Tensile Properties of Nanoclay (NC)-Acrylonitrile Terminated Polybutadiene (ATBN) Modified Epoxy Composites

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ABSTRACT

Brittleness of cured epoxy resins had limited its application in many fields. Modification of epoxy resin is one of the methods to improve the properties of cured epoxy resins. By using a liquid Acrylonitrile Terminated Polybutadiene (ATBN) rubber as a toughening had improved those weaknesses but it sacrifices the tensile properties. Therefore, in order to produce a toughened and strong mechanical properties of the m-Ep resin, nanofiller was introduced. In this work, the effect of nanoclay (NC) on the physical and mechanical properties of ATBN-m-Ep resin was studied. The different amount of NC is added to reinforce the modified epoxy (m-Ep) resin at the loading of 0.05%, 0.3%, 0.5%, 0.7% and 1% by weight. The mechanical properties of the nanocomposites were carried out by conducting the tensile testing. Meanwhile, for the physical testing, the hardness and density measurement was done. Results revealed that all the properties are improved except the tensile strain. An improvement was achieved at low filler

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loading. Based on the results, the optimum amount of NC was obtained at 0.05 wt. % and it is attributed to the some factors which are a good interaction between nanofiller and m-Ep interfaces and homogeneous dispersion of NC throughout the matrix resulting the formation of exfoliated NC reinforced ATBN-m-Ep composites.

Keywords: *Modification of epoxy resin, physical and mechanical properties, nanoclay, Acrylonitrile Butadiene Terminated Polybutadiene*

Introduction

Thermoset polymers have been widely used in engineering components, matrix and adhesives for natural and synthetic fiber reinforced composites due to their excellent mechanical properties as compared to those of thermoplastic polymers [1]. However, since they are usually brittle and vulnerable to crack, liquid rubbers such as Acrylonitrile Terminated Polybutadiene (ATBN) is introduced to the polymer matrices to increase the toughness properties, with compromise the strength of thermoset polymers.

The addition of ATBN to polymer often increases its toughness, however decrease the strength is due to the flexibility of ATBN chains in the epoxy networks [5]. ATBN has a tendency to form an elastomeric particles upon curing process. The formation of elastomeric particles was formed dual phases and it might happen due to the less polarity of ATBN in the polar thermoset resin. The polarity of ATBN represents to amount of hydrocarbon and epoxide parts which refers to the non-polar and polar respectively. Long hydrocarbon of polymer chains makes the ATBN less polar and hence reducing the interfacial adhesion. Therefore, it is a good way to reinforce the modified thermoset resin with inorganic nanofiller in order to increase the strength. Inorganic nanofillers have gained acceptance as a reinforcing agent because of their low cost and ease of fabrication. The incorporation of nano particle in the polymer matrices have been widely studied and numerous researchers agreed that the positive impact of nanofillers on the physical and mechanical properties of polymers. In this study, different amount of inorganic nanoclay (0.05wt.% - 1.0wt.%) was mixed with the modified epoxy resin (ATBN-m-DGEBA) to investigate the effect of filler loading on the physical and mechanical properties of composites.

Experimental

Materials

Diglycidyl ether bisphenol A (DGEBA) and aliphatic amine curing agent was purchased from Sigma Aldrich. Amine-terminated butadiene acrylonitrile (ATBN) is supplied by Scientific Polymer Products, Inc. The acrylonitrile content is 16%. This DGEBA resin was cured using aliphatic amine curing agent.

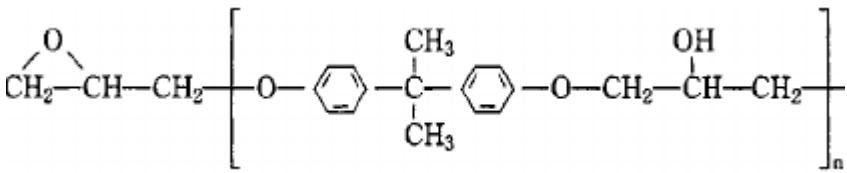

Figure 1: Structure of Diglycidyl ether bisphenol A (DGEBA)

The preparation of NC/ATBN-m-DGEBA Composites

ATBN was diluted in toluene and manually stirred for about 5 minutes to produce low viscosity of a homogeneous solution. The amount of toluene and ATBN used was determined by using the ratio 1: 2 respectively. Next, diluted ATBN was poured into DGEBA resin and mixed for 3 minutes. After that, NC filler was added into the modified resin (ATBN-m-DGEBA) and was continued stirred for another 3 minutes. Finally, the aliphatic amine curing agent was added and continually stirred for 5 minutes before pouring into the mold. The sample was cured at room temperature for 24 hours and further post-cured in the oven at 70^{O}C for 24 hours. The cured samples were cut according to the testing conducted.

In this study, six formulations were prepared to study the effect of nanoclay (NC) content on the physical and mechanical properties of ATBN-m-DGEBA resin properties as shown in Table 1.

Testing

Physical testing

Density is mass per unit volume of a material. It was conducted according to ASTM D792 and measured using a densitometer.

Hardness value of the composite samples was measured using the Rockwell machine by applying a specific load and then measure the depth of the resulting impression.

Table 1: Formulation of DGEBA/ATBN/NC Composites

Sample Designation	DGEBA: CA	ATBN (wt. %)	NC (wt. %)
F1	3.7: 1.3	5	0
F2	3.7: 1.3	5	0.05
F3	3.7: 1.3	5	0.3
F4	3.7: 1.3	5	0.5
F5	3.7: 1.3	5	0.7
F6	3.7: 1.3	5	1.0

Mechanical testing

Tensile test was performed in all six formulations of composite samples and five samples for each formulation were tested. The tensile test was conducted according to ASTM D 678 by using a tensile machine SHIMADZU AG-X Series at speed 5mm/min at room temperature. The sample dimension was 15cm x 2cm x 0.3cm (length x width x thickness). There were some data obtained from the tensile testing such as tensile strength, Young's Modulus and tensile strain.

Results and Discussion

Figure 2 shows the tensile strength of unfilled and filled nanoclay reinforced ATBN-m-Ep resin. The unfilled modified epoxy resin (Un-m-Ep) had a low tensile strength and it's value is approximately about 5.8MPa. Addition of 0.05wt% NC in the modified Ep (m-Ep) improved 34 times the tensile strength than the unfilled sample. It can be seen that there was no significant changes happen when 0.3wt% of NC loaded in the m-Ep. This condition can be interpreted by a good dispersion of NC particles which occurs at 0.05wt% - 0.3 wt% levels with repsects to the m-Ep resin causes an improvement in tensile strength. Better interaction seems to be effective for stress transferred between both NC and ATBN-m-Ep owing to the interfacial bonding effect of the flexible ATBN polymer chains resulting in an interdiffusion and

entanglement between the molecules of both ATBN chains and stiff 3-D networks of epoxy structure. However, up to 0.3wt% NC, the strength gradually decreases about 97%. Major reduction might be attributed due to the chances of forming aggregates at high loading of NC content. In other words, it can be said that the intercalation of NC in the modified matrix [1] gave a negative effect to the tensile properties.

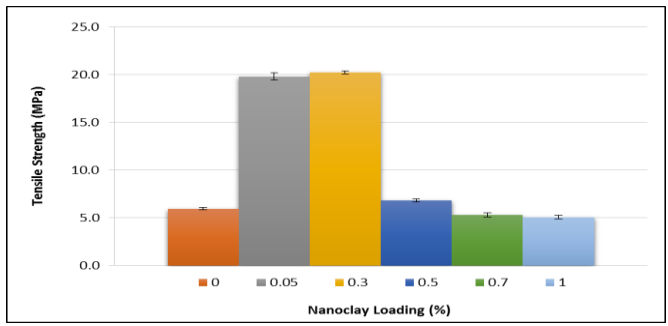

Figure 2: Effect of NC loading on Tensile Strength of composites

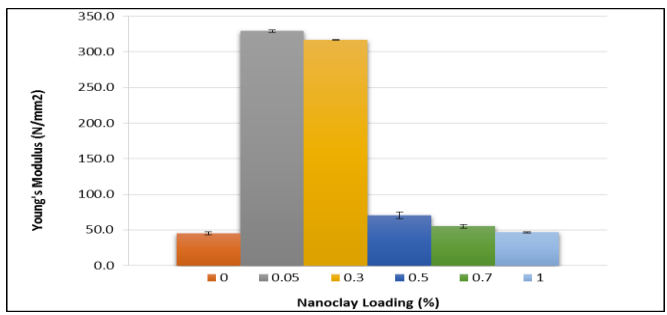

Figure 3. Young's Modulus of Unfilled and Filled NC reinforced ATBN-m-EP Composites

The effect of nanoclay content on the Young's Modulus is shown in Figure 3. The trend of Young's Modulus is same with the tensile strength condition. At 0.05wt% and 0.3wt% NC content displays the highest values as compared to other composite samples. It revealed that the incorporation of NC at that range would produce poor intercalation of NC particles causes the

modulus increases. A good dispersion of nanofiller throughout the m-Ep occurs at low filler loading due to strong chemical interaction between the metallic atoms (Silicon and Aluminum) of NC and oxygen and carbon atoms in the m-Ep resin [3] resulting a complete exfoliation which could contribute to the stiffening effect. Further NC loading (above 0.3 wt. %) led to the depression of the modulus. It can be stated that the reduction happens because the composites faced the agglomeration phenomenon where the agglomerated filler particles would produce micro-space between particles and thus reducing the modulus of composites.

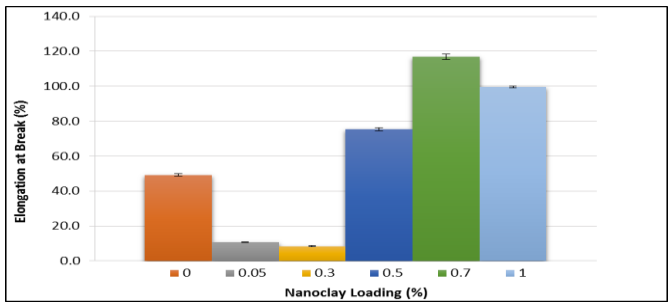

Figure 4: Effect of Nanoclay loading on the tensile strain of ATBN-m-Ep matrix

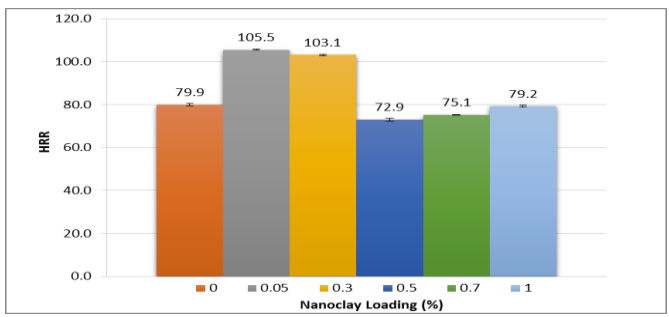

Figure 5: Effect of Nanoclay loading on the hardness of ATBN-m-Ep matrix

The effect of nanoclay (NC) content of the tensile strain response of the ATBN-m-Ep matrix is illustrated in Figure 4. As it can be seen that the trend of tensile strain is inversely proportional to the tensile strength. As mentioned earlier, a good dispersion happens at low filler loading (0.05 and 0.3wt.%) and thus, the composite structure became rigid so that it would

restrict the mobility of polymer chains. Once, the stress is applied to the composites, it easy to break. The tensile strain is related to the elasticity behavior and the inorganic fillers such as NC exhibits a lower elasticity and hence decreasing the tensile strain once it was incorporated in the modified epoxy resin. Further addition of filler would produce intercalated NC-modified epoxy resin and the increment of tensile strain could be due to the flexible effect of which dominated in the ATBN modified epoxy resin.

As shown in Figure 5, the hardness increases with the incorporation of NC filler from 0 to 0.3 wt. % in the exfoliated NC-modified epoxy composites. An exfoliated condition of NC throughout the modified matrix would form a rigid structure of uniformly-dispersed NC-m-Ep composites and thus, hardness increases. But further addition of NC (up to 0.3 wt. %) causes the hardness drastically decreased by 26.5% ± 4.

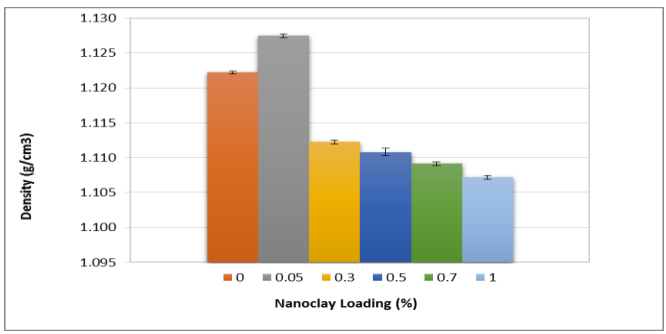

Figure 6: Effect of Nanoclay loading on the density of ATBN-m-Ep matrix

The density value of the NC reinforced modified epoxy composites is illustrated in Figure 6. A good interfacial bonding between NC particles and the m-Ep produces less porosity. As mentioned previously, the optimum amount of NC that shows a good dispersion condition (see Fig 7a.) in the m-Ep matrix was at 0.05 wt.% and thus, having higher density of sample as compared to other samples. In other words, it can be said that the NC filler not only acts as a reinforcing agent but at optimum amount of NC, NC particles plays as a pore-filling role to reduce the porosity [4] in the m-Ep resin and hence producing a denser polymer networks. However, the excessive amount of NC increased the porosity and hence decreased the density of nanocomposites. The formation of porosity could be due to the

filler-filler networks. Aggregated of filler particles in the m-Ep matrix is shown in Figure 7b.

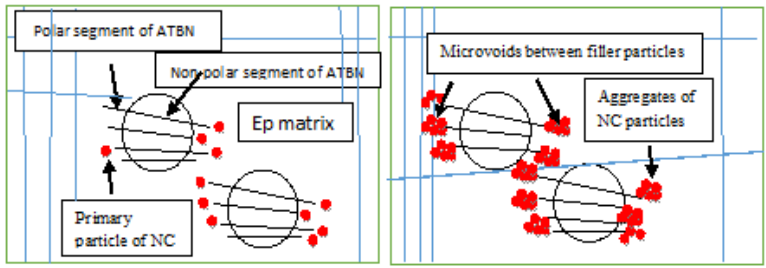

Figure 7: (a) At low filler loading (0.05 & 0.3 wt.%); b) High filler loading (0.5, 0.7,1.0wt. %)

Conclusion

In this work, the reinforcing effect of nanoclay on the modified epoxy resin (ATBN-m-DGEBA) resin was successfully carried by conducting the physical and mechanical testing. Addition at low NC loading improved the tensile strength, modulus, hardness and density properties due to good interaction bonding between filler and matrix interfaces and the exfoliated NC are throughout the m-Ep matrix resulting to the reinforcement of the composites.

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