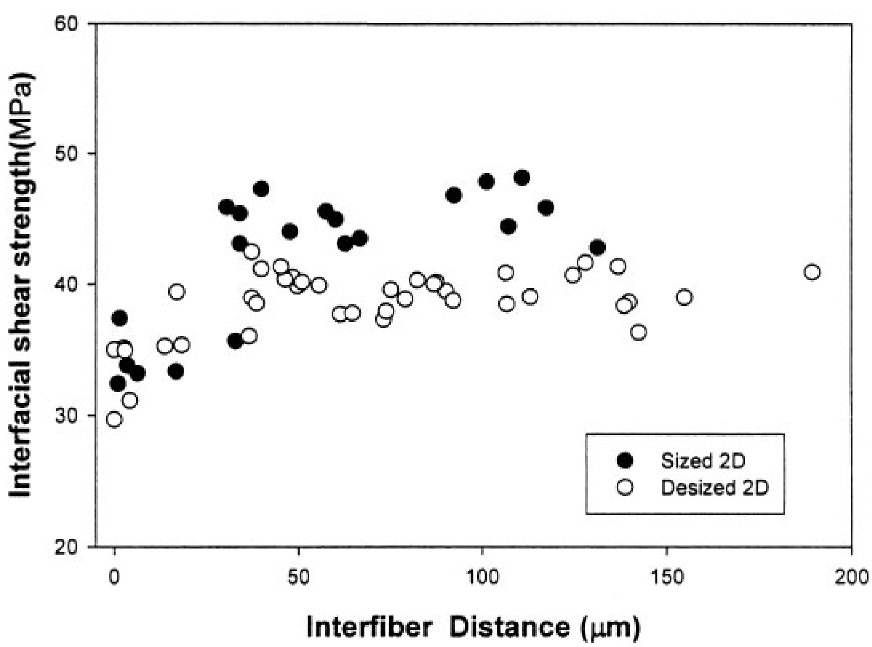
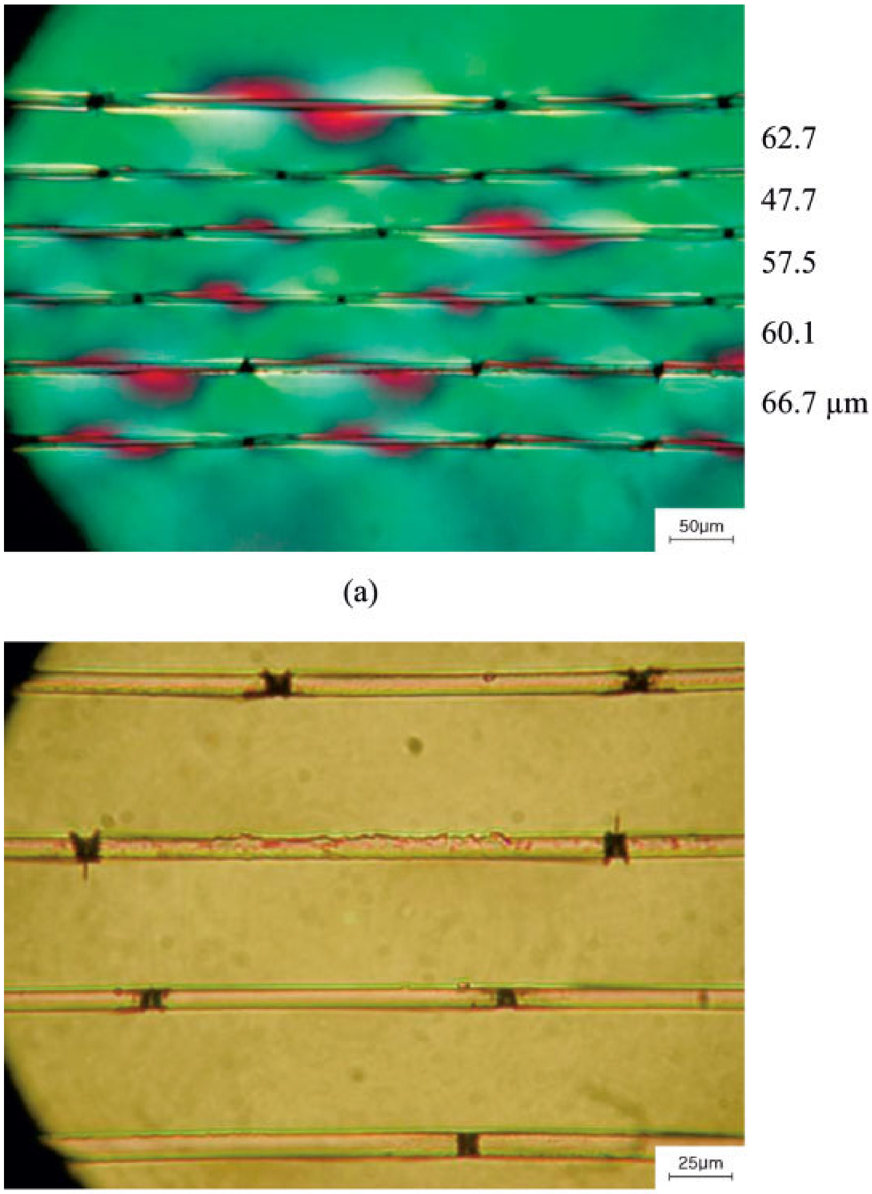


Figure 11 Interfacial shear strength versus interfiber distance in the fragmentation test.



(b)

Figure 12 Polarized and nonpolarized transmitted micrographs of fiber fragment in the fragmentation test of two-dimensionally arranged sized multi-E-glass fiber/epoxy resin. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

Figure 13 shows the polarized and nonpolarized transmitted micrographs of fiber fragment in the fragmentation test of desized multi-E-glass fiber arranged two dimensions/epoxy resin. Figure 13(a) shows the polarized photo and 13(b) is the enlarged photo of (a).

From this figure, we can see that all fibers were arranged very well and the stress distribution patterns and the locations of fiber break were also independent. And we can easily see that the surface of desized fiber was smooth and there were no matrix cracks.

In Figure 12 and 13, we could gain that each fiber of those samples has very similar interfacial shear strength with that obtained by single fiber fragmentation test as mentioned previously. From Figures 11, 12, and 13, we could conclude that if interfiber distance becomes >50 μm in the two-dimensional fragmentation test, the result is the same as that of the single fiber fragmentation test, regardless of fiber surface treatment.

From above-mentioned, we can conclude that in the multifiber fragmentation test, two dimensional fragmentation test shows the real values of the interfacial shear strength, in spite of any case. When interfiber distance is above 50 μm, the interfacial shear strength

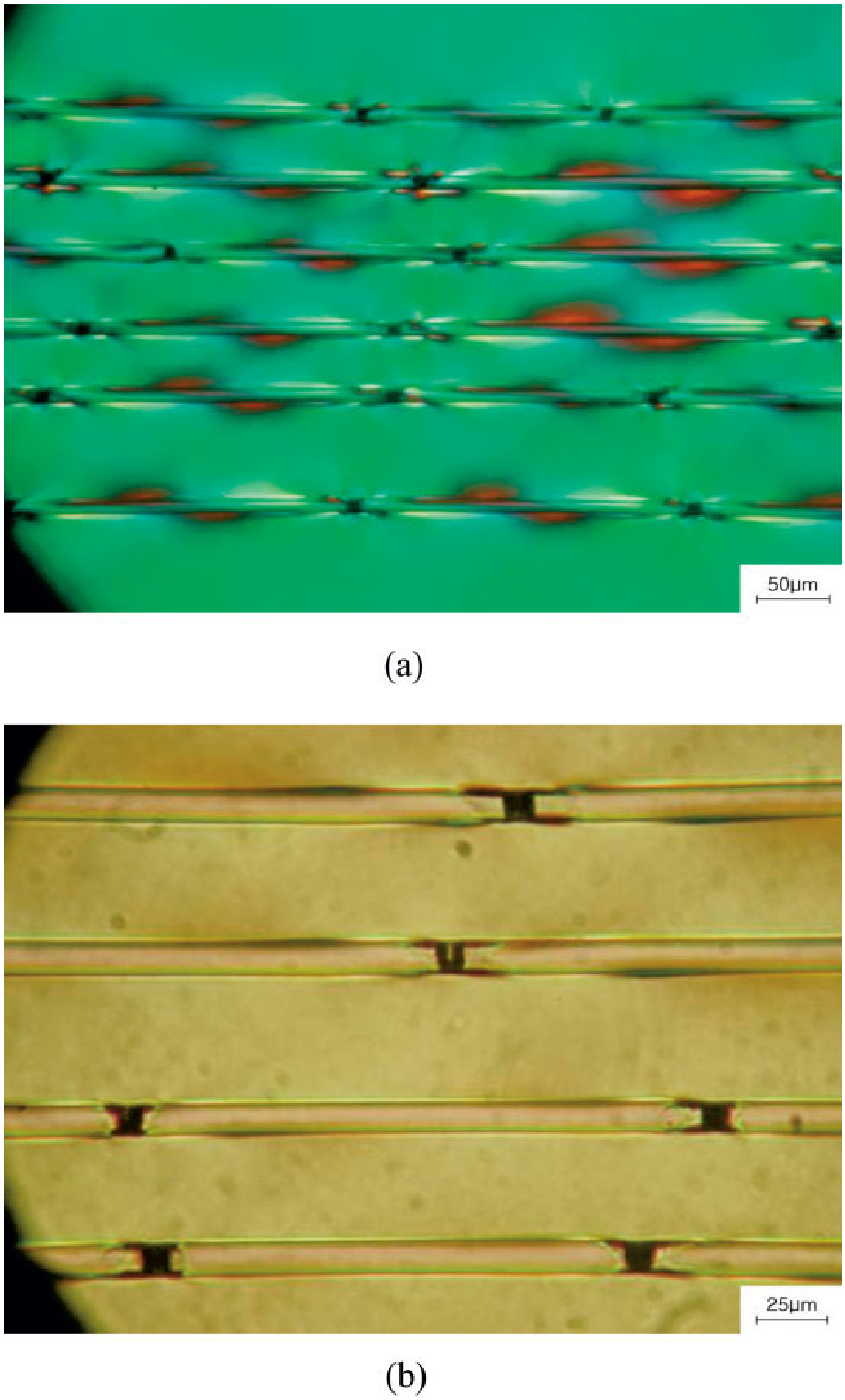


Figure 13 Polarized and nonpolarized transmitted light micrographs of fiber fragment in fragmentation test of two-dimensionally arranged desized multi-E-glass fiber/epoxy resin. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

value is the same as that obtained by single fiber fragmentation test.

CONCLUSIONS

In this paper, single fiber fragmentation test specimens and multiple fibers fragmentation test specimens with two dimensions were fabricated. The effect of interfiber distance on the interfacial properties in two-dimensionlly arranged multi-E-glass fiber/epoxy resin composites has been investigated. In addition, the effect of sizing on the interfacial properties has been studied and the findings made from this study can be summarized as follows.

1. Interfacial shear strength decreased with the decreasing of interfiber distance below 50 μm and the extent of the decreasing was more serious as the increasing of the number of adjacent fiber. This is probably due to the fact that the interface between the fiber and the resin was damaged by the breaking of adjacent fibers and the damage increased with decreasing the interfiber distance and the number of adjacent fibers
2. We can guess that the interfacial shear strength in real composites is much smaller than that of multifiber fragmentation sample with touched fibers.
3. In the two-dimensional fibers array fragmentation tests, when interfiber distance is above 50 μm, interfacial shear strength is the same as that obtained by single fiber fragmentation test.
4. When interfiber distance was small, the stress distribution pattern was shown like one fiber and when interfiber distance was large, over 50 μm, the stress distribution pattern was independent on between fibers.
5. The values of interfacial shear strength evaluated using two-dimensional fragmentation test are shown as real values regardless of fiber surface treatment, interfiber distance, and the presence of matrix cracks.

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