Experimental Verification of Interfacial Strength Effect on the Mechanical Properties of Carbon Fiber-Epoxy Composite

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Abstract— **The effects of carbon fiber–epoxy interfacial strength on the mechanical properties of the corresponding fiber-matrix composites are experimentally demonstrated in this work. Two composites containing different carbon fibers were tested: as-received fibers and fibers soaked in acetone to remove adhesive on their surfaces. The fiber surfaces were first characterized by scanning electron microscopy and time-of-flight secondary-ion mass spectrometry to verify removal of the adhesive. Further, single-fiber fragmentation tests were conducted to evaluate the fiber strength and the interfacial strength. The mechanical properties of the composites were evaluated via tensile testing under longitudinal and transverse loadings. The results show that interfacial strength does not decrease the mechanical properties of the composites under longitudinal loading. In contrast, under transverse loading, the interfacial strength significantly decreases the mechanical properties, specifically the ultimate tensile strength and toughness of the composites.**

Keywords— **interfacial strength; mechanical properties; fiber-matrix composite; tensile test**

I. INTRODUCTION

The mechanical properties of fiber-matrix composites have been predicted to be affected by not only the mechanical properties of their constituents but also the constituents' interfacial properties [1]. Interfacial properties, particularly interfacial strength (t_o), determine the effectiveness of stress transfer from the fiber to the matrix and vice versa, which is directly related to the mechanical properties of such composites [2], [3]. Numerous studies have predicted that a low t_o can result in fiber-matrix composites with poor mechanical properties [4], [5]. However, the literature contains few studies that have directly verified the effects of t_o on the mechanical properties of fiber-matrix composites.

Fig. 1a shows the components of fiber–matrix composite at the micro-level. Here, the interface is considered an important component that should be carefully characterized [6]-[8]. The t_o is usually evaluated by a micro-scale testing method such as the single-fiber fragmentation test (SFFT) [9], [10], the push-out test [11], the pull-out test [12], or the micro-bond test [13]. The primary purpose of such evaluations is to elucidate the effectiveness of fiber surface treatments with respect to increasing t_o [14]. Researchers have reported substantial improvements of t_o as a result of various surface treatment techniques, indicating that such improvements might improve the mechanical properties of fiber-matrix composites.

The mechanical properties of composites have been directly evaluated using tensile tests for ply composite specimens under both longitudinal and transverse loadings as shown in Fig. 1b and 1c. Depending on loading condition,

the failure mechanisms of the composites might be different. Under longitudinal loading, fiber is firstly broken followed by interfacial cracking or matrix cracking depending on the strength and fracture toughness of them [19]. In contrast, those cracks might firstly appear under transverse loading, which remains the fibers that do not bear the loading. In both longitudinal and transverse loading conditions, t_o surely determines the failure mechanisms, which directly affects the mechanical properties of the composites.

Substantial analytical and numerical studies have been conducted to elucidate the micro-level effects of t_o on the ply-level mechanical properties of fiber-matrix composites; these studies have focused on modeling the relationship between micro-mechanics and macro-mechanics [15]. Such studies can be conducted by analyses involving homogenization techniques [16]-[18]. By using these techniques, researchers can predict the effects of local micro-scale cracks at the interface on the mechanical properties of composites. Although these techniques appear to be promising for explaining the effect of t_o on the mechanical properties of fiber-matrix composites, the experimental validation of these techniques is still a challenging problem due to the scaling difference of evaluations between t_o and mechanical properties of the composites. Limited experimental studies on the investigation of the relationship between t_o and mechanical properties of the composite were previously conducted in which micro and ply levels are simultaneously analyzed. As a consequence, the high t_o values achieved using the previously reported fiber surface treatments are questionable because such treatments have not been demonstrated to positively affect the mechanical properties of fiber-matrix composites. Indeed, direct verification must be obtained via experiments in which the t_o and the mechanical properties of fiber-matrix composites are simultaneously evaluated.
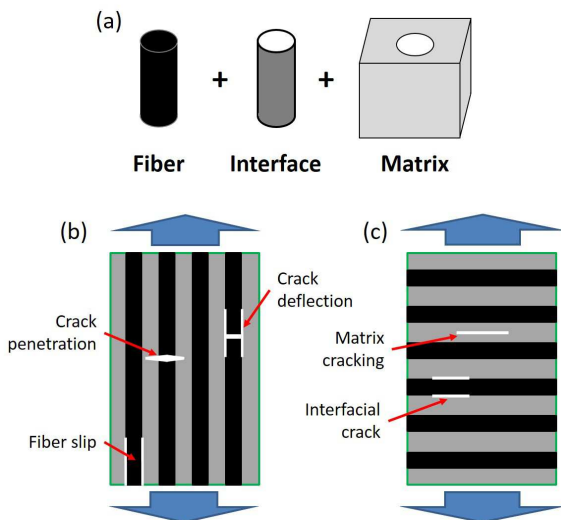

Fig. 1 Components of composite structures at the micro level (a) and illustrations of longitudinal loading (b) and transverse loading (c) applied to ply composites

In the present work, the effect of t_o was investigated experimentally. Two composites containing carbon fibers with different surface treatments were tested. The fiber surfaces were first characterized by scanning electron microscopy (SEM) and time-of-flight secondary-ion mass spectrometry (TOF-SIMS) [20]. The t_o and fiber strength (σ_f) were evaluated using SFFT. The mechanical properties of the ply fiber-matrix composites were then evaluated using tensile tests. The specimens were subjected to longitudinal and transverse loadings. In addition, the effects of t_o on the mechanical properties are comprehensively discussed.

II. MATERIAL AND METHOD

A. Characterization of the Fiber Surface

Two types of carbon fibers HTA 40 (TOHO Tenax Co. Ltd.) with different surface treatments were prepared. The first type was as-received carbon fibers obtained from the manufacturer which contain adhesive material on their surface. The second type was carbon fibers conditioned by being soaked in acetone for more than 5 hours and then rinsed with water. This treatment was conducted to remove any adhesive material on the carbon fiber surface.

The surface roughness of both carbon fibers was observed by using SEM. The purpose is to clarify alterations of the roughness due to the surface treatment. Further, TOF-SIMS analysis to verify that the adhesive material had been removed after the surface treatment was also conducted. The positive secondary ion mode was selected to investigate the material compound in the as-received and conditioned fiber surfaces. The positive ion has high sensitivity to hydrocarbon molecules that might be attached to the fiber surfaces during sizing process. These molecules aim to form strong bonding between carbon fiber and polymer matrix such as epoxy.

The σ_f and the t_o were subsequently evaluated by SFFT. The SFFT specimens contained single carbon fiber surrounded by epoxy (Konishi Chemical Co. Ltd). A shear lag model proposed by Kelly-Tyson was used to evaluate σ_f and t_o according to the equations shown in Equations 1 and 2, respectively [21]:

$$\sigma_f = \sigma_o \left(\frac{L_o}{L_c} \right)^{1/m} \tag{1}$$

$$t_o = \frac{\sigma_f d}{2L_c} \tag{2}$$

where σ_o is the characteristic stress at which fiber cracks appear, L_o is the measured area, L_c is the critical length, m is the Weibull modulus, and d is the diameter of the fibers. Details of the SFFT procedure are described elsewhere [10].

B. Tensile Testing

The tensile testing specimens used in this study were prepared using the carbon fibers previously characterized and the epoxy produced by Konishi chemical Co. Ltd. Ply composite specimens for tensile testing were prepared using the hand lay-up technique [22]. The specimens were designed to have low fiber volume fractions of 20% to ensure that the fiber surface appropriately contacted the matrix. Thus, the experimental results reflect the effects of t_o. Fig. 2 shows the dimensions of the specimens for tensile testing. The specimens exhibit a uniform fiber direction; they were cut for longitudinal and transverse loading tests.
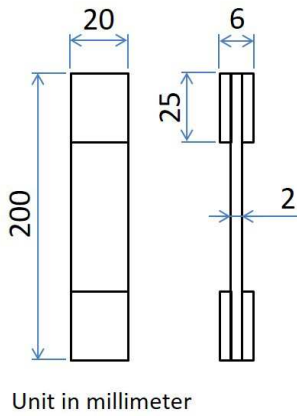
Fig. 2 Schematic showing the specimen dimensions

The detailed scenarios for tensile testing are reported in Table 1. An Instron tensile testing machine was used with a maximum load of 10 kN. A tensile speed of 2 mm/min at room temperature was applied to all specimens. The load and the displacement were recorded simultaneously until the specimen was broken. The stress-strain curve for each specimen was then obtained from the load-displacement results. From the curve, mechanical properties of the composite were analyzed.

TABLE I
TENSILE TESTING SCENARIOS

Loading type	As-received fiber–epoxy composite	Conditioned fiber–epoxy composite
Transverse loading	3 specimens (A1, A2, A3)	3 specimens (B1, B2, B3)
Longitudinal loading	3 specimens (A4, A5, A6)	3 specimens (B4, B5, B6)

III. RESULTS AND DISCUSSION

A. SEM Observation

The SEM micrographs are shown in Fig. 3. Figs. 3a and 3b clearly show that the surfaces of both the as-received fibers and the conditioned fibers have uniform porosities parallel to the fiber direction. These uniform porosities are possibly formed during the manufacturing process of the fibers. Later on, manufacturing the composite, the epoxy can fulfill these porosities that can create mechanical locking between the fibers and the matrix and increase t_o. Figs. 3c and 3d show SEM micrographs of the cross-sectional area of both the as-received fibers and the conditioned fibers. The surface roughness of both fibers was mostly unchanged, which means that the t_o due to mechanical locking might be retained even after the fibers' surface was soaked in acetone.

B. TOF-SIMS Analysis

The TOF-SIMS spectra are shown in Fig. 4. The intensity of the hydrocarbon peaks in the 20–60 Da range was substantially lower in the spectrum of the conditioned fiber surface compared with that in the spectrum of the as-received fiber surface. In particular, the peak intensities associated with $C_2H_n^+$ in the range from 27 to 30 Da, $C_3H_n^+$ in the range from 39 to 44 Da, and $C_4H_n^+$ in the range from 52 to 57 Da drastically decreased [23].
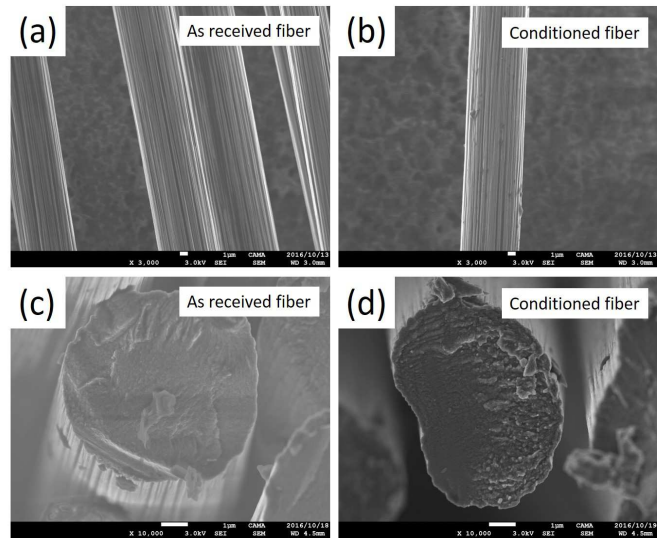

Fig. 3 SEM micrographs of the fiber surface of as-received fibers (a), the conditioned fibers (b), and the cross-sections of the as-received fibers (c) and the conditioned fibers (d)
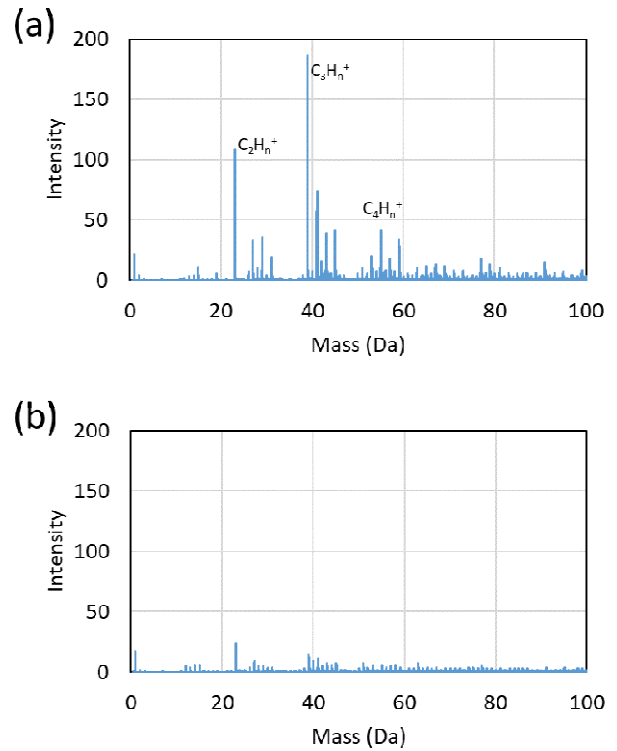

Fig. 4 TOF-SIMS spectra for the as-received fibers (a) and conditioned fibers (b). The decreased intensity of peaks in the range from 20 to 60 Da indicates that the hydrocarbon at the interface was eliminated

The presence of these peaks in the mass spectrum of the as-received fiber surface indicates that an adhesive material containing hydrocarbon molecules was added during the sizing process. It is noted that the exact adhesive compounds are secretly kept by the fiber manufacturer. However, after the fiber was soaked in acetone, no such hydrocarbon content was detected by TOF-SIMS analysis, which indicates that the acetone treatment successfully removed the adhesive. This observation also confirms that the effects of chemical bonding and mechanical locking on t_o can be practically separated by soaking the fiber in acetone to remove the adhesive material.

C. SFFT Results

Fig. 5 shows the σ_f and t_o values determined from the SFFT results. After the fibers were soaked in acetone, the values of the σ_f and the t_o decreased approximately 20% and 50%, respectively. Although the adhesive material was removed from the specimen with conditioned fibers, the low t_o still remains. Considering the fibers mainly contain stable carbon compound which is difficult to be reacted with another compound, a plausible explanation of the remaining t_o comes from a mechanical locking that prevents shearing-mode crack formation at the interface. The mechanical locking is most likely induced by surface roughness, which remains after the surface treatment, as observed in the SEM images in Fig. 3. Furthermore, by assuming that the stiffness of the fibers (E_f) did not change in response to the surface treatment and assuming that the fiber behavior was linear elastic, we calculated the fiber toughness (U_f) using Equation 3:

$$U_f = \frac{\sigma_f^2}{2E_f} \tag{3}$$

where the value of E_f is 240 GPa, as obtained from the TOHO Tenax Co. Ltd. data sheet. Thus, U_f values of 48 kJ/mm^3 for the as-received fibers and 30 kJ/mm^3 for the conditioned fibers were obtained using Equation 3. The U_f decreased by approximately 37.5% after the acetone treatment, which indicates that the adhesive material also substantially contributed to the value of U_f.
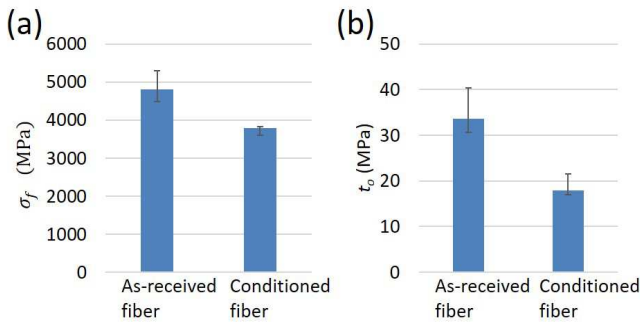

Fig. 5 Fiber strength (a) and interfacial strength (b) for the as-received and conditioned fibers [10]

D. Stress-Strain Curves

Stress-strain curves for tensile testing under longitudinal and transverse loadings are shown in Figs. 6 and 7, respectively. The stress-strain curves for three specimens of both the as-received and conditioned fibers show relatively consistent results. In the case of transverse loading testing, nonlinear stress-strain curves that might be caused by the epoxy properties were obtained for both the as-received fibers and the conditioned fibers. These results confirm that nonlinear behavior of the epoxy plays a dominant role in determining the mechanical properties of the composite in the transverse loading direction. Furthermore, the stress-strain curves of the specimens with conditioned fibers decreased to 50%. This result indicates that t_o strongly affects the mechanical properties of the composite under transverse loading. Considering the composite subjected to transverse loading is always to be the weakest condition, the

t_o unavoidably determines the overall performance of the composite as load-bearing structures.
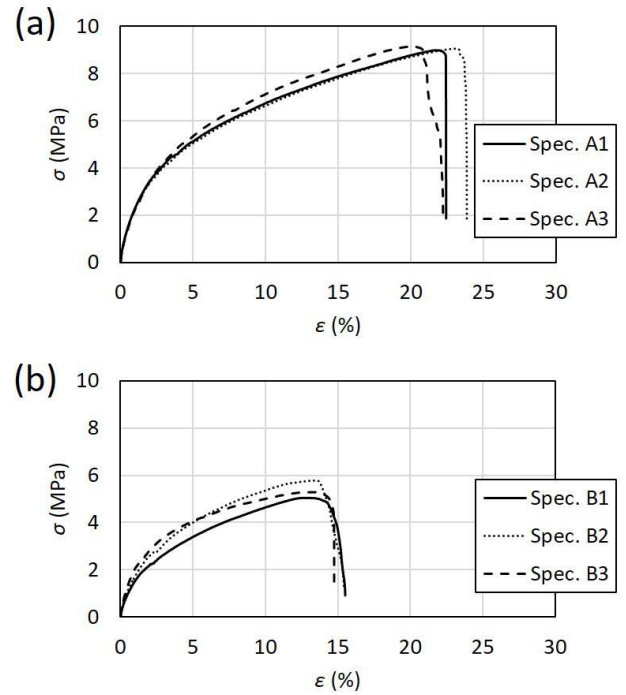

Fig. 6 Stress-strain curves of the as-received fiber–epoxy composite (a) and the conditioned fiber–epoxy composite (b) tested under transverse loading
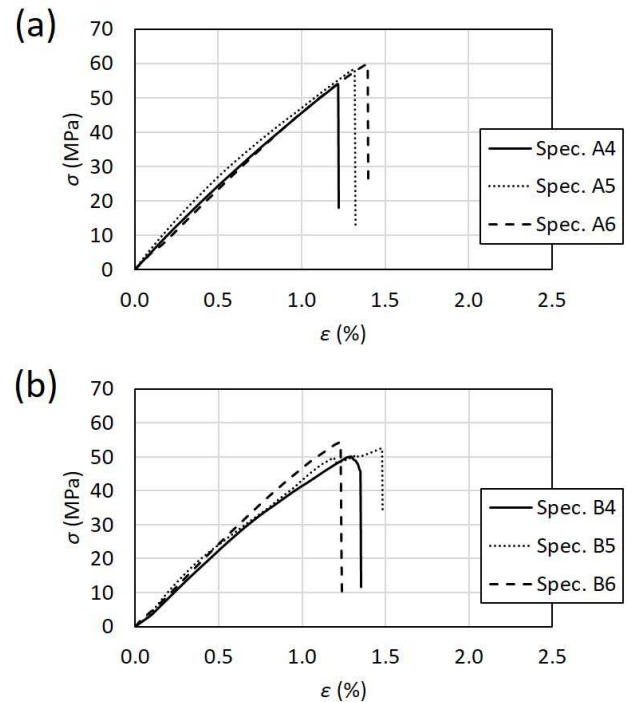

Fig. 7 Stress-strain curves of the as-received fiber–epoxy composite (a) and the conditioned fiber–epoxy composite (b) tested under longitudinal loading

The tensile test results for the composite under longitudinal loading differed slightly from those for the composite under transverse loading. As evident in Fig. 7, linear stress-strain curves were obtained from tensile testing results of specimens with both as-received fibers and

conditioned fibers. These results are related to the linear behavior of the fibers, which dominantly affect the curves. Under longitudinal loading, the t_o did not substantially decrease the mechanical properties of the composite. Notably, however, the elastic modulus of the composite also slightly changed, which might be caused by ineffective stress transfer between the matrix and the fibers [3].

E. Alteration of Mechanical Properties

The mechanical properties, i.e., the ultimate tensile strength (σ_{uts}) and toughness (U), of the composites under longitudinal and transverse loadings were calculated on the basis of the stress-strain curves obtained via tensile tests. outs refers to the maximum stress that appears in the curves, whereas U is defined as a total area under the curves. The calculated values of σ_{uts} and U are then plotted in Figs. 8 and 9, respectively.

Fig. 8 clearly shows that the σ_{uts} under longitudinal loading was only slightly changed. This result is explained on the basis of the mechanism of composite failure under longitudinal loading, where fiber breakage might first occur. Before fiber breakage, the interface does not usually bear relatively high stress; thus, fractures at the interface are avoided. In fact, whether a decrease in σ_{uts} is caused by a low t_o or by a low σ_f is uncertain because, under the longitudinal loading condition, fracture occurs via the shearing mode, which can cause mechanical locking that generates additional t_o. The effect of mechanical locking becomes more dominant for rough fiber surfaces. By contrast, σ_{uts} under transverse loading drastically decreases as a consequence of a low t_o because the interface bears high stress to transfer the load from the matrix t_o the fiber. At this stage, t_o plays a very important role and can directly determine σ_{uts}. The opening mode of fracture at the interface might also contribute to a decrease in σ_{uts}, where mechanical locking does not effectively prevent fracture of the interface.

As shown in Fig. 9, U exhibits tendencies similar to those observed for σ_{uts}. The U drastically decreases under transverse loading because the loading cannot be transferred from the matrix t_o the fibers as a consequence of a weak interface. Thus, the fibers cannot be deformed to absorb the strain energy from external loading. However, U only slightly decreases under longitudinal loading because the fibers are deformed and broken, resulting in the strain energy from the external loading being absorbed. This result is interesting because low to might not substantially cause low U.

Under the scenario of composite failure, after cracks form in the fibers, a low t_o can cause the cracks to first deflect at the interface before penetrating into the matrix. However, a high to causes the cracks to directly penetrate into the matrix. Consequently, the surface area of the cracks generated as a result of a low t_o is much larger than that of the cracks generated as a result of a high t_o, which means the absorbed energy required t_o create the cracks is substantially greater in the case of a low t_o. The slight decrease of U might only be caused by the decrease of U_f in response to the surface treatment rather than by a low t_o.

We also note that according to Fig. 9, the values of U are much higher when the composite is subjected to transverse loading rather than longitudinal loading. This result arises from the ductile properties of the matrix, which enables the absorption of strain energy via matrix deformation. Under longitudinal loading, the fiber cracks cause stress concentration in each point of the composite, which in turn causes the composite to break under the low strain. Thus, the total strain energy absorbed by the composite is relatively low.
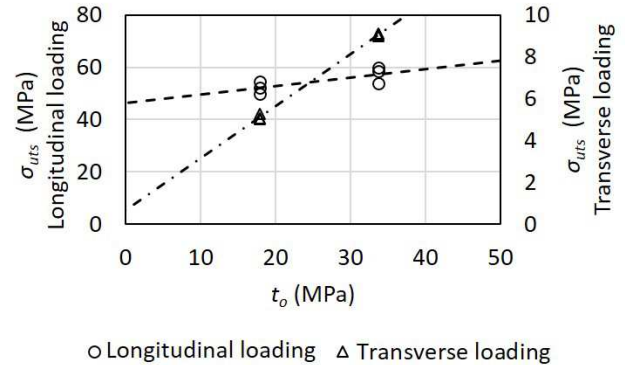

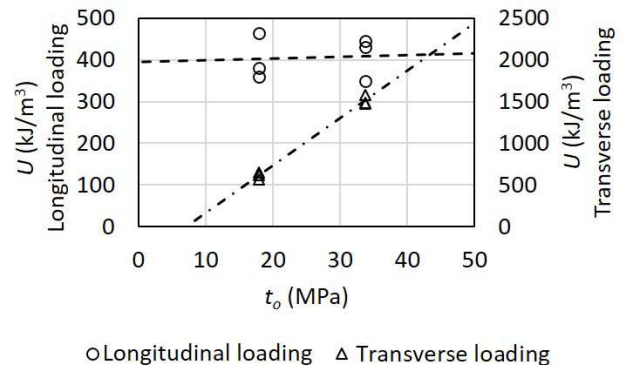Fig. 8 Tensile strength of the composite versus interfacial strength

Fig. 9 Toughness of the composite versus interfacial strength

On the basis of these experimental results, t_o is demonstrated to strongly affect the mechanical properties of carbon fiber–epoxy composite, i.e., its σ_{uts} and U, particularly under transverse loading. Low t_o can drastically decrease the σ_{uts} and U. In contrast, the role of t_o under longitudinal loading is still questionable. Depending on the failure mechanisms, low t_o might give benefit to the mechanical properties of the composite because the deflection of fiber crack to the interface prior matrix cracking can increase the strain energy absorption, which delays final failure of the composite. However, when the interfacial cracks appear prior fiber breaks, which is possible for the extremely small t_o, slip failure mechanism at the interface might occur that causes the mechanical properties of the composite drastically decrease.

In the engineering applications, the composite is also designed for absorbing the strain energy as much as possible such as for crashworthiness structures and for blast impact protectors. In these cases, the defects of the composite structures cannot be avoided since the load imposed to the structures is much larger than σ_{uts}. In the context of the design problem for this case, increasing U of the composite is more beneficial than the σ_{uts}. Thus, the t_o should not be too high for allowing the fiber crack deflected to the interface. In

general, controlling t_o values in the manufacturing process of the composite is important to meet requirements of the design structures.

IV. CONCLUSIONS

The effect of interfacial strength on the mechanical properties of a fiber–matrix composite was revealed. The interfacial strength strongly affected the composite under applied transverse loading: the ultimate tensile strength and toughness of the composite decreased by 50% when the interfacial strength was decreased 50%. This relationship might be a consequence of the failed transfer of the load from the matrix to the fibers, rendering the fibers unable to effectively participate in bearing the load and resulting in the appearance of cracks at the interface under low loading. By contrast, the mechanical properties of composites under longitudinal loading only slightly changed in response to changes in interfacial strength because the failure scenario of the composite under longitudinal loading allows fibers to break before cracks occur at the interface or in the matrix. The diminished mechanical properties of the composite are mostly caused by poor mechanical properties of fibers owing to the surface treatment. The role of interfacial strength in determining the mechanical properties of the composite was confirmed in this study.

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