Mechanical Properties of Epoxy Matrix Composites Toughened by Liquid Epoxidized Natural Rubber (LENR)

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Abstract: Epoxy matrix widely used in polymer composites as a reinforcement material due to its outstanding performance. Epoxy matrix exhibit good mechanical and thermal properties such as good stiffness, dimensional stability and high heat distortion temperature. However, it is known that epoxy had brittleness and exhibit low toughness. This study investigates the mechanical properties of brittle epoxy resin when modified by liquid epoxidized natural rubber for toughening purposes. Liquid epoxidized natural rubber (LENR) was introduced to the epoxy with five different loadings of 1%, 3%, 5%, 7% and 9% by weight. The mechanical strength (flexural strength, flexural modulus, tensile strength, tensile modulus and impact strength) of the rubber toughened epoxy composites were investigated. The results showed that the addition of liquid epoxidized natural rubber (LENR) had improved the flexural modulus, flexural strength and impact strength by 47%, 40%, and 22% respectively at 3% loading.

Keywords: epoxy, liquid epoxidized natural rubber, mechanical properties, toughened

1. Introduction

Epoxies are defined as well-established thermoset cross-linked polymers in which the cross-linking is derived from reactions of the epoxy group. Commercial epoxy resin contains aliphatic, cycloaliphatic, or aromatic back bones. The most widely used is epichlorohydrin and bisphenol-A derived resin. Epoxies were developed, principally by Ciba AG (Switzerland) and the Dow and Raynolds Co. (USA) [1-3]. Epoxy resins consist of three-dimensional networks or agglomerates of moderate molecular weight. As the molecular polymerize through crosslinking, their rotational and rotational freedom is reduced, which reduces the chances of the primary bonds to be set up with adjacent molecules. The tensile and comprehensive strength of epoxy resins are increased if the distance between crosslinks is shortened. Shorter crosslink distances imply high volume concentration of epoxy group and hence a higher probability of reaction of all the epoxy groups. Epoxy resin also have good properties with good stiffness, specific strength, dimensional stability, high heat distortion temperature and chemical resistance [1, 4-5, 21]. For these reasons, it is also the most common polymer used in fibre and filler reinforced polymers for structural applications. However, epoxy is known to be brittle with poor mechanical properties, in terms of strength and toughness, thus preventing the use of this polymer in applications that require stability and high mechanical performance.

Many researchers reported that to toughen epoxy resin, a common method is introducing particles in the resin, including liquid rubbers, thermoplastics, copolymers, silica nanoparticles, silicate layers, core shell particles, and combinations of these [6-10]. Synthetics rubbers containing methyl, hydroxyl, carboxyl, anhydride or thiol groups potentially react with the epoxy resins. Barcia et al. [11] used hydroxyl terminated polybutadiene (HTPB) as surface modifier in carbon fiber reinforced, epoxy matrix composites. Rubbers with carboxyl groups in the chain however may be cured with the epoxy resins. Nigam et al. [13] studied the changes in mechanical properties of epoxy cresol novolac (ECN) resin by liquid carboxy terminated copolymer of butadiene acrylonitrile (CTBN) modification.

Extensive study had been carried out to increase the toughness, and these include using glass beads, alumina...
The mechanical tests were carried out as follows the sample conditioned at 20°C and 65% RH. A three-point flexural test and tensile test was performed on a Testometric machine model M350-10CT according to ASTM D790-96 and ASTM D 638 M-9 1a, respectively. Five specimens of dimensions 127 mm × 12.7 mm × 3.2 mm were prepared. Sample were used to obtain the average value for flexural strength and modulus with loading head speed of 1.35 mm/min and span length was fixed at 50 mm. All tests were performed at room temperature. To determine the mechanical performance of impact strength, Izod test was carried out using Digital Impact Testing RR/MT according to standard test method of ASTM D256-9 1a. Six samples of rectangular with dimensions of 60.3mm × 12.7 mm × 3.2 mm, with a notch at the center were used for this testing.

Table 1 Compounding formulation used in Epoxy/LENR blend preparation

<table>
<thead>
<tr>
<th>Epoxy</th>
<th>LENR</th>
<th>Hardener</th>
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<tbody>
<tr>
<td>70 phr</td>
<td>1.00 - 9.00 phr</td>
<td>30 phr</td>
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mechanical performances at the same extent of cross linking.

![Fig. 1 Flexural strength of epoxy rubber toughened composites.](image1)

![Fig. 2 Flexural modulus of epoxy rubber toughened composites.](image2)

### 3.2 Tensile Strength of the Composites

The tensile strength and tensile modulus of the composite are shown in Figure 3 & 4. At 1% addition of LENR, the tensile strength seems to be improved about 20%. Compared with neat epoxy, the addition of 1% LENR will absorb more energy, it may explain the improved in tensile properties and the toughening and strengthening mechanism of the polymer matrix [3-4, 16]. However, the tensile strength start to decrease at higher addition of LENR. This was due to higher amount of rubber content will decrease the stiffness of the composite. The same trend also observed with tensile modulus. Tensile modulus is a measure of the stiffness of an elastic isotropic material and is a quantity used to characterize materials. Due to elastomeric behavior of LENR, the decreases in modulus was expected when it is added to the thermoset resin [2]. This reduction in tensile strength is due to the incompatibility and poor interfacial adhesion between the epoxy and the LENR in the blend system. It also may due to faster reaction between hardener and epoxy than any possible reaction between LENR and epoxy/hardener mixture [3-4].

![Fig. 3 Tensile strength of epoxy rubber toughened composites.](image3)

![Fig. 4 Tensile modulus of epoxy rubber toughened composites.](image4)

### 3.3 Impact Strength of the Composites

The impact properties of the composites were also given in Figure 5. Since epoxy is a brittle polymer, rubber modification increases its impact strength. When LENR was added to the neat epoxy, the impact strength of the neat epoxy was increased from 5.49 to 6.38 KJ/m². Moreover, with the addition of 3% LENR, the maximum of impact strength value was obtained (7.07 KJ/m²). It was about 22% improvement. This increase impact strength may also be correlated with the toughness improvement [3]. LENR addition can be attributed to the increases in entanglement between epoxy resin, and the increase in main chain mobility. Under low loading rates, the entanglement increases the toughness of the specimens with the flexibility effect of LENR molecules [3-4].

### 4. Summary

In this study, the effects of LENR in epoxy were investigated on mechanical properties. Composites with different LENR ratio were prepared by controlling the micro bubbles formation before curing process. Mechanical tests show epoxy-LENR composites exhibit better modulus of elasticity (MOE), modulus of rupture (MOR) and impact strength than neat epoxy composites. The results have shown that the mechanical behavior of
neat epoxy was improved with LENR addition. The results have shown that the addition of liquid epoxidized natural rubber (LENR) improved the flexural modulus and flexural strength by 47% and 40%, respectively, at 3% of LENR addition. 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 matrix led to flexibility and toughness to increase. Furthermore, the formations 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. It was observed that the optimum percentage of LENR addition was at 3% addition of LENR.

References