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

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ABSTRACT: The effect of interfiber distance on the interfacial properties in three-dimensional multi-E-glass fiber/epoxy resin composites has been investigated using fragmentation test. In additions, the effect of the fiber surface treatment on the interfacial properties has been studied. The interfacial shear strength decreased with the decreasing the interfiber distance at the range of under 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 adjacent fiber breaks and the damage increased with closing the interfiber spacing and the number of adjacent fiber. It was found that the interfacial shear strengths saturated when the interfiber distance was over 50 μm, the ones were saturated regardless of fiber surface treatment and the ones were in close agreement with those of the single fiber fragmentation test. Finally, the interfacial shear strength evaluated using three-dimensional fragmentation tests are shown as real values in-site regardless of fiber surface treatment, interfiber distance and existing of matrix cracks. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 116: 1483–1490, 2010

Key words: interfacial shear strength; interfiber distance; three-dimensional fragmentation test; damage; matrix crack

INTRODUCTION

It is well known, because of the large differences in mechanical properties between the reinforcing fibers and polymer resin, external stresses were transferred from polymer resin to fiber through the interface in fibrous reinforced composites. Most of the external load is carried by the fibers. So the interface between the fiber and the polymer resin is very important part in the fibrous composites and the exhibition of original fiber performance depends mainly on the interfacial properties in fibrous reinforced composites.

The interface plays a very important role in determining the final performance of the fibrous composites. Especially, the interfacial shear strength is one of the most fundamental factors in evaluating the mechanical properties and durability on the specific environment of the fiber reinforced composites.[1] In principle, the interfacial shear strength will be able to be controlled by the suitable combination of fiber, matrix resin and fiber surface modification, etc. Then, it is also very important to evaluate exactly the interfacial shear strength thus controlled.

For the last a few decades, many techniques such as the pull-out,[2–5] microbond,[6–10] fragmentation,[11–18] and indentation[19] test methods have been developed to try to measure the interfacial shear strength correctly.

Among the number of techniques mentioned previous, 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 easy to prepare sample comparing the other techniques. But currently, it takes $\sim$ 4 h to test a single sample. Because of this problem, new approaches have been developed to overcome, many works[11,20–23] have been done exploring the effects of testing multiple fibers in a fragmentation test.

Most of the current multifiber researches have focused on using laser Raman spectroscopy (LRS) as a detection tool for directly measuring the strain in broken and unbroken fibers.[24–36] 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 restricted to fibers with aromatic character that are Raman active. Hence, this detection technique has limited application to glass fibers.

The effect of interfiber distance on the interfacial properties in E-glass fiber/epoxy resin composites

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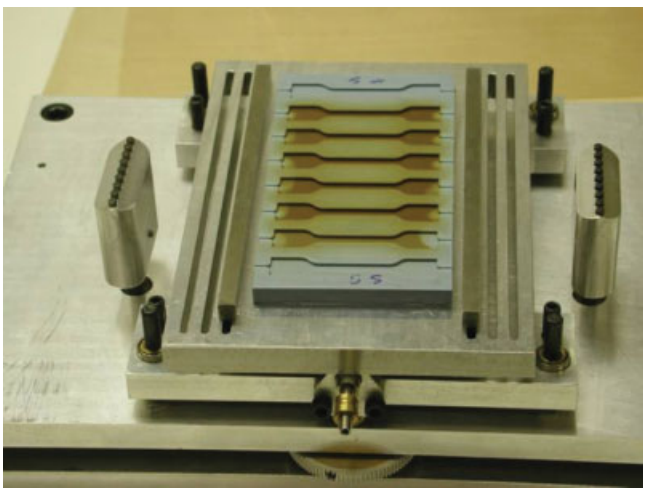

Figure 1 Apparatus of fiber arranging for multifiber fragmentation test specimen. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

using single, two fiber and two dimension fragmentation test specimen with various interfiber distance were investigated by authors.[37–39] From the previous works, it were found that when the interfiber distance was over 50 μm, interfacial shear strength was the same as the single fiber fragmentation test and when the one was under 50 μm, the one decreased with decreasing the interfiber distance.

The objective of this research is to study whether the results gained at previous work might be applied in three dimension fragmentation test.

In this article, multiple fibers fragmentation test specimens with three dimension were fabricated.The effect of interfiber distance on the interfacial properties in three dimension arranged multi-E-glass fiber/epoxy resin composites was investigated. In additions, the effect of sized and desized fiber in model composites on the interfacial properties has been studied.

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 of 50°C for 24 h. The matrix resin and hardener used were epoxy [diglycidl ether of bisphenol A (DGEBA), Epon 828, shell co.] and meta-phenylene diamine (m-PDA, Fluka chemical co.).

Preparation of fragmentation test sample

The sample preparation for single and multifiber testing was similar to that described by Drzal at al.,[14] and further details on specimen preparation are as follows.

The silicone (GE silicone RTV-664) mold with eight dog bone-shaped cavities (see Fig. 1) was used for the preparation of fragmentation test sample. Each cavity in the mold has sprue slots with width of 400 μm in the center of each end to aid in aligning the fiber in the center of the cavity. Single fiber was placed through the sprue slots of a silicone mold by hand, and multifibers with two and three dimension were placed using specific designed device as can be seen in Figure 1.

The method of preparation of fragmentation test sample with two and three-dimensional multifibers are as follows; first of all, two dimensional fragmentation test sample was fabricated as follows. First, double stick tape was placed on both side of long stick and outer of rotational rode with combs shape in Figure 1. Each fiber was placed on rotational rode with combs by hand alternatively. When six fibers were placed on it, double stick tape was placed on it to fix temporarily and then, by turning the screw under the device the fibers became closer each other [Fig. 2(a,b)]. It was turned the screw about 10° per each step and then waited for 10 min. to prevent fiber break. Like this, it was repeated until when the distance of each fiber was about 50 μm. And then, it was put down the arranged fibers into the slots of silicone mold with care [Fig. 2(c,d)]. The equally

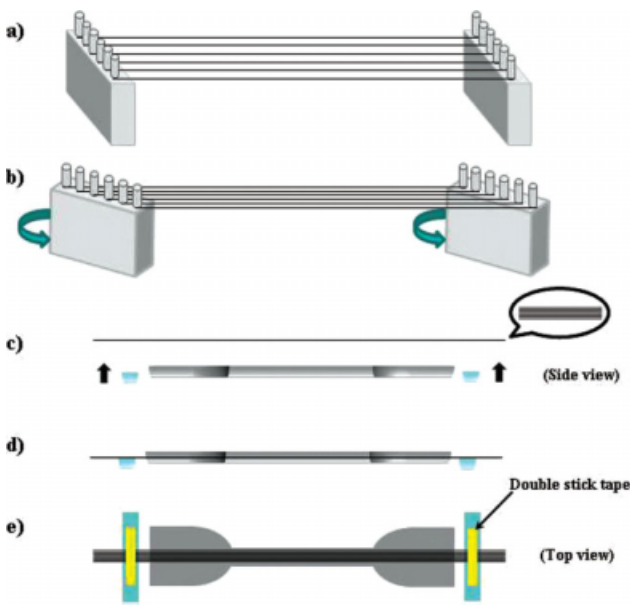

Figure 2 Schematic of multifiber arranging for fragmentation test specimen. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

arranged fibers were fixed temporarily by the double stick tape on the two long rectangular sticks in Figure 2(e).

Three-dimensional fragmentation test sample was fabricated by twice repeating of previous mentioned course at the same cavity of silicone mold. The thickness of the double stick tape was about 100 μm. In the three dimensions fragmentation test specimen, the height between two layers was designed to have about 100 μm. And then, arranged fibers were fixed by putting a small drop of five minutes epoxy resin (Hardman Adhesives) at the far end of each sprue slot. When fiber density was high like three-dimensional fragmentation test sample, the epoxy glue was hard to be infiltrated owing to high viscosity. So, it was used methylene chloride to lower the viscosity of epoxy resin as a diluent.

The multifiber specimens were prepared with an epoxy resin cured using meta-phenylene diamine. One hundred grams of DGEBA and 14.5 g of m-PDA were weighed out in separate beakers. To lower the viscosity of the resin and melt the m-PDA crystals, both beakers were placed in a vacuum oven (Fisher Scientific Isotemp Vacuum Oven, model 281 A) set at 65°C. After the m-PDA crystals were completely melted, the silicone rubber mold containing the fibers was placed into another vacuum oven (Fisher Scientific Isotemp Vacuum Oven, model 281 B) that was preheated to 75°C at –20 kPa, for 20 min. This last procedure dries the mold and minimizes the formation of air bubbles during the curing process. At ~ 9 min before the preheated molds were removed from the oven, the m-PDA is poured into the DGEBA and mixed thoroughly. The mixture was placed into the vacuum oven and degassed for ~ 7 min. After 20 min, the preheated molds were removed from the oven and filled with the DGEBA/m-PDA resin mixture using 10 mL disposable syringes. The filled molds were then placed into a programmable oven (Blue M, General Signal, model MP-256-1, GOP). A cure cycle of 2 h at 75°C followed by 2 h of post curing at 125°C was used. And then, it was allowed the sample to cool to room temperature in the oven before removal. After sample was removed from the silicone mold, and those samples where the fiber was not aligned and broken during processing were discarded. In fact, unlike single fiber fragmentation test specimen, it was hard to gain good sample having equalized interfiber spacing in two and three-dimensional fragmentation test specimen. In the three-dimensional samples, the heights between two layers were about 50 to 80 μm.

Fragmentation test

Fragmentation test was carried out by using fragmentation test machine as shown in Figure 3. Before each testing, samples were polished to avoid stress concentration at the edge part of sample by emery paper of nos. 800 and 2400. And then samples were marked the standard gauge length of about 10mm to measure real strain by permanent pen with blue color. When sample was installed, the grip was adjusted not much tight. The sample was loaded in tension by the sequential application of step strain. The number of total strain step was 28 and total strain was 2.4 mm. The strain rate was 85 μm/sec 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 applications of successive step strain was 10 min. After 28 steps, sample was unloaded and measured every fiber fragment length within gauge length.

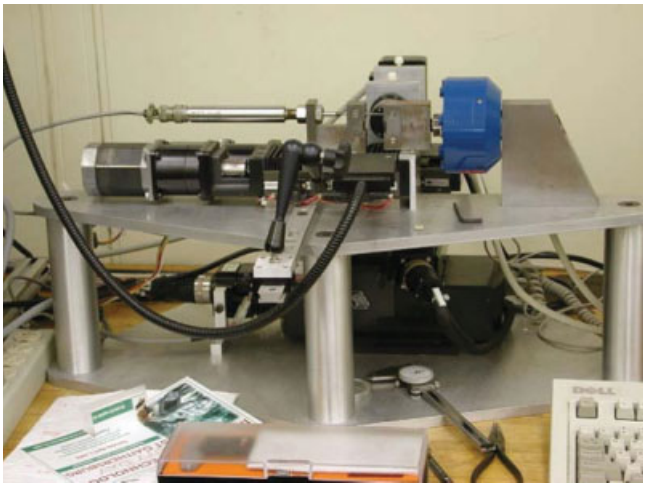

Figure 3 Automated fiber fragmentation testing machine. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

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 ~ 1.6 was used to improve image clarity.[38] Refraction index of the oil was almost same one 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 cal

The interfacial shear strength was calculated using following equation (1).[12]

The distribution of fragment lengths have been determined to be satisfactorily described by a two-

TABLE I
Tensile Strength of Single E-Glass Fiber

Fiber	Sized fiber	Desized fiber
Tensile strength (GPa)	2.10 ± 0.57	1.63 ± 0.44

parameter Weibull analysis, causing the expression for the interfacial shear strength τ, to be come

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

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

Γ is 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, it was used to the single fiber tensile strength at 20 mm of gauge length. One goal of this study is to discuss about the effect of interfiber distance on the interfacial properties in fragmentation test.

RESULTS AND DISCUSSION

Single fiber fragmentation test

The Table I shows the tensile strengths of sized and desized E-glass single fiber. The tensile strength was measured on single fiber with gage length of 20 mm at a crosshead speed of 2.0 mm/min. using a tensile tester equipped with a load cell of 100 g.

Before tensile testing, the diameter of every sample was measured with an optical micrometer (VIA-100, Boeckeler).

The tensile strength of sized fiber is shown stronger than one of desized fiber. This is probably due to the fact that desized fiber is easier to get damage when they rub against each other during processing operations. In many cases, it is known that the most important factor determining the tensile strength of the glass fiber is the damage of fiber surface.

Table II shows the interfacial shear strengths of single fiber fragmentation test and the values were average and standard deviations.

Multifibers fragmentation test

Figure 4 represents the plot of interfacial shear strength according to interfiber distance in the several E-glass fiber/epoxy resin fragmentation test. It

TABLE II
Interfacial Shear Strength of E-Glass Fiber/Epoxy Resin

Fiber	Sized fiber	Desized fiber
Interfacial shear strength (MPa)	46.70 ± 2.59	40.69 ± 2.60

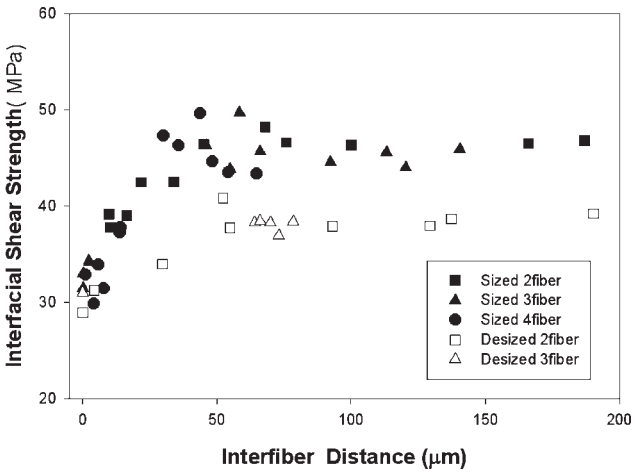

Figure 4 Interfacial shear strength vs. interfiber distance in the fragmentation test of the different fiber number.

was shown that the interfacial shear strength increased until interfiber distance became about 50 μm, and then the ones were saturated regardless of sized and desized fiber.

That is, in case of the interfiber distance of under 50 μm, the interfacial shear strength decreased with decreasing the interfiber distance and the extent of the decreasing was more serious as the increasing of the number of adjacent fiber.[37–39] This is considered that the interface was damaged and become weak by the adjacent fiber breaks and the extent of the damage increased with closing the interfiber spacing and the number of adjacent fiber. Thus, it is reasonable to assume that interfacial shear strength in real composites is much smaller than that of multifiber fragmentation sample with touched fiber.

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

The damaging factors considered owing to fibers break are strain energy release, stress transfer and stress concentration, etc.[37–39] In case of sized fiber sample, the extent of stress concentration depends on mainly exiting matrix crack [see Fig. 9(a)]. 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 the one in the desized fiber fragmentation test.

Figure 5 shows the result of multifiber fragmentation test to investigate the interfacial shear strength of real composites. It was shown that the interfacial shear strength decreased with the number of touched fiber. This is another reasonable data to assume that interfacial shear strength in real composites is smaller than that of multifiber fragmentation sample with touched fiber.

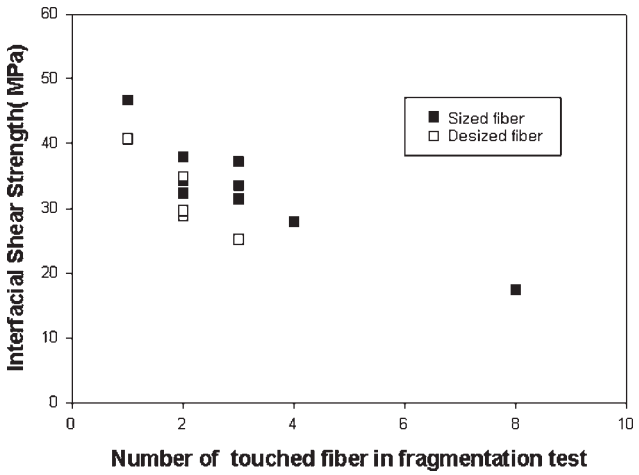

Figure 5 Interfacial shear strength vs. number of touched fiber in fragmentation test.

Figure 6 presents the micrograph of fiber fragment in the eight fiber fragmentation test. From this figure, it was shown that all fiber was arranged very close each other and three dimensionally and the locations of fiber break were similar. The latter is probably that fiber break occurs within damaged interface induced by the adjacent fiber break.[39]

Figure 7 reveals the polarized transmitted light micrographs of desized multi-E-glass fiber/epoxy resin fragmentation test at saturation. It was found that interfiber spacing was not equally arrays. In fact, the preparation of good sample with equal interfiber distance is extremely difficult. It could be arranged the fiber with equal interfiber spacing using special tool as shown in Figure 1. It was shown that when the interfiber distance is small, the stress distribution pattern is shown like one fiber

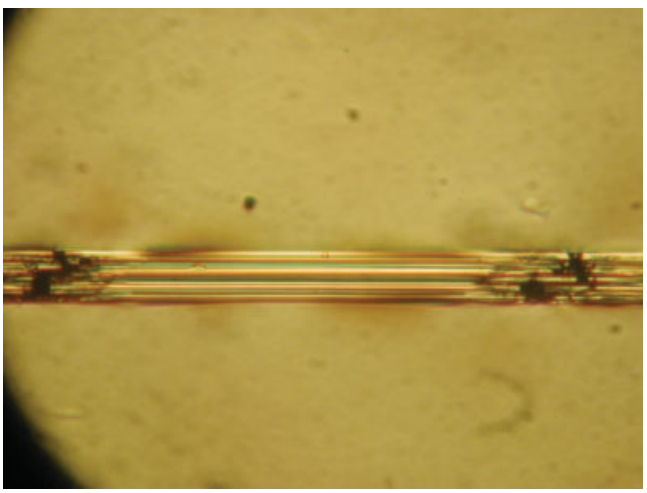

Figure 6 Micrograph of the sized eight glass fiber/ epoxy resin fragmentation test at saturated. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

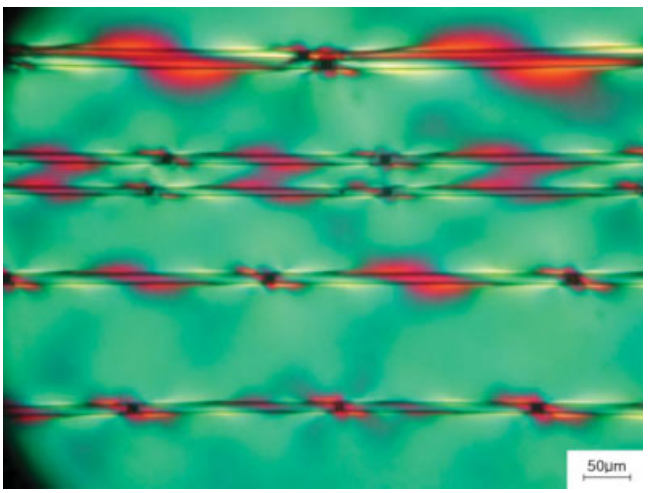

Figure 7 Polarized transmitted light micrograph of the desized E-glass fiber/epoxy resin fragmentation test at saturated. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

with big fiber diameter and when the interfiber distance is large, the stress distribution pattern is independent of between fiber breaks.

From the Figure 4, it can be easily seen that when the interfiber distance was over 50 μm, interfacial shear strength became independent values. Therefore, in two and three-dimensional fragmentation test, if the interfiber distance was at least over 50 μm, it was considered nothing the effect of interfiber distance on the interfacial shear strength.

For example, in Figure 7, it was used the interfiber distances were 0, 17, and 154.9 μm from the upper of the figure respectively. Unless otherwise noted, interfiber distance means as the same mentioned previously.

Figure 8 represents the plot of interfacial shear strength according to interfiber distance in the two dimensions multi-E-glass fiber/epoxy resin fragmentation test. The interfacial shear strength increased until when the interfiber distance became about 50 μm, and then the ones were saturated regardless of sized and desized fiber. In the whole region, the interfacial shear strength of sized was bigger than the one of desized fiber. However, in case of the interfiber distance of under 50 μm, some of the interfacial shear strengths of sized fiber were seen smaller than the ones of desized fiber. This is probably the effect of matrix crack with being long as previously mentioned damaged mechanism. Figure 8 also shows that if interfiber distance becomes over 50 μm in the two dimensions fragmentation test regardless of fiber surface treatment the result is the same as the one of the single fiber fragmentation test.

Figure 9 represents the micrographs of the fiber fragment in the three-dimensional fragmentation test

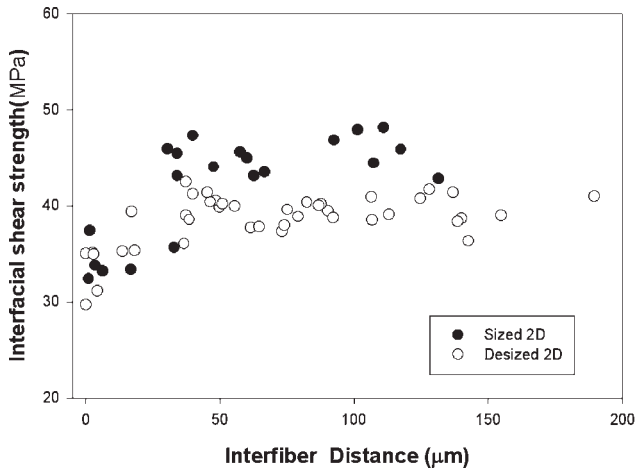

Figure 8 Interfacial Shear Strength vs. Interfiber Distance in the fragmentation test.

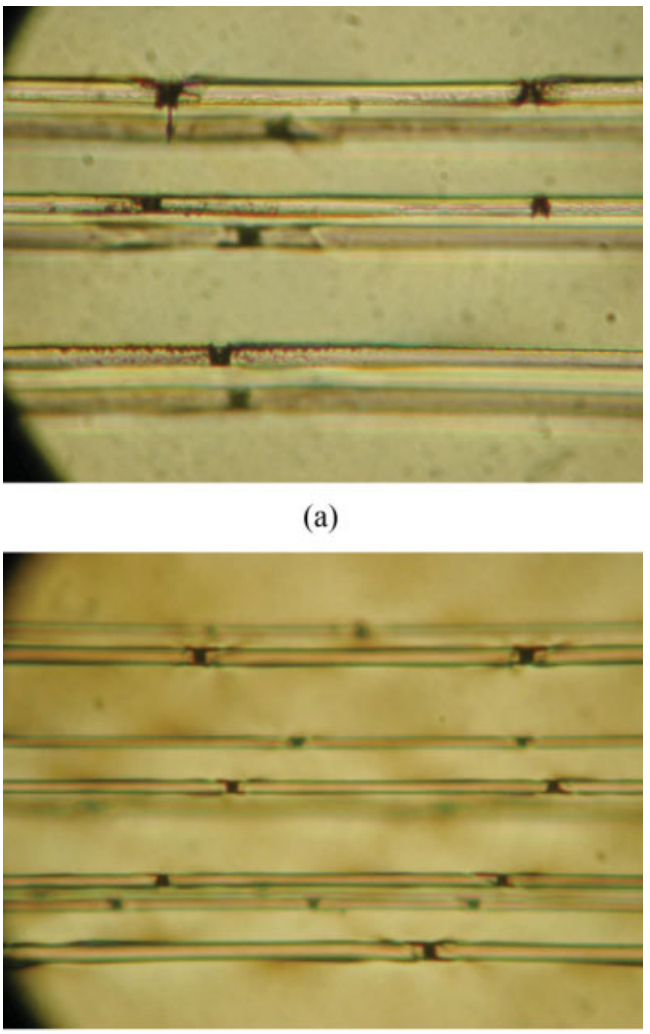

Figure 9 Microphotographs of the three dimension fragmentation test specimen at saturation (a) Sized fiber (b) Desized fiber. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

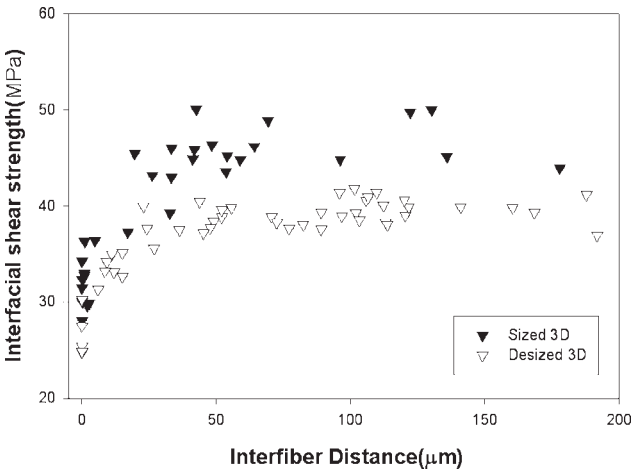

Figure 10 Interfacial shear strength vs. interfiber distance in the three dimension fragmentation test.

sample. At this figure, (a) is sized and (b) is desized fiber of fragmentation test. It can be seen the samples of sized and desized fibers were made to three dimensions and there were matrix cracks in sized sample, but there was no matrix crack in desized sample.

Figure 10 shows the plot of interfacial shear strength according to interfiber distance in the three dimensions multi-E-glass fiber/epoxy resin fragmentation test. It was also shown that the interfacial shear strength increased until when the interfiber distance became about 50 μm, and then the ones were saturated regardless of sized and desized fiber. In the whole region, the interfacial shear strength of sized was shown bigger than the one of desized fiber. But in case of the interfiber distance of under 50 μm, some of the interfacial shear strengths of sized fibers were found smaller than the ones of desized fiber like in Figure 8. This is also considered that the effect of matrix crack with being long as previously mentioned in the damaging factors. There were matrix cracks in the sized fiber fragmentation test specimen.

Figure 11 reveals the plot of interfacial shear strength according to apparent interfiber distance in the three dimensions multi-E-glass fiber/epoxy resin fragmentation test. In this figure, an apparent interfiber distance means apparent interfiber distance between the different two layers, whereas interfiber distance described previous meant the distance of fibers between the same layers. From this figure, it can be seen that interfacial shear strengths are independent of apparent interfiber distance regardless of sized and desized fiber in the whole region without some values at folded fibers. The distance of most samples between the upper and lower layers was measured from 50 to 80 μm before testing. The results of those samples were good accordance with the ones of single and two dimensions multifiber

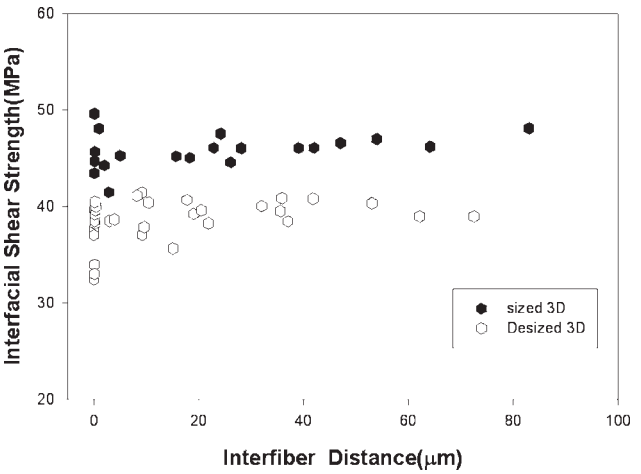

Figure 11 Plot of Interfacial shear strength vs. apparent interfiber distance of different layer in three dimension fragmentation test.

fragmentation test. However, in desized 3D fragmentation sample, some of folded fibers between two layers were lower values than the average values. The actual interfiber distances of samples with lower values were found under 50 μm, it were 34.8, 38, 4, and 30.0 μm.

Eventually, in the three dimensions fragmentation test, when interfiber distance is over 50 μm, interfacial shear strength is the same values as single fiber fragmentation test.

The values are true regardless of interfiber distance. But it was hard to measure the fiber fragment when fibers were folded between the upper and the lower layers. If the fibers of two layers are arrayed well alternatively, the interfacial shear strength similar with one of the single fiber fragmentation test will be gain.

From above mentioned, it was found that the interfacial shear strength in the several fiber fragmentation test, two and three-dimensional fragmentation test show the real values despite any case. When interfiber distance is over 50 μm, the value is the same as one of single fiber fragmentation test.

CONCLUSION

In this article, multiple fibers fragmentation test specimens with three dimensions were fabricated. The effect of interfiber distance on the interfacial properties in three dimension arranged multi-E-glass fiber/epoxy resin composites has been investigated. In additions, the effect of sized and desized fiber in model composites 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 the interfiber distance at the range of under 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 adjacent fiber breaks and the damage increased with closing the interfiber spacing and the number of adjacent fiber.
2. Interfacial shear strength in real composites is probably much smaller than that of multifiber fragmentation sample with touched fibers.
3. In the several, two and three-dimensional fibers array fragmentation tests, when interfiber distance is over 50 μm, interfacial shear strength is the same values as single fiber fragmentation test.
4. The interfacial shear strength evaluated using three dimensions fragmentation test is shown as real values in-site regardless of fiber surface treatment, interfiber distance and existing of matrix cracks.

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