Pertanika J. Sci. & Technol. **20 (1)**: 129 – 137 (2012)

ISSN: 0128-7680

Effect of Epoxidized Natural Rubber on Mechanical Properties of Epoxy Reinforced Kenaf Fibre Composites

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ABSTRACT

Kenaf fibre that is known as *Hibiscus cannabinus*, L. family *Malvaceae* is an herbaceous plant that can be grown under a wide range of weather conditions. The uses of kenaf fibres as a reinforcement material in the polymeric matrix have been widely investigated. It is known that epoxy has a disadvantage of brittleness and exhibits low toughness. In this research, liquid epoxidized natural rubber (LENR) was introduced to the epoxy to increase its toughness. Kenaf fibres, with five different fibre loadings of 5%, 10%, 15%, 20% and 25% by weight, were used to reinforce the epoxy resins (with and without addition of epoxidized natural rubber) as the matrices. The flexural strength, flexural modulus and fracture toughness of the rubber toughened epoxy reinforced kenaf fibre composites were investigated. The results showed that the addition of liquid epoxidized natural rubber (LENR) had improved the flexural modulus, flexural strength and fracture toughness by 48%, 30%, and 1.15% respectively at 20% fibre loading. The fractured surfaces of these composites were investigated by using scanning electron microscopic (SEM) technique to determine the interfacial bonding between the matrix and the fibre reinforcement.

Keywords: Biocomposite, kenaf fibre, natural rubber, mechanical properties, toughened

INTRODUCTION

Conventional fibre reinforced composites, such as carbon fibres and glass fibres, can contribute to environmental problems in disposal through incineration. To overcome these problems, environmentally friendly composites are required by utilizing natural fibres as reinforcement in the composites (Marthur, 2006). Mechanical properties of the fabricated composites are governed by reinforcement and matrix properties, as well as the interfacial adhesion between them. Fibre reinforced polymer receives considerable demands in structural application, automotive, recreation and sports tool, as well as furniture and aerospace. Meanwhile, natural fibres such as jute, sisal, coir and banana are major renewable resource materials that are abundantly available throughout the world, especially in the tropical region such as India, Sri Lanka, Thailand, Indonesia, Bangladesh and Philippines. These plant fibres have a great potential to be used as reinforcement in polymer composites (Saheb & Jog, 1999). The attractive features of these fibres include low cost, light weight, high specific modulus, renewability and biodegradability (Ray *et al.,* 2001a; Ray *et al.,* 2001b). For example, kenaf (*Hibiscus cannabinus*, L. family *Malvaceae)* is an herbaceous annual plant that can be grown under a wide range of weather conditions, and kenaf is economically not expensive

Received: 15 January 2011
Accepted: 1 April 2011
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and it is also ecologically biodegradable (Nishino *et al.,*2003). However, kenaf fibres have low strength to resist the failure. This particular behaviour causes fracture in the composite materials to catastrophically occur without any warning. Therefore, composite based rubber-toughened polymer matrix provides extra toughness and delays the failure through dispersion mechanism in the material (Hazleen, 2007).

Epoxy resins are a large family of resins that represent some of the high performance resins available in the market; they are characterized by high chemical and corrosion resistance as well as good mechanical and thermal properties once cured (Ray & Rout, 2005; Saadati *et al.,* 2005). In many applications, however, they have disadvantages of brittleness with poor resistance to crack propagation, low impact strength and low toughness. A number of research have been carried out to increase the toughness, and these include using glass beads, alumina trihydrate and silica. The most successful work involved the addition of a suitable rubber, such as liquid amine-terminated (ATBN) (Saadati *et al.,* 2005), carboxyl-terminated (CTBN) (Calabrese & Valenza, 2003; Thomas *et al.,* 2007); Auad *et al.,* 2001; Jang & Yang, 2000), hydroxyl-terminated (HTBN) (Ozturk *et al.,* 2001; Thomas *et al.,* 2008; Kaynak *et al.,* 2005), epoxy-terminated (ETBN) copolymers of butadiene and acrylonitrile (Thomas *et al.,* 2007), and hydroxyl-terminated and epoxy-terminated polybutadiene to interpenetrating the polymer networks (IPN) structures (Saadati *et al.,* 2005). Another potential method is by addition of suitable rubbers that are copolymers with variable acrylonitrile contents to the uncured epoxy resins. The effects of the acrylonitrile content of the copolymer type, the molecular weight, the concentration and the solubility parameter of the rubber and its functional end groups on the fracture toughness and impact strength have been studied. It has been reported that the modification of epoxy resins by the addition of rigid particle leads to a significant reduction in cost and a considerable improvement in the mechanical, thermal and electrical properties of resin (Saadati *et al.,* 2005; Jayle *et al.,* 1996). Meanwhile, the variation of parameters such as volume fraction of the filler, particle size, modulus and strength of the filler, as well as resin-filler adhesion and toughness of the matrix, leads to improvement in the toughness of resins.

MATERIALS AND METHODS

Materials

The epoxy (Morcote BJC 39) materials used in this study were supplied by Vistec Technology Sdn. Bhd. The kenaf fibres were supplied by Symphony Advance Sdn. Bhd., sieved with sizes ranging from 125 - 355 µm. Polymer used was epoxidized natural rubber (ENR) which was supplied by Rubber Research Industries Malaysia (RRIM). Liquid epoxidized natural rubber (LENR) was prepared using photochemical degradation technique in the polymer laboratory, UKM, according to the method described by Abdullah and Zakaria (1989).

Composite Preparation

Kenaf fibre-epoxy composites were fabricated in the form of plate using a stainless steel mould measuring 165 × 165 × 3 mm. Firstly, the kenaf fibre and the epoxy (with and without LENR) were mixed using mechanical stirrer and with the hardener for a few minutes after that. The mixture was poured into the mould. The mould was then placed between the heated platens of a hot press of 8 MPa pressure at 100°C for 25-50 minutes, depending on the percentages of the fibres used. Then, the plates were cut into the sizes, depending on the types of testing to do, using a diamond blade cutter.

TEST METHODS

Flexural Test

The flexural strength and flexural modulus were determined according to ASTM D790-96. A gauge length of 100 mm was employed with a cross-head speed of 5 mm/min using Shimadzu Universal Testing Machine (model: Autograph AG-X 50kN). The value of the flexural strength and flexural modulus would be determined from the graph of stress-strain curve.

Fracture Toughness Test

The composites were tested for fracture toughness according to ASTM D5045. A span of 80 mm was employed maintaining a cross-head speed of 2 mm/min using Shimadzu Universal Testing Machine. The fracture toughness of the kenaf fibre reinforced epoxy composites can be calculated using Equations 1 and 2.

$$K_{IC} = \frac{PS}{BW^{3/2}} \cdot y \qquad \text{(Equation 1)}$$

$$y = 3\sqrt{(a/w)} \cdot \frac{1.99 - (a/w)[1-(a/w)][2.15 - 3.93\,(a/w) + 2.7\,(a/w)^2]}{2\,[1 + 2\,(a/w)][1 - (a/w)]^{3/2}} \qquad \text{(Equation 2)}$$

where P is the maximum load, S is the span length, a is the notch length, B is the specimen thickness, w is the specimen width, and y is the geometry correction factor.

Morphological Examination

The surface morphology was examined using scanning electron microscope (Philips XL 30) on the impact fractured samples. The fractured samples were coated with a thin layer of gold to avoid electrostatic charging during examination.

RESULTS AND DISCUSSION

Flexural Properties of the Kenaf Fibre Reinforced Epoxy Composite

The flexural strength and flexural modulus for the composites, with and without addition of LENR are shown in *Fig. 1* and *2*, respectively. In general, the flexural strength for kenaf fibre reinforced epoxy composites with the addition of LENR seems to be improved with increasing of fibre content up to 20wt%. For the kenaf fibre reinforced epoxy composites, without addition of LENR, there is an increment in the flexural strength up to 10 wt% of the fibre loadings. Higher fibre loading causes a decrease in the flexural strength of the composites. John and Venkata (2004) have reported that decreases in the mechanical properties for higher fibre loading are due to the poor adhesion between the matrix and the fibre and the presence of voids at the resin-fibre interface (John & Venkata, 2009).

In comparison, flexural strength and flexural modulus for the kenaf fibre reinforced LENR modified epoxy composites were higher than the composite without LENR, except at low wt%. It was observed that the optimum flexural strength was obtained at 20wt% fibre loading for the kenaf fibre reinforced epoxy with the addition of LENR. Meanwhile, the flexural strength of 20wt% of kenaf fibre reinforced epoxy, with the addition of LENR composite, was found to be 25% higher than that of the neat epoxy, whilst the flexural modulus was 60% higher than that of the neat epoxy composite.

As shown in *Fig. 1*, the flexural strength of the composites at 20wt% of fibre loading for kenaf fibre reinforced epoxy with the addition of LENR composite was 56.33 MPa as compared to 43.42 MPa for kenaf fibre reinforced epoxy without addition of LENR composites. The improvement was about 30%. The interfacial strength of the composites and the flexural strength of the matrix are the important factors that determine the flexural strength of a composite. In the case of the kenaf fibre reinforced epoxy with LENR composite, the modification of epoxy with LENR contributed to a strong interface and hence improved the flexural strength of the composite. This could be seen from the SEM micrograph shown in *Fig. 4*.

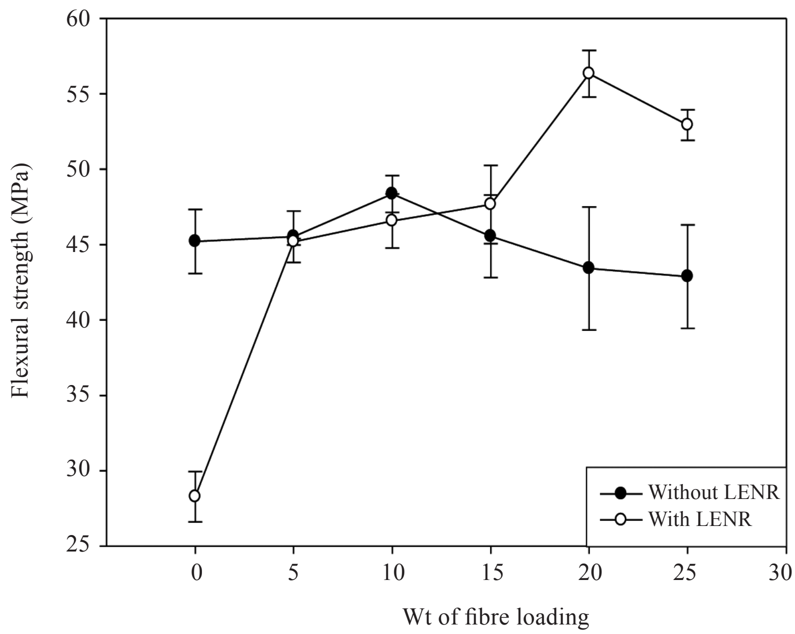

Fig. 1: Flexural strength versus weight percentage of fibre loading used for kenaf fibre reinforced epoxy composite (with and without LENR)

As illustrated in *Fig. 2*, the optimum flexural modulus of the composites at 25wt% of fibre loading for the kenaf fibre reinforced epoxy with the addition of LENR composite was 3.4 GPa compared to merely 2.3 GPa for the kenaf fibre reinforced epoxy composites without the addition of LENR. The improvement was about 48%, and this was mainly due to toughening caused by the LENR modification of the epoxy matrix, which gave rise to good energy absorbing capacity. Rubber domains were formed after the addition of LENR had been found to improve flexibility and toughness. The reactions of LENR used to modify the epoxy matrix, the ring opening reaction between the LENR hydroxyl end group, and hardener may lead to chain extension leading to flexibility and toughness (Kaynak *et al.,* 2005). Furthermore, cross linking during the reaction produces higher chain extensions, and therefore, specimens containing both kenaf fibre and LENR should show better mechanical performances at the same extent of cross linking.

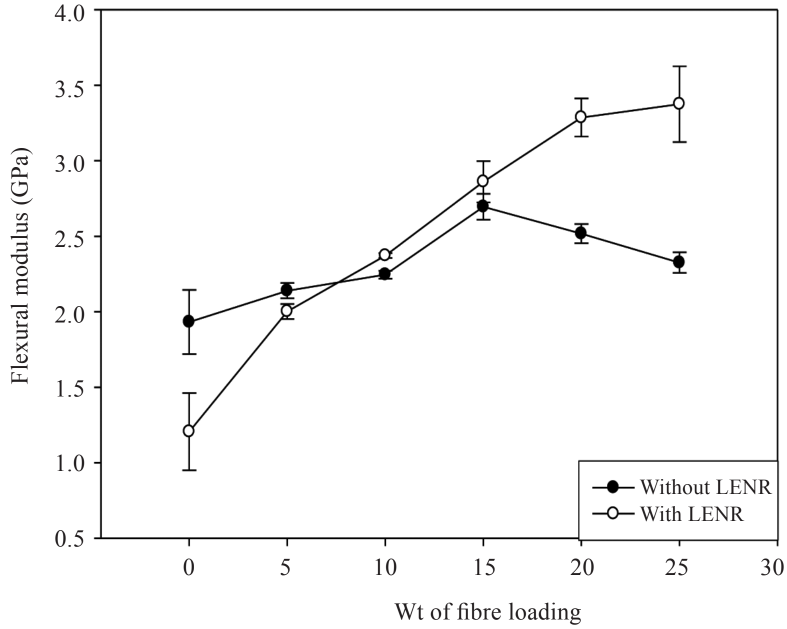

Fig. 2: Flexural modulus versus weight percentage of fibre loading used for kenaf fibre reinforced epoxy composite (with and without LENR)

Fracture Toughness Properties of the Kenaf Fibre Reinforced Epoxy Composite

The fracture toughness properties of the composites are stipulated in *Fig. 3*. The composites, which were prepared with epoxy and addition of LENR as a matrix, showed improvement of the fracture toughness at all weight percentages of fibre loadings. The fracture toughness was relatively higher for the kenaf fibre reinforced epoxy composite as compared to the neat epoxy. The optimum fracture toughness of the kenaf fibre reinforced epoxy, with addition of LENR composite at 20wt% fibre loading, was 2.64 MPa*m$^{1/2}$ as compared to 2.61 MPa*m$^{1/2}$ for the kenaf fibre reinforced epoxy composite without the addition of LENR. This indicated an improvement of 1.15 %. It could also be clearly seen that the fracture toughness for 20wt% fibre loading of the kenaf fibre reinforced epoxy with the addition of LENR composite was 10.9 % higher than the neat epoxy. It could be expected that the composites made using rubber modified epoxy showed improvement in the fracture toughness. This improvement in the toughness was attributed to the rubber particles that enhanced shear localization by acting as stress concentrators. Meanwhile, hydrostatic tension ahead of the crack tip caused rapid cavitations of the rubber. The voided damage zone then blunts the crack, which behaves as if it possesses a much larger crack tip radius. Thus, a larger plastic zone gets associated with this crack and this is the source of toughening effect. The rubber particles that are bonded to the matrix can bear the load in triaxial tension. Thus, the interfacial interaction of rubber particles with the matrix epoxy is desirable for the toughness property which can be attributed by the pre-reaction of the rubber, and thereby improves the toughness by increasing the miscibility of rubber into the epoxy matrix. Hence, some amount of rubber goes into the epoxy matrix and act as plasticizer. If rubber is incorporated into the epoxy network, it will act as a flexibilizer. Both these effects will increase the ability of the matrix to deform under shear (Thomas *et al.,* 2008).

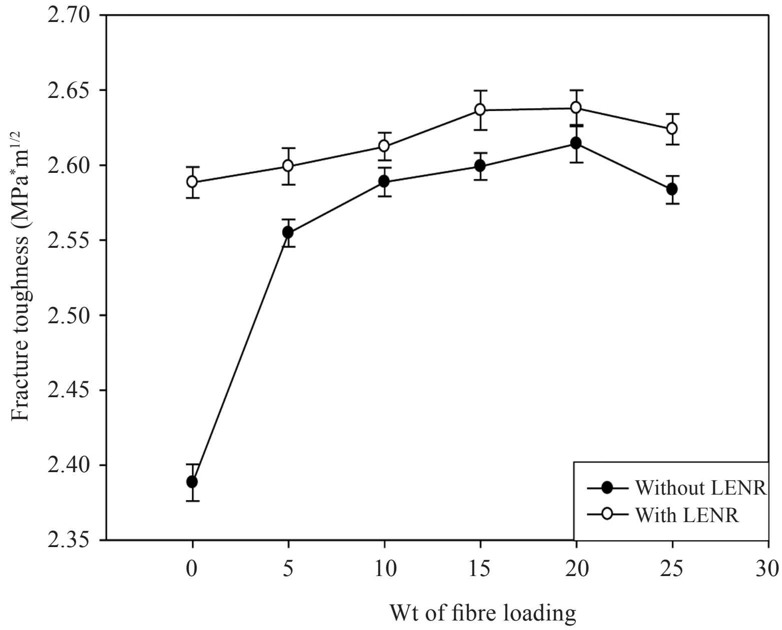

Fig. 3: Fracture toughness versus weight percentage of fibre loading used for kenaf fibre reinforced epoxy composite (with and without LENR)

Morphology of Impact Fractured Surfaces

The fractured surfaces of the specimens were examined using the scanning electron microscope (SEM). The typical fracture surfaces of the kenaf fibre reinforced, with and without LENR modified epoxies, are shown in *Fig. 4*. The composites epoxy without the addition of LENR showed smooth and rivery fractured surfaces with ripples. The relative smoothness of the fractured surface indicates that no significant plastic deformation had occurred irrespective of the presence of some shear deformation lines. The morphology of the LENR modified epoxy shows that the rubber particles are well dispersed in the epoxy matrix. Nonetheless, the fractured surfaces are not very smooth, indicating a ductile manner of the fracture. Yang and Li (1987) have reported that the size of the stress-whitened zone or the amount of deformation lines is proportional to the increase of toughness in the material. The relatively distorted shape of rubber domains in these epoxy resin matrices is supposed to be attributed to the higher amount of plastic deformation. The deformation lines are propagated through rubber domains, promoting stress transfer between the particles and the epoxy matrix. The same finding has also been reported by Thomas *et al*. who operated the yielding process throughout the matrix, whereby, a homogenous distribution of smaller particles is necessary (Thomas *et al.,* 2008). This morphological structure in *Fig. 4* is believed to be responsible for the higher toughness performance of the kenaf fibre reinforced LENR modified epoxy composites. Thus, the uniformly distributed rubber particles act as stress concentrators and exhibit higher toughness than the kenaf fibre reinforced unmodified epoxy composites.

CONCLUSIONS

This work clearly shows that introducing LENR to epoxy matrix can improve the mechanical performance of the kenaf fibre reinforced epoxy composites. The results have shown that the

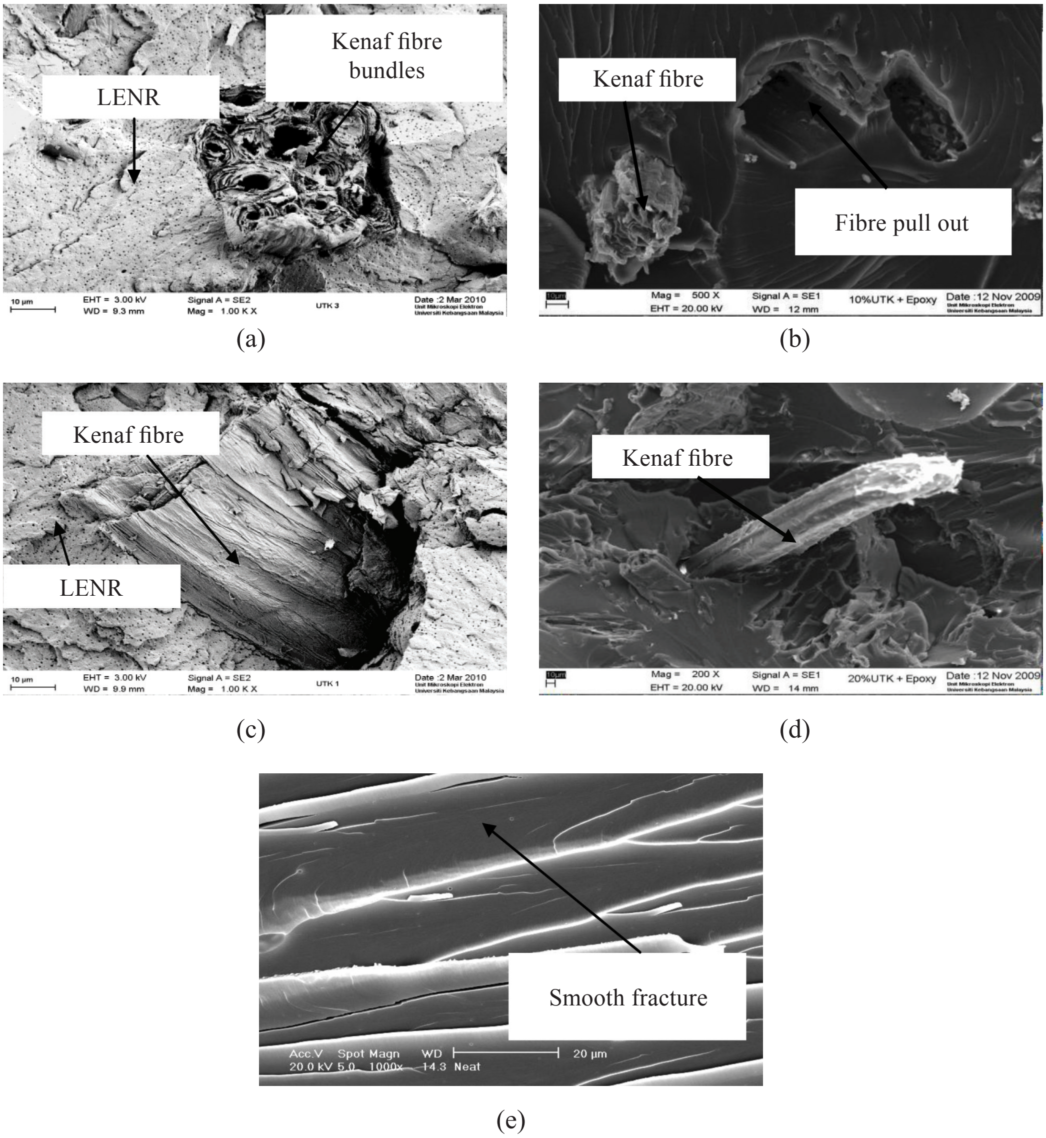

Fig. 4: The SEM micrographs of the fractured surface of kenaf fibre-epoxy composites for; (a) 10% fibre loadings with LENR; (b) 10% fibre loadings without LENR; (c) 25% fibre loadings with LENR; (d) 20% fibre loadings without LENR, and (e) neat epoxy

addition of liquid epoxidized natural rubber (LENR) improved the flexural modulus, flexural strength and fracture toughness by 31%, 30% and 28%, respectively, at 20wt% of the fibre loadings. When LENR reacted with epoxy during mixing process, the path of the reaction could change. This reaction could cause the flexibility and toughness to increase. Furthermore, the formations of rubber domains in the epoxy matrix led to flexibility and improved toughness. The distorted shape of rubber domains in the epoxy resin matrices was attributed to the higher amount of plastic deformation. These deformation lines are propagated through rubber domains, promoting stress transfer between the particles and epoxy matrix, and this consequently increases its toughness.

ACKNOWLEDGEMENT

This work was supported by the MOSTI under grant 03-01-02-SF0223. The authors are grateful to the KPT, Universiti Kebangsaan Malaysia and Universiti Teknologi MARA for sponsoring this research.

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