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Interfacial adhesion improvement of plain woven carbon fiber reinforced epoxy filled with micro-fibrillated cellulose by addition liquid rubber

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Abstract In certain application of fiber reinforced polymer composites fracture resistance is required. The aim of this study was to improve the interfacial adhesion between plain woven carbon fiber (CF) and epoxy matrix filled with microfibrillated cellulose (MFC) modified with carboxyl-terminated butadiene acrylonitrile (CTBN) as liquid rubber. CF/Epoxy/MFC/CTBN composite was characterized by different techniques, namely, tensile, bending, fracture toughness (mode I) test, and scanning electron microscope (SEM). The results reveal that at a fiber content 1% of MFC and 10% CTBN, initiation and propagation interlaminar fracture toughness in mode I improved significantly by 96 and 127%, respectively, which could be attribute to strong adhesion between filled epoxy, CF, and rubber. This can be explained by SEM at given weight as well; SEM images showed that in front of the tip, fiber breakage during initiation delimination as well as the extensive matrix deformation between fibers accounting for increase fracture toughness.

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Introduction

Composite materials have been extensively applied to the areas of aerospace, aircraft, sports, and military industries. Composite materials formed by natural fibers and polymeric matrices constitute a current area of interest in composites research. A great development in this field has been noticed, mainly driven by the automotive industries. Cellulose, the most abundant natural homopolymer, is considered to be one of the most promising renewable resources and an environmentally friendly alternative to products derived from the petrochemical industry. Plant derived cellulose has been widely used as either reinforcement [1–3] or matrix [1] and even as the sole component to prepare all-cellulose composites [4].

Recently the identification of nano-sized cellulose microfibrils which called microfibrillated cellulose (MFC) increases the choices of fibers and expands their use because of their excellent mechanical properties in composites. Using MFC as reinforcement with polymer matrix gives significant effect on the mechanical properties for the composite [5–9]. Takagaki et al. [10] showed that addition of 0.1, 0.3 wt% MFC contribute slightly to improve the static properties of MFC–CFRP–epoxy composite. They also stated that addition of MFC contribute to improve fatigue life of the composite. Zimmerman et al. [3] reported that MFC-reinforced hydroxypropyl cellulose (HPC) prepared by film casting has three times higher tensile modulus and five times higher tensile strength compared with the matrix without reinforcement. Nakagaito and Yano [11] showed that nanocomposites produced by compression molding of MFC sheets impregnated with phenol formaldehyde (PF) resin have high bending strength and modulus comparable to magnesium alloy. Nanocomposite materials based on MFC and melamine formaldehyde (MF) resulted in high mechanical damping, showing their potential to be used as loudspeaker membranes [12]. Suryanegara et al. [13] showed that the addition of MFC increased the tensile modulus of crystallized neat polylactic acid (PLA) by 42% and the strength by 14% at an MFC content of 20 wt%.

However, the low interlaminar strength of composite laminates is one of the major disadvantages, which delayed the widespread use of composite laminates in primary aircraft structures. Interlaminar fracture is one of the major problems for fiber composites. Its occurrence greatly reduces the stiffness of a structure, often leading to catastrophic failure during service [14]. For this reason, many efforts have been made to improve the interlaminar strength. Okubo et al. [15] stated that if small amount of MFC added into the bamboo fiber composite, tangled MFC fibers prevented the growth of micro-crack along the interface between bamboo fiber and matrix. Okubo et al. [16] showed significant improvements in the strain energy until fatal failure when the PLA matrix enhanced with 1 wt% of MFC. Naoya et al. [17] showed that addition of small amount of MFC into PLA/bamboo short fiber (PLA/BF) composites contribute to improve the fracture toughness and impact strength.

In this study, polymer matrix composites (PMCs) are produced using an epoxy resin as matrix. Epoxy resins exhibit low shrinkage, excellent adhesion to a variety of substrate materials [18], high temperature performance, chemical resistance and reactivity with a wide variety of chemical curing agents [19]. The resin forms a highly crosslinked network structure having relatively high stiffness and glass transition temperature (T_g) with high chemical resistance. However, the inherent toughness of the network polymer is low [19]. Elastomeric modification is one of the most frequently used and widely accepted methods for improving properties of epoxy networks. The copolymer of acrylonitrile and butadiene with end carboxyl functional groups, carboxyl-terminated butadiene acrylonitrile (CTBN), can react with the epoxide groups, and hence are popularly employed as a modifier to epoxy. A high level of interfacial adhesion and property improvements are achieved by this elastomer [20, 21]. Bussi and Ishida [22] studied the mechanical properties of blends of diglycidyl ether of bisphenol-A (DGEBA) based epoxy resin and hydroxyl terminated, internally epoxidized polybutadiene rubber. In order to improve mechanical properties, the epoxidized rubber was prereacted with an excess diepoxide to achieve better bonding between the rubber particles and the epoxy continuous phase. They also observed that without rubber prereaction, almost no improvement in the value of critical stress intensity factor, K_{IC} (fracture toughness) was achieved. However, K_{IC} increased with increasing prereacted rubber concentration. From dynamic mechanical analysis (DMA) studies, it was also concluded that the sample containing prereacted rubber exhibited lower T_g than the sample containing the unmodified rubber. Nigam et al. [23] studied the changes in mechanical properties of epoxy cresol novalac (ECN) resin by liquid CTBN modification. Tensile, flexural, and impact strength showed an increasing trend up to 10% CTBN, beyond which a rapid fall took place. From SEM analysis, it was mentioned that well dispersed rubber particles (average size 6 µm) in the epoxy matrix caused restricted plastic deformation. Ratna and Banthia [24] synthesized low molecular weight liquid carboxyl-terminated poly(2-ethylhexyl acrylate) (CTPEHA) and examined its effect on the adhesive properties of epoxy resin. They stated that optimum properties were obtained at about 10 ± 15 phr (phr stands for parts per hundred parts of epoxy resin) concentration of modifier. The great majority of the studies [25–27] involve the chemical modification of epoxy resin with reactive liquid rubber, particularly CTBN copolymer. The micro-structure formed consists of an elastomeric phase finely dispersed in the epoxy matrix with the elastomeric particle diameter of few micrometers or less. These particles enhance the toughness of the unmodified epoxy considerably with only a minimal modification to thermal and mechanical properties [28]. Carbon fiber reinforced plastic (CFRP) is a type of reinforcement which applied in this study. Carbon fibers offer the highest specific modulus and highest specific strength of all reinforcing fibers that makes them suitable for such applications [29]. In previous work [30], we studied the interlaminar fracture toughness for CFRP/epoxy modified with MFC as hybrid reinforced composite. We concluded that initiation and propagation interlaminar fracture toughness in mode I improved significantly by 80 and 44%, respectively, with addition 2 wt% of MFC to CFRP/epoxy composite [30]. The main objective of the present study was to improve the interlaminar fracture toughness of CFRP/epoxy filled with MFC by addition CTBN as liquid rubber as well as study the effect of natural fiber on carbon fiber reinforced epoxy modified with liquid rubber. The effect of addition liquid rubber on CFRP/epoxy filled with MFC on mechanical properties has been investigated.

Experimental

Material

MFC (Celish KY110G, water slurry containing 10 wt% fiber, Daicel Chemical Industries, Ltd., Japan) was used as filler. Plain woven carbon cloth (Pyrofil TR3110M: Mitsubishi Rayon CO., LTD.) was used as reinforcement. Epoxy resin and modified aliphatic polyamines (Japan Epoxy Resins Co., Ltd) were used as matrix and curing agent, respectively. Liquid rubber CTBN copolymer (Nipol DN601, acrylonitrine 20%, Zeon Chemicals, Japan) was used as modifier.

Preparation of MFC

As the MFC (Celish) water slurry containing 10 wt% fiber, the water was removed from the aqueous suspension of MFC by solvent exchange with ethanol. The content of MFC in ethanol was adjusted to be 0.1 wt%. MFC containing ethanol is filtered by vacuum pump to obtain the sheet of MFC. The filtered sheet of MFC was stirred with additional amount of ethanol then sonicated for 10 min by using ultrasonic homogenizer.

Sample preparation

MFC 0.0, 0.5, 1 wt% were prepared to examine the effect of addition cellulose fibers on the mechanical properties of the composite modified with CTBN. The desired amount of epoxy was added into MFC ethanol suspension mixed for 5 min. The modifier content was 10 and 20 wt% based epoxy. Epoxy MFC rubber mixtures were mixed at room temperature for 10 min. The mixture heated for 3 days at 85 °C in an electric oven. The calculated amount of curing agent was added and the mixture was stirred for 5 min and then degassed in a vacuum oven for 10 min. The mixture was hand lay-upped with plain woven carbon fiber which its fraction of volume was the 50 ± 2%. Five specimens were prepared for each test.

Tensile tests

The tensile properties of samples were measured using an Shimadzu Autograph universal testing machine. The specimen gage length was 100 mm and the testing speed was set to 1 mm/min. The specimen dimension was 200 × 25 × 2 according to JIS K7073 [31], glass fiber reinforced plastic (GFRP)/epoxy tabs were attached at both ends of specimen by adhesive.

Bending test

Flexural properties were measured according to JIS K 7074 [32]. Three point bending tests were conducted at room temperature. Dimensions of the specimen for the bending test were 2 mm in thickness, 15 mm in width and 100 mm in length. The span was set to 80 mm.

Fracture toughness test

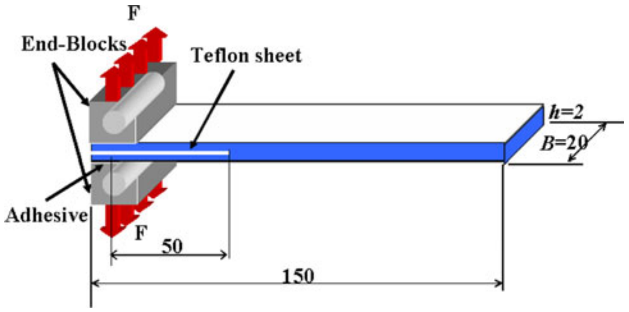

Fig. 1 Geometry of DCB specimen (all dimensions in mm)

The double cantilever beam (DCB) mode I fracture specimen (JIS K 7086 [33]) was employed to characterize the delimination resistance. The corrections for the end block, DCB arm bending and root rotation were considered. DCB tests are conducted using a universal mechanical testing machine. The recommended specimen size is at least 150-mm long and 20-mm wide with an initial crack length (i.e., the length of the insert from the load line) of 50 mm (see Fig. 1). Hinges of the same width as the specimen were attached to allow load application. The mode I interlaminar fracture toughness G_{IC} and G_{IP}, for each MFC and CTBN content was calculated using the modified compliance calibration (MCC) method [34]. The MCC methods were calculated from Eq. 1:

$$G_{\mathrm{IC}} = \frac{3m}{2(2h)}\left(\frac{P_{\mathrm{c}}}{B}\right)^{2}\left(\frac{BC}{N}\right)^{2/3}F,$$

$$G_{\mathrm{Ip}} = \frac{3m}{2(2h)}\left(\frac{P_{\mathrm{p}}}{B}\right)^{2}\left(\frac{BC}{N}\right)^{2/3}F, \tag{1}$$

where G_{IC} is the fracture toughness at initial crack stage, G_{IP} is the fracture toughness at propagation stage, P_{p} is the applied load, C is the compliance corresponding to each crack length, a is the crack length, P_{c} is the initial maximum load, B is the specimen width, $2h$ is the thickness in equation, N is the end block correction factor, F is the large displacement correction factor, and m is the slope of a plot of $(BC/N)^{1/3}$ versus $(a/2h)$.

Dynamic mechanical analysis

Seiko DMS 6100 instruments with a chuck distance of 20 mm was used to perform the DMA studies in order to evaluate tan δ and T_{g}. DMA gives both storage modulus and loss modulus characteristics as a function of temperature. The measurements were carried out at a heating rate of 2 °C/min from 30 to 250 °C at fixed frequency of 1 Hz. The samples were rectangular bars of sizes 40 × 10 × 2 mm.

Scanning electron microscopy observations

Fractured surfaces obtained from mechanical tests were examined by scanning electron microscopy (SEM) using

JSM-7001FD equipment. Prior to SEM observation, all samples were sputter coated with a thin layer of gold to avoid electrical charging.

Results and discussion

Tensile properties

The effect of the CTBN contents on the strain behavior of CF/epoxy composites filled with MFC was investigated up to their failure. Figures 2 and 3 show the tensile strength and young's modulus, respectively. For unfilled composite, the tensile strength decreased with addition 10% CTBN and maintained its decrease at 20%. A gradual increase for composites filled with 1% MFC at 10% CTBN followed by slight decrease with addition 20% CTBN. This may be due to the increase in the relative amount of dissolved rubber as rubber content increases [28]. Although there is tendency to increase strength for composite filled with addition of MFC, the increase was no significant. The Young modulus, determined from the typical stress–strain curve, showed little decrease for composites modified with 10% CTBN followed by insignificant increase for composites filled with MFC at 20% CTBN.

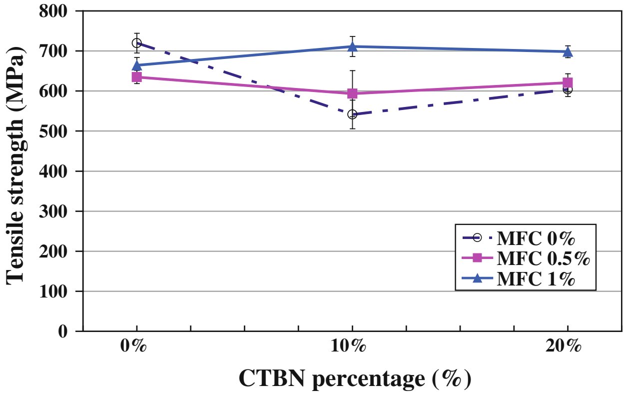

Fig. 2 Tensile strength of each CTBN weight at different ratios of MFC

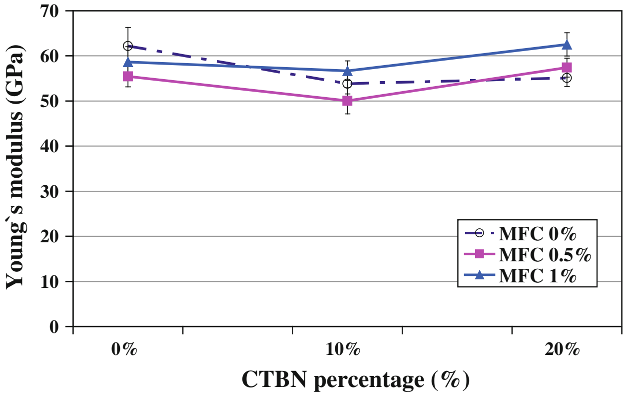

Fig. 3 Young's modulus of each CTBN weight at different ratios of MFC

Flexural properties

The flexural properties of the CF/epoxy composites filled with MFC with addition CTBN in this study are shown in Figs. 4 and 5. It can be seen that flexural strength is slight increased with addition 10% CTBN to CF/epoxy which could be considered as insignificant effect whereas the addition of MFC to the CF/epoxy at same content of CTBN does not show any increase. A gradual fall in flexural strength was noted as the concentration of the elastomer phase increased to 20%. Also, this may be due to the increase in the relative amount of dissolved rubber as rubber content increases, which could be attributed to the fact that the strength and modulus of rubber is much lower than that of the epoxy matrix as well as carbon fiber. In addition, low modulus rubber particles act as stress concentrators and decrease the yield strength [35]. Unmodified CF/epoxy filled with 0.5 wt% MFC showed higher flexural

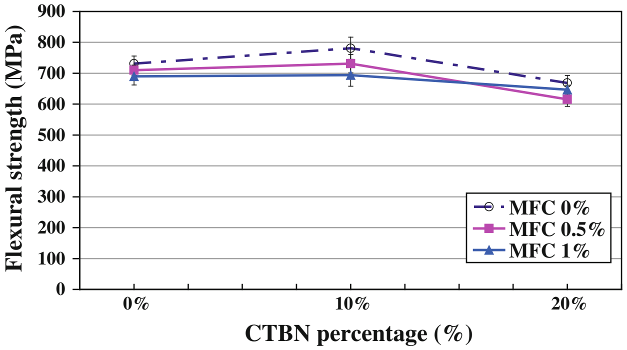

Fig. 4 Flexural strength of each CTBN weight at different ratios of MFC

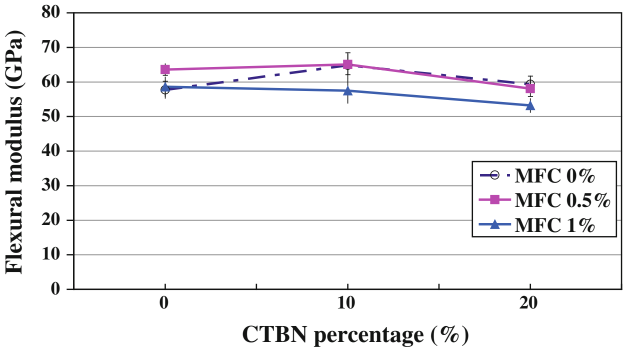

Fig. 5 Flexural modulus of each CTBN weight at different ratios of MFC

modulus compared with unfilled unmodified composite by about 10% maintained its value with addition 10% CTBN followed by a gradual decrease with addition of 20% CTBN.

Dynamic mechanical analysis

Figures 6 and 7 show the DMA data for CF/epoxy composite with varying amounts of CTBN. The glass transition temperature (taken as maximum of the tan δ curve at 1 Hz) was 162 °C for CF/epoxy (Fig. 6), which was equal to the values for other systems [36, 37]. The addition of 10 and 20% CTBN in the unfilled matrix decreased the T_g of the matrix phase by about 7 and 19 °C, respectively. The difference in the T_gs was purely due to the difference in the crosslink density [38]. Similar results have been reported by Verchere et al. [39] for epoxy terminated butadiene/acrylonitrile rubber in an aliphaticamine cured DGEBA. Three effects may be responsible for this observation. One possibly is due to the presence of a small amount of unreacted monomer that plasticize the CTBN. The T_g of epoxy is 162 °C (by DSC) which is higher than the T_g of the CTBN, so this affect cannot be attributed to plasticization by epoxy alone. Alternatively, the lowering of the CTBN rubber T_g may be due to compositional heterogeneity of acrylonitrile in the copolymer chains, which causes compositional fractionation of the acrylonitrile butadiene copolymer during the phase precipitation process. This could cause the nonpolar, butadiene-rich component of the CTBN copolymer to be preferentially expelled from the more polar matrix thus leading to a reduction in the T_g of the dispersed phase because the butadiene-rich CTBN has a lower T_g. A third explanation, offered by Verchere et al. [39], is that after Curing at elevated temperatures and subsequent cooling, thermal hydrostatic stresses are applied to the rubber inclusions due to differences in the thermal expansion coefficients of the rubber and matrix phases. This is believed [39] to raise the free volume in the rubber phase and hence decrease T_g. Compared to unfilled unmodified CF/epoxy composite, filled CF/epoxy with 0.5 and 1 wt% MFC showed increase in T_gs of the unmodified matrix phase by about 12 and 7 °C, respectively. At 10% addition of CTBN, the T_g of filled CF/epoxy with 0.5 wt% MFC is the same as unfilled composite, while with 1 wt% MFC decreased by about 4 °C. This suggests that filled composite with MFC does not increase T_g with addition of CTBN. Figure 7 shows the temperature dependence of the storage modulus (E') of CF/epoxy filled with MFC at 10 and 20% CTBN. The figure shows that the storage modulus of CF/epoxy below T_g is around 5.4 GPa. It is worth to point out that addition of 20% CTBN with MFC fiber content 0.5 and 1 wt% contribute to increase storage modulus compare to unfilled unmodified CF/epoxy indicating that addition MFC to modified epoxy contribute to better heat resistance at given weight. For unmodified CF/epoxy composite, the addition of 1 wt% MFC slightly improved the storage modulus of composite from 5.4 to 5.7 GPa at 40 °C (glassy state). The figure also shows that storage modulus of samples modified with 10% CTBN has tendency to decrease but improved with addition MFC confirming the effect of addition MFC to CF/epoxy contribute to better heat resistance.

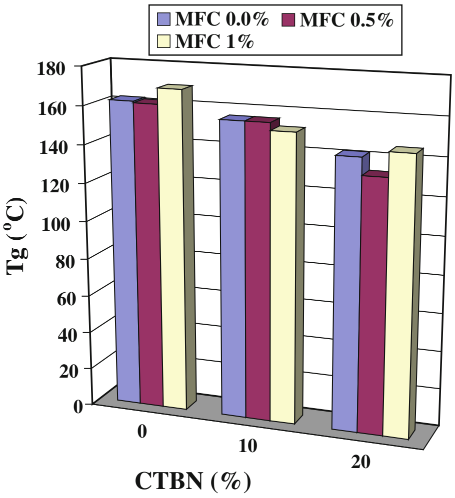

Fig. 6 Glass transition temperature at each CTBN weight at different ratios of MFC

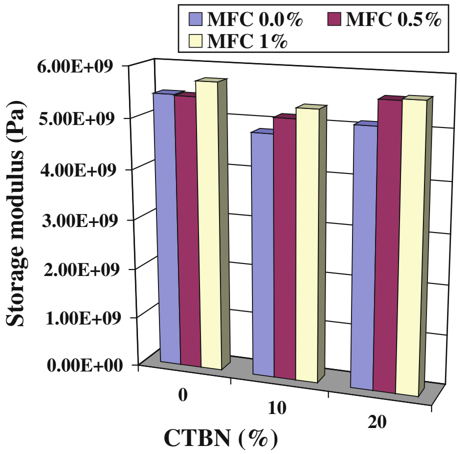

Fig. 7 Storage modulus at 40 °C for each CTBN weight at different ratios of MFC

Mode I interlaminar fracture toughness

DCB tests were performed and the mode I interlaminar fracture toughness, G_{Ic} was determined. The typical load displacement curves recorded during the interlaminar fracture test for CF/epoxy composite filled with MFC

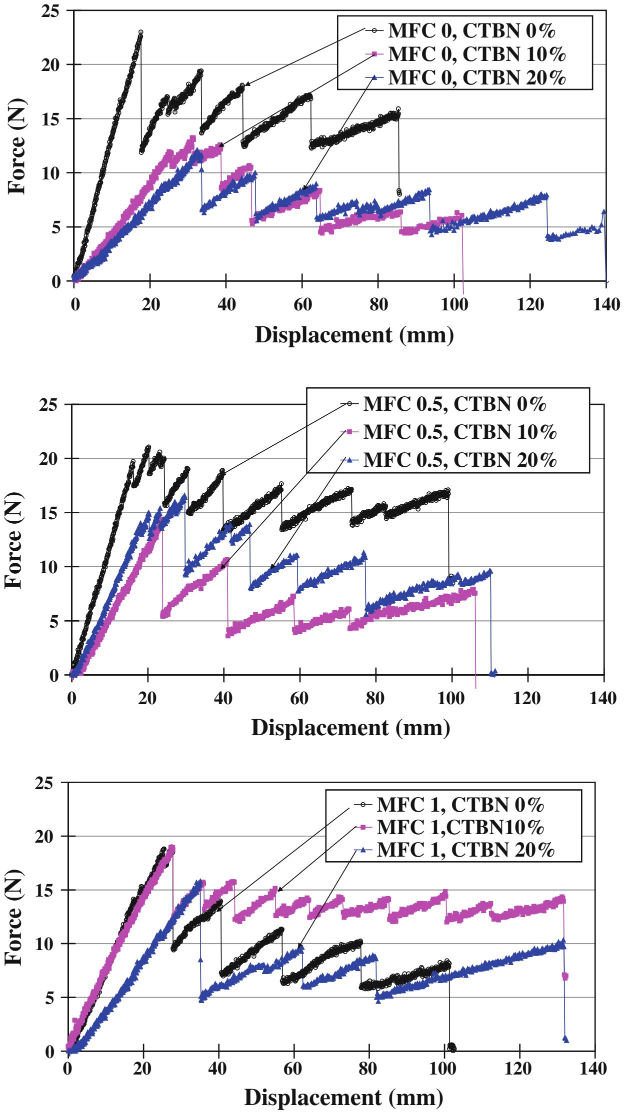

Fig. 8 Force displacement curves for each content of MFC and CTBN

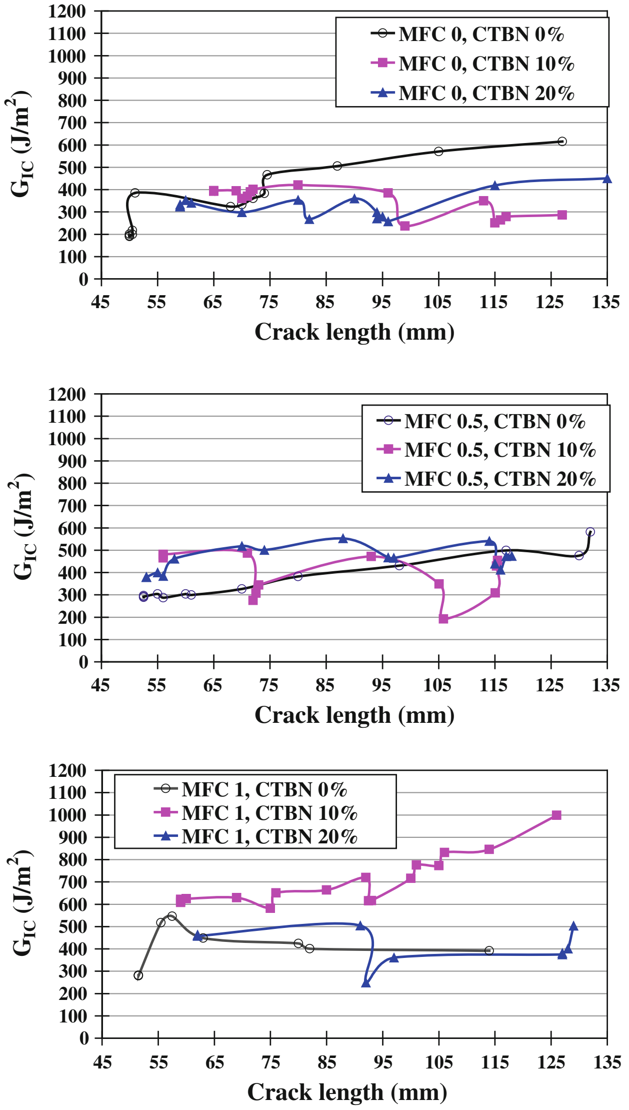

Fig. 9 R-curves for each content of MFC and CTBN

contents modified with CTBN are shown in Fig. 8. Step by step variations in the crack opening load of all specimens are observed. Severe steps are clearly observed for unfilled unmodified CF/epoxy. It is seen that the specimens filled with MFC contents showed more stable and gradual crack growth than unfilled specimens. It was more pronounced that at 1% MFC and 10% CTBN, the crack propagates more stable and gradually as well as almost all peaks rather than first peak showed closed values which could be attribute to higher crack closure force exerted incorporating MFC and CTBN with carbon fibers.

The delamination resistance curves (R-curves) are drawn between crack length (*a*) and the corresponding fracture toughness as shown in Fig. 9. High deviations are noted particularly for some fabric composites, probably as a consequence of the complex translaminar crack growth mechanism acting in the material. It has been observed that inside unfilled unmodified resin rich layers the crack growth rate propagates smoothly as a result of the relatively low tenacity of the polymeric phase with carbon fiber; on the other hand, in the resin rich layers filled and modified with MFC and CTBN, the crack is deflected and pinned by the reinforcing obstacles so that more energy is required, resulting in increasing fracture toughness.

The G_{Ic} value corresponding to first crack initiation is determined from the load point at which the initiation of delamination is microscopically observed on the specimen edge. In this investigation, both delamination initiation and delamination propagation mode I fracture toughness values are plotted in Figs. 10 and 11. The delamination initiation mode I fracture toughness values reported throughout this

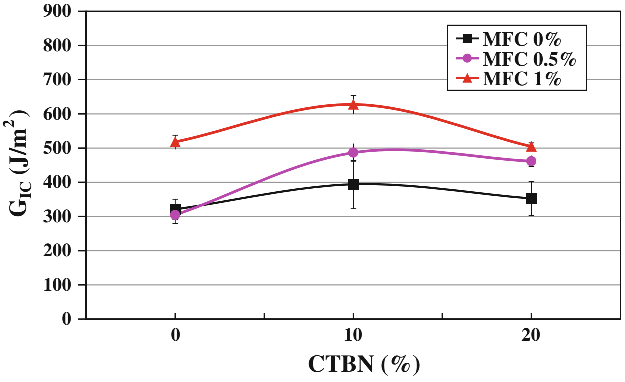

Fig. 10 Initiation fracture toughness for each weight of CTBN at different contents of MFC

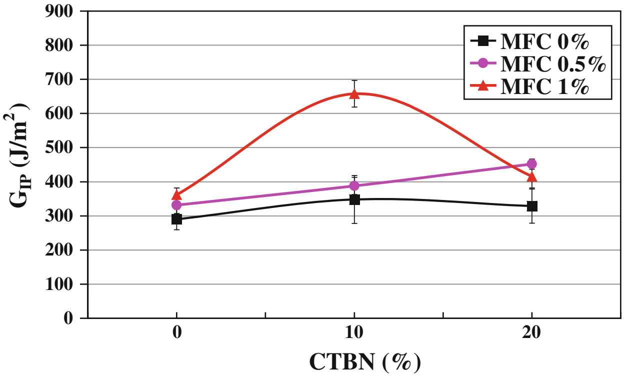

Fig. 11 Propagation fracture toughness at each weight of CTBN for different contents of MFC

investigation correspond to first peak load in the load–crack opening displacement curves, while the delamination propagation mode I fracture toughness values are taken from the plateau region of the R-curves [40, 41].

Various reasons such as intra-laminar delamination, fiber-bridging, micro-cracking, residual stresses, or a combination of these effects of lamina at interface caused the development of transverse intralaminar and unstable crack propagation in DCB tests [42]. The experimental results show that addition of CTBN to composite filled with MFC affects the G_{IC} and G_{IP}. As shown in Fig. 10, the initial fracture toughness was increased about 61% from 320 J/m^2 for unfilled unmodified CF/epoxy to 517 J/m^2 for 1% wt addition of MFC while with addition 10% of CTBN, the fracture toughness was increased by 23% from 320 J/m^2 for unfilled unmodified CF/epoxy to 394 J/m^2 using MCC method. The improved interlaminar fracture toughness is attributed to improved fracture toughness of rubber modified adhesives, originating from the additional toughening mechanisms provided by the rubber particles, such as tearing, cavitation, shear band formation, and fracture of rubber particles [43]. Although the rubber modified composite showed increase in fracture toughness of composite, it imparted only moderate improvements in interlaminar fracture toughness. An established explanation for the poor translation of the toughness of rubber modified resin into composite interlaminar fracture toughness is mainly due to the suppression of the toughening effect in a thin resin film between reinforcing fibers or between substrates, which constrain plastic deformation near the crack tip [44].

A more significant increase was observed for the initial fracture toughness with addition of 1% MFC with addition 10% of CTBN, the initial fracture toughness increased about 96% from 320 J/m^2 for unfilled unmodified CF/epoxy to 627.4 J/m^2. The propagate fracture toughness was further improved by 127% from 289 J/m^2 for unfilled unmodified CF/epoxy to 657 J/m^2. This increase could be attributed to crack deflection processes at nano fiber obstacles in a matrix which proposed to play an important role in the toughening [45]. The theory was described by Faber and Evans [46, 47]. It is assumed that a crack can be deflected at an obstacle and that it is forced to move out of the initial propagation plane by tilting and twisting. Tilting and twisting subsequently continues at further particles if the crack follows a three-dimensional pathway. Therefore, an increase in total fracture toughness of a composite can be expected if this mechanism holds. However, the crack deflection theory cannot fully explain the toughening effect provided especially by the BC nano-fibers. These results suggest that CF/epoxy composite filled with 1% MFC modified with 10% CTBN gives better results which agree with other researches [22, 23]. The fracture toughness decreased with addition 20% of CTBN, this may be attributed to the lowering of cross-linking density in the modified samples. During the curing of epoxy resin, phase separated rubber domains shall occupy the space in between the reaction sites, thereby impairing the cross-linking reaction at that particular site. This, in turn, reduces the cross-linking density of cured systems. Thus, the overall cross-linking density changes by the incorporation of more rubber [48].

SEM observations

The micrographs in Fig. 12a, CF/epoxy filled 0.5 MFC modified 10% CTBN, show stepwise topography at the end of the insert film and fiber/matrix interface debonding, indicating that delamination initiation was dominated by the failure of the fiber/matrix interface. The distinct feature of the fracture surface is matrix deformation between clean fibers devoid of the matrix, indicating tearing (or drawing) of the matrix and interfacial debonding.

Two distinct regions exist in Fig. 12b showing micrograph of CF/epoxy filled 1% MFC modified 10% CTBN:

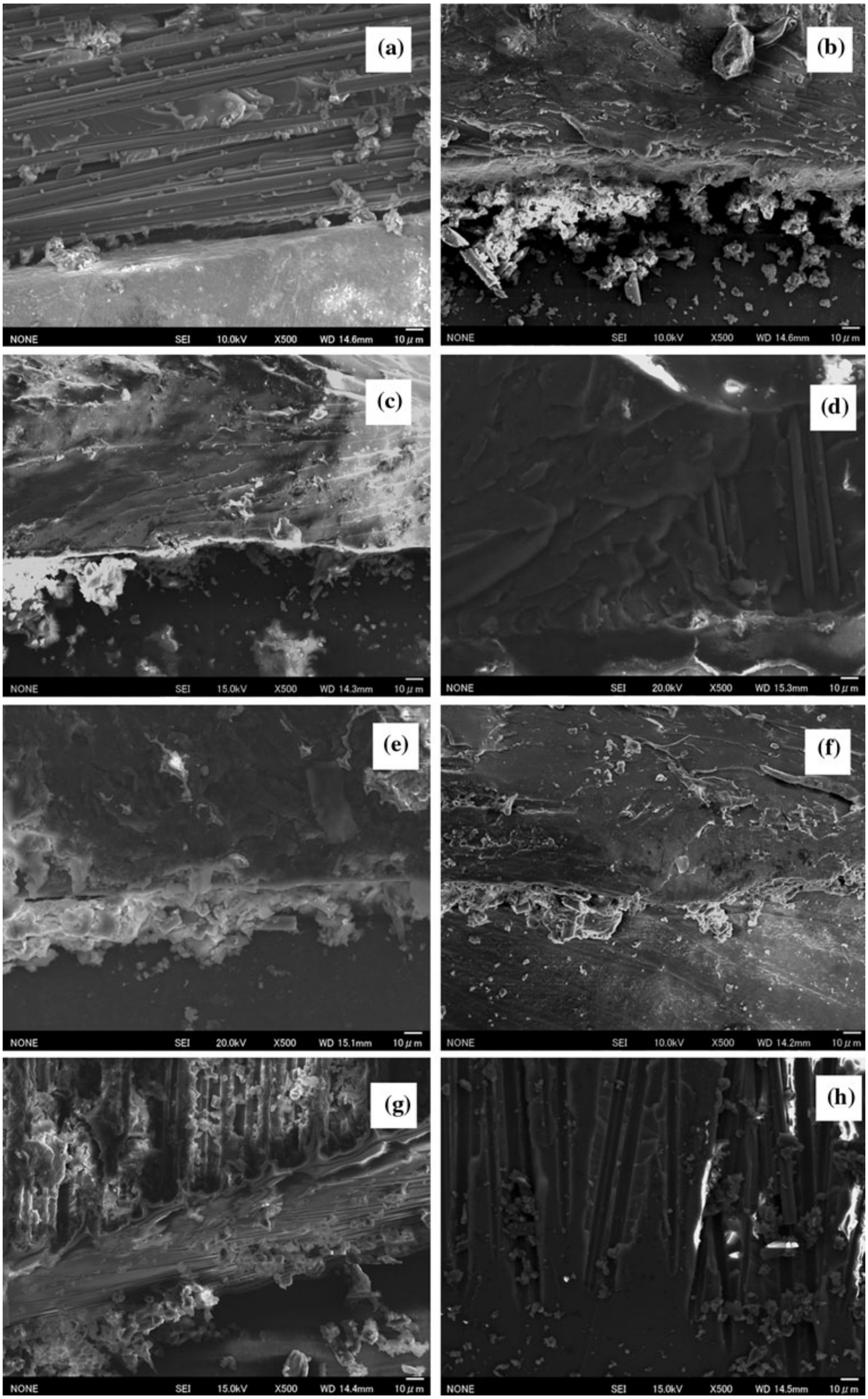

Fig. 12 SEM for different ratios of MFC %/CTBN%, (**a**) 0.5/10, (**b**) 1/10, (**c**) 0.5/20, (**d**) 1/20, (**e**) 0.5/0, (**f**) 1/0, (**g**) 0/10, and (**h**) 0/20 showing the delamination direction at insert tip from the bottom to the top

the first is the rough region showing reinforced adhesion, while the second one in front of the tip of film insert contains remnants of the CFRP substrate. The rough areas of the CFRP suggest a stronger bond exists at the interface as a result of the strengthening effect resulting from the dispersed nanofillers with liquid rubber. In front of the tip, fiber breakage takes place during initiation delimination. Fiber breakage results from fiber–polymer interaction, fiber–fiber interaction, and fiber contact with surfaces of processing equipment. Fiber–polymer interaction promoted a large number of carbon fiber to break at the fracture surfaces resulting in consuming substantial fracture energy.

In Fig. 12c, the fracture surface at CF/epoxy filled 0.5 MFC modified 20% CTBN consists mainly of resin microflow lines and river patterns, which are characteristic of brittle cleavage matrix fracture. However, the river marks in the matrix fracture area of the composite appear not to coincide with the macroscopic crack growth direction, which may indicate various directions of local microcrack growth due to interference of fibers during interlaminar crack growth. The matrix deformation between fibers with interfacial debonding remains the dominant fracture feature accounting for increase fracture toughness.

Figure 12d shows initial delimination of CF/epoxy modified filled 1% MFC modified 20% CTBN. Although a few isolated areas of the cohesive matrix fracture consisting of shoulder like topographic pattern having different radius of curvature can be found, the matrix deformation between fibers with interfacial debonding remains the dominant fracture feature.

The matrix failure for CF/epoxy modified filled with 0.5 and 1% MFC as shown in Fig. 12e and f appears very similar to the plastic deformation associated with shear lip formation for fracture surface of CF/epoxy filled 1% MFC modified 10% CTBN (see Fig. 12b). The extensive matrix deformation between fibers with interfacial debonding as well as fiber breakage noted to be the dominant fracture feature.

Figure 12g and h show stepwise topography at the end of the insert film and fiber/matrix interface debonding for CF/epoxy modified 10 and 20% CTBN, indicating that delamination initiation was dominated by the failure of the fiber/matrix interface. The stepwise fracture surfaces were caused by the interaction of initiation of two delamination fronts, one at the fiber/matrix interface and the other from the insert film, suggesting that the delamination initiated first at the fiber/matrix interface rather than from the resin-rich area at the tip of the insert film for DCB specimens with a tough matrix but a weak fiber/matrix interface [49].

Figure 12g show that addition 10% CTBN to CF/epoxy resulted in high wettability leading to improve interfacial bonding between fiber and matrix. With addition 20% CTBN the fracture occurred predominantly at the fiber/matrix interface as reflected by the bare fibers and cavities left by carbon fibers on the fracture surfaces as shown in Fig. 12h, which indicates poor fiber/matrix adhesion. Although some broken carbon fibers are observed on the fracture surfaces, the fiber-bridging mechanisms, common in mode I fracture of unidirectional composites [50], appear not to be a major energy-absorbing mechanism for the plain weave composite.

Conclusion

In this study, we focused our attention to improve the interfacial adhesion between plain woven carbon fiber (CF) and epoxy matrix filled with MFC modified with CTBN as liquid rubber. The work concentrated on the experimental determination of the tensile, bending, dynamic mechanical, and fracture toughness properties of the CF/epoxy/BC/CTBN composite. The results reveal that a given weight of MFC fiber content 1 and 10% CTBN plays a major role to improve initiation and propagation interlaminar fracture toughness in mode I significantly by 96 and 127%, respectively, which could be attribute to strong adhesion between filled epoxy, carbon fiber, and rubber. This can be explained by SEM at given weight, in front of the insert film, fiber breakage takes place during initiation delimination resulting in consuming substantial fracture energy as well as the rough areas of the CFRP confirm a stronger bond between fiber and matrix. Although there is a slight increase in tensile strength for composite filled with addition of MFC, the increase was no significant. Addition of 20% CTBN with MFC fiber content 0.5 and 1 wt% contribute to increase storage modulus which confirm that MFC contribute to better heat resistance at given weight.

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