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The overall effect of reactive rubber nanoparticles and nano clay on the mechanical properties of epoxy resin

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ABSTRACT

Epoxy resin, a thermoset polymer matrix used for technical applications; exhibit some outstanding properties such as high modulus, high chemical resistance and high dimension stability. However, the high crosslink density of epoxy makes this material brittle with low impact strength and poor resistance to crack propagation, which limits their many end use applications. It is an important objective to explore new routes toward toughening of epoxy resins without affecting stiffness, strength, and glass temperature. The main objective of this work is to incorporate reactive rubber nanoparticles (RRNP) and organically modified nanoclay (Cloisite-30B) into epoxy matrix with the aim of obtaining improved material with higher toughness without compromising the other desired mechanical properties. Epoxy hybrids nanocomposites containing RRNP, Cloisite-30B and RRNP/Cloisite-30B mixture were synthesized and characterized to compare the different properties which normally result from the use of single filler and hence aiming to improve toughness/stiffness balance.

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1. Introduction

Epoxy polymers are widely used for the matrices of fibre-reinforced composite materials and as adhesives. When cured, epoxy polymers are amorphous and highly-crosslinked (i.e. thermosetting) polymers. This microstructure results in many useful properties for structural engineering applications, such as a high modulus and failure strength, low creep, and good performance at elevated temperatures. However, the structure of such epoxy polymers also leads to a highly

undesirable property in that they are relatively brittle materials, with a poor resistance to crack initiation and growth.

So, toughening of brittle epoxy thermosets has been intensively studied in the past few decades since the lack of toughness is one of the major reasons limiting their more widespread engineering applications (Hsieh, Kinloch, Masania, & Taylor, 2010).

One of the most successful strategies of improving the fracture toughness of epoxy is to introduce a second phase into the epoxy matrix. In these systems, the fracture

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toughness can be increased by forming multiphase morphology with the ability to initiate various toughening mechanisms during crack growth (Bashar, Sundararaj, & Mertiny, 2011).

One of the successful methods used to toughen (EP) epoxy is the incorporation of the rubber phase into the brittle epoxy matrix, which may be achieved by the use of reactive liquid rubber or preformed rubber particles. The compatibility between liquid rubber and resin must be matched carefully in order to achieve phase separation during cure and simultaneously provide adequate interfacial adhesion (Pascault, 1995). The phase-separated rubber particles are presumed to act as stress concentrators initiating energy absorbing “toughening” processes. According to the literature, the most common micromechanical processes responsible for the increase in fracture toughness are localized shear yielding of the epoxy matrix, plastic void growth in the matrix, which is initiated by cavitation or debonding of the rubber particles, and rubber particle bridging behind the crack tip (Yi, Wiedmaier, & Schmauder, 2015).

The early advances of McGarry and the re-searchers at B.F. Goordrich were the compatibility matching that has been achieved by varying molecular architectures and reactive end groups of liquid rubbers, such as butadiene acrylonitrile copolymers containing carboxyl (CTBN), amine (ATBN), or epoxy (ETBN) reactive end groups. Other elastomeric modifiers that have been studied included acrylate elastomers (Ratna & Banthia, 2004) and polysiloxanes. Liquid polyethers like poly(propylene oxide) (Harani, Fallahi, & Bakar, 1998) or poly(tetrahydrofuran) have also been used as toughening agents. Modification of their end groups in order to tailor phase separation behavior and adhesion to the matrix via chemical bonds have easily been achieved. However, improvement of fracture toughness by the addition of liquid rubber modifiers is frequently associated with softening of the matrix due to matrix flexibilization. This is attributed to the fact that the modulus of the modifier is much lower than the modulus of the matrix.

Nanocomposite technology using organophilic layered silicates as an *in-situ* route to nano-reinforcement offers new opportunities for the modification of thermoset micro-mechanics. Large improvements of mechanical and physical properties including modulus (Kojima et al., 1993), barrier properties, flammability resistance, and ablation performance have been reported for this type of material at low silicate content. In principle, it should be possible to compensate matrix flexibilization via matrix reinforcement using organophilic layered silicates.

Polymer/layered silicate nanocomposites were first developed by researchers from Toyota based on the thermoplastic polyamide-6 (Sharmin et al., 2011) and have been extended to thermosets by Giannelis and Pinnavaia (Messersmith & Giannelis, 1994). Polymer/layered silicate composites are usually divided into three general types: conventional composites with the silicate acting as a filler on the microscale, intercalated nanocomposites based on the insertion of a polymer in between the silicate layers which remain in a long-range order and exfoliated nanocomposites in which individual silicate layers are dispersed in the polymer matrix. Only few attempts have been made so far to combine liquid rubbers and layered silicates to achieve hybrid nanocomposites. Many

patents are dealing with a particulate of inorganic fillers which have been extensively used in combination with liquid rubbers to enhance the mechanical properties of epoxy resins and other polymeric matrices in industry (MÜlhaupt, 1994). Also another family of epoxy hybrid composites is materials containing both glass beads and liquid rubbers (Michael, DiBerardino, & Raymond, 2009).

Therefore, the main objective of this work is to incorporate both reactive rubber nanoparticles (RRNP) and organically modified nanoclay (Cloisite-30B) into epoxy matrix with the aim of obtaining improved material with toughness higher than neat epoxy, epoxy/clay and epoxy/RRNP hybrids without compromising the other desired mechanical properties. Epoxy hybrid nanocomposites containing both fillers; organo-modified montmorillonite (o-mmt) and reactive rubber nanoparticles (RRNP); were synthesized in order to combine their enhancing properties which normally result from the use of the single fillers and hence aiming to improve toughness/stiffness balance. Although the fracture and deformation behavior of the epoxy–nanocomposites have been intensively studied at the macro-level over the past decade, less attention has been paid to the stress transfer mechanism in the nano to micro-level behavior of nanoclay and nanorubber interactions with epoxy.

2. Experimental methods

2.1. Materials

Oleic acid is a fatty acid (Adwic co) as a surfactant, potassium hydroxide (TOK co), toluene (fision, AR), chloroform (Aldrich, 99.8%) are all of analytical grade and were used as received. Deionized water was used for all latices. Divinylbenzene (Merck, inhibited by tert-butyl catechol) as a crosslinker was washed over 10 wt% NaOH solution. Styrene-co-butadiene rubber (SBR-latex) (Sika Co.), benzoyl peroxide (BPO) (moistened with 25% water, Loba Chemie), ammonium persulphate (Morgan, 98%), and HCL (37%, Aldrich CO), methanol (95%, Aldrich CO) were used as received.

In our research investigation, we used SBR-latex; supplied by Sika Company; which has the formulations shown in Table 1. This latex contains anionic and nonionic surfactants; (Sodium dodecyl sulfate, SDS with HLB = 8) and (Polyoxyethylene (100) stearyl ether, Brij 700 with HLB = 19), respectively. The HLB value represents the tendency of an emulsifier (surfactant) to act as an oil-soluble or as a water-soluble type of emulsifier (Becher, 1965). The HLB values for the SBR latex surfactants indicate both oil and water soluble substances,

Table 1 – Chemical composition of commercial SBR-latex (Ohama, 1995).

| Material | Parts by weight |
|-----------------------|-----------------|
| Styrene | 64.0 |
| Butadiene | 35.0 |
| Vinyl carboxylic acid | 1.0 |
| Nonionic surfactant | 7.0 |
| Anionic surfactant | 0.1 |
| Water | 105.0 |

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