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Synthesis And Characterization Of Liquid Natural Rubber As Impact Modifier For Epoxy Resin

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Abstract

Liquid natural rubber (LNR) with a molecular weight $\bar{M}n = 16\times10^3$ was prepared by the depolymerization of deproteinized natural rubber latex (DPNR). The liquid natural rubber (LNR) was characterized by FTIR and H'NMR spectroscopic analysis. LNR was premixed with the epoxy resin (EP) and cured with a diamine curing agent for 1 h at 100 °C and post cured at 110 °C, for 2 h in air oven. The modified EP containing different contents of LNR (5, 10, 15 and 20 phr) were evaluated. Thermal, mechanical and morphology properties were determined. The fracture toughness (K_{IC}) of both unmodified and modified EPs were determined on static loaded single edge notched (SEN-B) specimens at room temperature. The glass transition temperatures (T_g) of the modified EPs were decreased with increasing LNR content. The strengths and modulus of EPs were slightly reduced with the incorporation of LNR. The effect was also reflected in the significant increase in the tensile strain of modified EP. Fracture toughness of the EP was observed to increase with the presence of LNR. The toughening effect became more apparent as the testing speeds were increased from 1 to 500 mm/min. Fracture surface analysis by scanning electron microscopy (SEM) revealed the presence of a two-phase morphology.

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Key words: Epoxy resins, Liquid natural rubber (LNR), Modified Epoxy Resin, Toughening, Impact Modifiers

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1. Introduction

Epoxy resins (EPs) are considered as one of the most important classes of thermosetting polymers. They have been used extensively as high performance adhesive composite materials, due to their outstanding mechanical and thermal properties such as high modulus, tensile strength, low creep, high glass transition temperature, high thermal stability, and moisture resistance [1]. In the cured state, EPs are brittle materials that have fracture energies some two orders of magnitude lower than modern thermoplastics and

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other high performance materials [2]. In order to remain competitive as materials of choice for many applications such as adhesives and composite matrices, epoxies should be modified to improve their fracture toughness.

One of the successful methods to toughen EPs is the incorporation of rubber phase into the brittle epoxy matrix, which may be achieved by the use of reactive liquid rubber or preformed rubber particles [3]. Carboxyl, epoxy and amine terminated butadiene-acrylonitrile copolymers (CTBN, ETBN and ATBN) are widely used as toughening agents. These rubbers are initially miscible with the epoxy but during the polymerization the rubber phase separates due to slight immiscibility with the matrix. At proper concentration of rubber, the dispersed rubber phase can improve the toughness without a significant decrease in the other properties of epoxies [4]. The improvement the toughness of rubber-toughened epoxies has been associated with three main toughening mechanisms; crazing, shear banding and elastic deformation of rubber particles. These mechanisms can either act alone, or together, to produce the toughening effect [5]. Many efforts have been made to understand the behavior of the dispersed rubber phase, which separates from the resin before gelation. Sultan and McGarry found that the toughening mechanisms are affected by the rubber particle size [6]. Chen and Jan extensively studied the effect of matrix ductility on the fracture behavior of rubber toughened EPs [7]. Verchere *et.al.*, showed shear deformation of the matrix as the major toughening mechanism [8]. Bagheri and Pearson studied the role of blend morphology in rubber-toughened polymers [9].

Evaluation of effective crosslink density of matrix resin, measurement of glass transition temperature T_g, cure characterization through differential scanning calorimetry (DSC) and dynamic mechanical analysis (DMA) of diglycidyl ether of bisphenol -A (DGEBA) modified with CTBN liquid rubber have also been reported [10]. Recently, some of saturated liquid rubber such as polysulphide [11], acrylate [12], silicone [13] and polyurethane [14] have been reported in the literature as replacement for CTBN. An attempt to toughen the EP using a polyurethane (PU) prepolymer as a modifier via interpenetrating network (IPN) grafting has also been reported [15]. For this purpose, a PU prepolymer has been synthesized based on hydroxyl-terminated polyester resins and used as a modifier for EP at different concentration. Ratna and Banthia [16,17] showed that carboxyl terminated poly(2-ethylhexylacrylate) (CTPEHA) liquid rubber can be used as an impact modifier for EP cured with ambient temperature hardener. However, carboxyl terminated oligomers can only be synthesized by bulk polymerization, which is difficult to control. Qian *et.al.,* [18] studied the synthesis and application of core-shell rubber particles as toughening agents for epoxies. The effect of epoxidized natural rubber (ENR 50 mol %) on the curing behaviors and adhesive strengths of an epoxy (DGEB-A) and dicyandiamide/2-methyl imidazole system was studied by Hong and Chan [19]. Many works in toughening of EP have been reported [10- 20].

This paper reports on our attempt to synthesis reactive liquid natural rubber (LNR) and to investigate its effect as impact modifier for EP. The present work attempts to discuss thermal, mechanical and morphology properties of fracture surface of the above mentioned LNR modified EP.

2. Materials

Deproteinized natural rubber latex (DPNR) having 22 % dry rubber content was supplied by Rubber Research Institute Malaysia (RRIM). Formic acid 98 %, hydrogen peroxide 30 %, methanol was obtained from Fisher Scientific UK. Clear EP 331 with epoxide equivalent weight 182-192 was a liquid diglycidyl ether of bisphenol A, and curing agent was clear epoxy hardener 8161 (isophorone diamine) with amine value 260-284 (mg KOH/gm). Both EP and curing agent were obtained from Euro chemo-pharma, Sdn. Bhd.

3. Experimental

3.1. Preparation of liquid natural rubber (LNR)

The 500 g of deproteinized natural rubber latex (DPNR) was stabilized by adding appropriated amount of nonionic surfactant and sodium chlorite, with stirring at 50 ºC for 2 h. 100 ml of hydrogen peroxide was then added to the mixture. After few minutes the pH was reduced to a value of 5 by adding small amount of diluted formic acid (20 %). The depolymerization reaction took place at 50 ºC with stirring for 14 days. The appropriate amount of methanol was added to the mixture to precipitate LNR.