Effect of Mixing Parameter on Epoxy-Organoclay Montmorillonite Composites: Flexural and Microscopy Analysis

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Abstract. The effect of two mixing parameters during preparation of epoxy composites on speed 250 rpm and 500 rpm, mixing time 60, 90, 120, 150 and 180 minutes were observed in this study by flexural and microscopy analysis. Preparation epoxy composites were based on epoxy diglycidyl ether of bisphenol A (DGEBA) and montmorillonite (OMMT) filler. It was found that mixing speed at 500 rpm performed higher flexural strength as compared to 250 rpm at mixing time 150 minutes. This was due to an improved mixing speed by appropriate time as observed through Scanning Electron Microscopy (SEM).

1. Introduction
Epoxy is well known matrix materials used commonly in composites development due to their ease processing. Method of preparation nanocomposites is very important in ways to produce a good nanocomposites. The processing of epoxy nanocomposites have varies techniques depends on types of applications. Some literature referred as synthesis nanocomposites. There are four general approaches to synthesized layered silicates/polymer nanocomposites i) Solution approach, ii) In-situ polymerization, iii) Melt intercalation and iv) Sol gel technology. However, numerous processing technique highlighted in-situ polymerization and it have been practiced by many authors in an attempt to obtain a homogeneous dispersion of nanofiller in the nanocomposite. In-situ polymerization by mechanical mixing are the most common technique that used to synthesized epoxy -clay nanocomposite [1]. It has been reported widely for advanced epoxy nanocomposites filled nanoclay such organo-montmorillonite. H.Alamri in his research epoxy nanocomposites used mechanical mixer for 10 minutes with rotation speed 1200 rpm [2]. A.Jumahat and W.Liu prepared their sample at same duration time 120 minutes at 400 rpm and 1000 rpm respectively [3-4]. While, I.Isik in his study epoxy montmorillonite composites used 120 minutes to achieved desired properties at no specific rotation speed [5]. However, less of researcher reported on the effect of parameter of mixing and time of mixing towards mechanical properties.
In this study, the effect of time and rotation speed of stirrer was considerably investigate at optimum montmorillonite content on epoxy diglycidyl ether of bisphenol A (DGEBA) [6].

**Experimental**

1.1. Material
Epoxy matrix resin, diglycidyl ether bisphenol-A (DGEBA) was sourced from Euro Pharma (M) Sdn Bhd. Organo modified montmorillonite (OMMT) used was surface modified contains 25-30 wt% octadecyldiamine is from Sigma Aldrich, and curing agent Isophoronediamine (IPDA) were sourced from Grechem Material Technology.

1.2. Sample Preparation
The mix ratio of epoxy and hardener amount was determined by stoichiometry. OMMT was dried in vacuum oven at temperature 60°C for 24 hours before mixing onto epoxy resin. Then epoxy and OMMT were mixed together for 60, 90, 120, 150, 180 minutes at mixing speed of 250 rpm and 500 rpm (Refer Table 1). Followed by adding curing agent IPDA and degassed for about fifteen minutes using the two stage vacuum pump before poured onto the silicon mold and cured at room temperature.

| Sample | Epoxy: IPDA | Filler | Speed (rpm) | Time (minutes) |
|--------|-------------|--------|-------------|----------------|
| A      | 100:23      | 3 phr  | 250 rpm     | 60, 90, 120, 150, 180 |
| B      | 100:23      |        | 500 m       |                |

1.3. Testing and Characterization
Three point flexural test were performed at Instron machine according to ASTM D790 with specimen dimensions 130×20×5 mm at crosshead speed of 1 mm/min and support span of 50 mm to study the flexural strength and modulus. Scanning electron microscope (JEOL Ltd., Japan) were tested at an accelerating voltage 10-20 kV to study the fracture surface of specimens. The specimen were coated with platinum coating.

2. Results and Discussions

2.1 Flexural Strength and Modulus
Effect of mixing times to mechanical stirring speed at 250 rpm and 500 rpm was investigated by three point bending testing. Figure 1 shows an ultimate flexural strength at different mixing time for 250 rpm and 500 rpm in bar graph and the data attained was displayed in table 1. It was observed at minimal mixing time 60 minutes the flexural strength for 250 rpm and 500 rpm does not show very significant differences where the value is about 7.0 MPa and 7.8 MPa respectively. However as mixing time increase to 90 minutes and 120 minutes the strength start to increase from 7.2 MPa to 8.2 MPa for rotation speed 250 rpm. The same situation applies for the rotation speed 500 rpm until the mixing time 150 minutes and reaching the highest bending strength at 13.7 MPa. It was observed that after 150 minutes the flexural strength was decreased to 9.0 MPa differs by 250 rpm that decrease after 120 minutes.

The bending response through modulus elasticity also shows the similar trend in which the maximum bending strength of 250 rpm and 500 rpm at 120 minutes and 150 minutes respectively has achieved highest modulus. It was observed a longer mixing time more than 150 minutes decrease the modulus elasticity.
Table 2. Values of flexural strength and modulus elasticity of different mixing time and speed for epoxy organo-montmorillonite.

| Rotation Speed | 250 rpm | 500 rpm | 250 rpm | 500 rpm |
|----------------|---------|---------|---------|---------|
| Mixing Time (minutes) | Flexural Strength (MPa) | Modulus Elasticity (MPa) | Flexural Strength (MPa) | Modulus Elasticity (MPa) |
| 60 | 7.0 | 7.8 | 1001 | 1020 |
| 90 | 7.2 | 8.2 | 1032 | 1065 |
| 120 | 8.2 | 10.0 | 1053 | 1060 |
| 150 | 7.0 | 13.7 | 1144 | 1549 |
| 180 | 5.8 | 9.0 | 600 | 895 |

Figure 1. Effect of mixing time on different speed content on flexural strength of epoxy organo-montmorillonite composites.

Overall, the highest bending strength and modulus achieved at mixing time of 150 minutes with mechanical stirring speed 500 rpm may be explained by the fact that dispersion of organo-montmorillonite in the epoxy matrix is better at high velocity within the range time. It is because the at 500 rpm it promotes a good radial flow for drawing the material to be mixed from the top and the bottom with high turbulence at high shearing forces to lead pre-intercalation process efficiently [7]. The enhancement in bending strength also may be ascribed by effectiveness mixing time and speed for better dispersion of organo-montmorillonite filler throughout the matrix that helps increasing of interaction matrix/filler interface. Meanwhile, the lower flexural strength of epoxy organo-montmorillonite in mechanical stirring 250 rpm were expected due to non-homogeneity within the matrix by low shearing forces in short time mixing and tend to agglomerate such degrade the interfacial adhesion between the matrix and filler as can be seen in fractographic Figure 2(a).

2.2 Fractograph Analysis

Image of the fracture surface effected by three point bending testing for the optimum flexural strength achieved by mechanical mixing speed 250 rpm and 500 rpm have been analysed using scanning electron microscope (SEM) and shown in Figure 2. Generally a rougher surface is seen upon adding organo-MMT into epoxy matrix [8–11]. The fracture surface of 250 rpm show considerably different fractographic features as shown in Figure 2 (a) and magnify image at in (b) compared to figure 2 (c) for 500 rpm at 150 minutes and magnify image in (d). As can been seen from figure 2 (d) lower speed 500 rpm, it promote much rougher than 250 rpm. According to Lin et al. (2006) more energy was required for a rougher surface to break which support the high mechanical properties obtained for epoxy organo-montmorillonite at mechanical mixing speed 500 rpm [12].
Figure 2. SEM micrograph of the fractured surface epoxy organo-montmorillonite at optimum flexural strength for 250 rpm and 500 rpm.

3. Conclusions
In this study, the influence of mixing speed and varies time from 60 to 180 minutes was studied by means of flexural properties and fractographic analysis. The samples was prepared using mechanical stirrer at speed 250 rpm and 500 rpm. It was observed that mixing speed at 500 rpm performed higher flexural strength as compared to 250 rpm at mixing time 150 minutes.

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References
[1] Zunjarrao S C et al 2006 J. Mater. Sci. 41 2219.
[2] Alamri H et al 2013 Comp. Part A Appl. Sci. Manufact. 44 23
[3] Jumahat A et al 2012 Pro. Eng. 41 1607.
[4] Liu W et al 2005 Comp. Sci. Tech. 65 307.
[5] Isik I et al 2003 Polymer 44 6371.
[6] Daud Y M et al 2016 AIP Conf. Proc. 1885 1.
[7] Daud Y M et al 2016 Key Eng. Mater. 673 55
[8] Castrillon P D et al 2015 Comp.Struc. 133 70.
[9] Subramanyan A K et al 2007 Comp. Part A Appl. Sci. Manufact. 38 34
[10] Zainuddin et al 2010 Mater. Sci. Eng. A 527 7920.
[11] Lin L Y et al 2006 Comp. Sci. Tech. 66 2116.
[12] Lin J et al 2006 Comp.Struc. 7 30.