The effect of clay and mixing conditions to fire resistant and mechanical property of unsaturated polyester – glass fiber composites by a vacuum bagging method

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Abstract. Glass fiber reinforced polymer (GFRP) composites have a weakness in fire resistant property. The addition of clay can improve the fire resistant property of GFRP. However, a good mixing is needed to disperse the clay. This research aims to observe the effect of clay and mixing conditions on fire resistance and impact strength of GFRP/clay composites. Unsaturated polyester (UP), glass fiber and 3-Aminopropyltriethoxysilane were used as a matrix, a reinforcement and a compatibilizer, respectively to build the composites. Unsaturated polyester-clay-silane were mixed using a magnetic stirrer with a variation of rotation speed and mixing duration. The composites were fabricated using a vacuum bagging method. The composition of clay was varied from 1, 2, and 3 wt%. The rotation speed and mixing time were varied at 100, 150, 200 rpm and 60, 90, and 120 minutes, respectively. UP-glass fiber was used as a comparison material. The results showed that composites with 1 wt% clay content was not burned with the burning rate value of 0 mm/min. There were no significant effects of variables on impact strength in between the boundaries that attributed to different mechanisms such as shear and diffusion that worked together to disperse the clay. The shear action should work at high speed rotation and short mixing time, while diffusion at low speed rotation and long mixing time.

1. Introduction
Glass Fiber Reinforced Polymer (GFRP) composites are widely used in many fields, such as aircraft, automotive and construction materials [1-3]. It was evidence that unsaturated polyester (UP) has been used as a matrix for GFRP composites, because the composites had good mechanical properties and UP had lower price compared to others thermoset polymers [4]. However, GFRP is not resistant to fire. From previous researches it turned out that glass fibers reinforced UP had poor fuel resistance properties [5]. Therefore, the addition of a filler as a flame retardant was necessary.

One filler that had been proved could improve GFR-UP fire resistant is clay [5]. Although, the hydrophilic property of the surface of clay caused poor interaction or interface bond with the matrix. Therefore, a compatibilizer, such as silane, was needed to facilitate clay dispersion in polymer [6]. Moreover, the addition of silane could improve the mechanical properties of GFR-UP [7].
The clay dispersion in polymer also depends on processing method [8]. Mixing process was one of commonly used method to disperse clay in polymer. Rotation speed and mixing time are process parameter in mixing process which could affect nanocomposite performance [9]. Magnetic stirrer was one of tools which have rotation speed and mixing time parameter. The rotation speed produces shear tension. The higher speed rotation could produce a higher shear tension which could facilitate clay dispersion. However, a high shear tension could make the composites degradation [10, 11]. Therefore, the varieties of process parameters were required to find the effect of mixing process on GFR-UP fire resistant and mechanical properties.

The specimen preparation can also affect GFR-UP property according to fiber and nanocomposite laminating. Vacuum bagging is one of laminating process which has the following advantages; has comprehensive pressure distribution, the amount of resin content that can be controlled, having customized mold, having low amount of void, and being not dependent on operator [12].

The objective of this research was study the effect of clay addition and mixing conditions on fire resistance and impact strength of GFR-UP/clay nanocomposites. The processing variables were clay content, rotation speed and mixing time.

2. Experimental method

2.1. Materials

Unsaturated polyester (UP) Yukalac 123 B - EX (IV) and E-glass fiber with chopped strand mat type (CSM) as first filler were supplied by PT Justus Kimia Raya. Montmorillonite (Cloisite 20A) or MMT C20A as second filler was provided by Southern Clay and silane (3-Aminopropyltriethoxysilane) as compatibilizer was produced by Sigma Aldrich.

2.2. Preparation of nanocomposites

Firstly, MMT C20A was sieved and dried at 80 °C for 24 hours. UP, clay, and silane were stirred manually for 4 minutes. The clay composition was varied as low, medium, and high (1, 2, and 3 wt%). The composition of silane was 0.5 wt%. Then, the solution was mixed by magnetic stirrer method. Methyl Ethyl Ketone Peroxide (MEKP) was also added, and mixed manually for ± 40 secs. The materials were processed at eleven conditions that are shown in Table 1.

| Sample | Clay (wt%) | Rotation speed (RPM) | Time (minute) |
|--------|------------|----------------------|---------------|
| Control | 0          | 0                    | 0             |
| 1      | 0          | 100                  | 60            |
| 2      | 1          | 100                  | 120           |
| 3      |            | 200                  | 60            |
| 4      |            | 200                  | 120           |
| 5      |            | 100                  | 60            |
| 6      |            | 100                  | 120           |
| 7      | 3          | 200                  | 60            |
| 8      |            | 200                  | 120           |

After mixing, specimens were prepared by vacuum bagging. At first, hand lay-up technique was applied, with nine layers CSM, then the composite was covered by peel ply, perforated film, breather and vacuum bag respectively. Vacuum method was applied with -1 bar pressure at room temperature for 3 hours. Then, the samples were cut according to test standard (UL 94H and Impact Charpy ISO 179).

3. Characterization
Flammability and impact strength test were conducted to analyze fire resistance and mechanical properties with ASTM D635 (UL 94H) and Impact Charpy ISO 179 standard. The flammability test was carried out 5 specimens using methane gas. Impact strength test was carried out to 5 specimens using the Ceast Resil Impactor Junior with speed of 3.7 m/s, pendulum energy 15 Joule, and correction energy 0.05 Joule. In this research, the composites of contained 1 wt% clay and 3 wt% clay were identified as “low” “high” compositions respectively.

4. Results and discussion

4.1. Effect of clay, mixing rotation speed, and time on fire resistant

In this research, it was found that UP with 1 and 3 wt% organoclay can improve the fire resistant up to 100%. Table 2 shows that the low clay composition samples at low speed and high speed, generate the similar burning rate which is 0 in result. In compared to previous study, polyamide 6 with 2 wt% organoclay could improve 2.4% in fire resistant property [13].

Table 2. Burning rate and impact strength GFRP / clay composites.

| Sample | Clay (wt%) | Rotation speed (RPM) | Time (minute) | Burning rate (mm/min) | Impact Strength (kJ/m²) |
|--------|------------|----------------------|---------------|-----------------------|------------------------|
| Control | 0          | 0                    | 0             | 8.5                   | 129 ± 12               |
| 1       | 1          | 100                  | 60            | 0                     | 121 ± 12               |
| 2       | 1          | 100                  | 120           | 0                     | 128 ± 12               |
| 3       | 2          | 200                  | 60            | 0                     | 153 ± 17               |
| 4       | 3          | 200                  | 120           | 0                     | 133 ± 10               |
| 5       | 3          | 100                  | 60            | 8                     | 143 ± 21               |
| 6       | 3          | 100                  | 120           | 0                     | 137 ± 13               |
| 7       | 3          | 200                  | 60            | 0                     | 123 ± 15               |
| 8       | 3          | 200                  | 120           | 6.7                   | 13711                  |

4.2. Effect of clay, mixing rotation speed, and time on fire resistant

Figures 1(a) and (b) show that the result of impact test value selection. Samples with “low” and “high” clay composition were selected to study the comparison. In low composition clay samples, the diffusion and shear mechanism worked well. Meanwhile, in high composition clay samples, the diffusion and shear mechanism did not work well because probably there was an agglomeration by visual looking and it decreased the impact value. In the previous research diffusion and shear worked well together [11]. The difference value between in low and high composition clay samples was not substantial. It was proven that diffusion and shear mechanism had no significant effect in impact value.

Figure 1. Impact strength GFRP/clay composites (a) low level of clay, (b) high level of clay.
There are two variables which affect the process of polymer mixing, namely diffusion and shear. Shear mechanism would work at the high-speed rotation and short time. In the other hand, diffusion mechanism would work at the low speed rotation and longer time [10, 11]. In this research shear and diffusion did not show positive result.

5. Conclusion
It could be concluded that additional clay composition with percentage of 1wt% improved the fire resistant property by producing unburned GFR-UP composites. With the burning rate value of 0 mm/min. In the other hand, the shear and diffusion mechanism did not affect significantly the impact strength values for both composites 1 wt% and 3 wt% clay compositions.

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