Sawdust Flour Reinforced Epoxidized Natural Rubber: Effect Of Trans-Polyoctylene Rubber on Curing Characteristics and Tensile Properties

Omar S Dahham¹, N Z Noriman¹, R Hamzah¹, S Z Syed Idrus², Z Shayfull³,⁴, T Adam¹,⁵, H Jaya¹

¹ Center of Excellence Geopolymer and Green Technology (CEGeoGTech), Faculty of Engineering Technology (FETech), Universiti Malaysia Perlis (UniMAP), Level 1 Block S2, UniCITI Alam Campus, Sungai Chucuh, Padang Besar, 02100, Perlis, Malaysia.
² School of Human Development and Techno-communication (iKOM), Green Advanced Computing and Technology (GREAT) Research Group – CEGeoGTech, Universiti Malaysia Perlis (UniMAP)
³ School of Manufacturing Engineering, Universiti Malaysia Perlis, Arau, 02600, Malaysia
⁴ Green Design and Manufacture Research Group, Center of Excellence Geopolymer and Green Technology (CEGeoGTech), Universiti Malaysia Perlis, Kangar, 0100, Malaysia
⁵ Institute of Nano Electronic Engineering (INEE), Universiti Malaysia Perlis (UniMAP), 01000 Kangar, Perlis, Malaysia

E-mail: niknoriman@unimap.edu.my

Abstract. In this work, the compatibilization effect of trans-polyoctylene rubber (TOR) on the cure characteristics, tensile properties and morphology of epoxidized natural rubber/sawdust flour (ENR-25/SD) compounds was evaluated. Five different TOR content (2, 4, 6, 8 and 10 phr) was used. Results showed that the increasing of TOR content caused decreasing on scorch and cure time. Moreover, the maximum torque has improved. The minimum torque showed decreasing as TOR content increased indicating that the rubber compound became less viscous and easy to process. The addition of TOR, particularly high TOR content (10 phr) into rubber compounds has increased the interfacial adhesion between SD and ENR-25, which in turn improved the tensile strength and modulus of the rubber compounds. The elongation at break has decreased as TOR content increased indicating that the samples became less flexible. SEM micrographs proved that the high TOR content presented stronger SD/ENR-25 than low TOR content.

1. Introduction
Nowadays, wood polymer Composites is used extensively as a raw material in building such as fences, railings, windows, doorframes, park benches and other. This type of composite gives numerous advantages such as low cost, dimensional stability, good mechanical properties, long service life and less deterioration compared to natural wood itself [1,2]. The sawdust of wood is often used as filler in the production of wood polymer Composites due to the environmental friendly, bio-degradability...
characteristics and low cost. Furthermore, sawdust fibers are less abrasive to processing machine and
abundantly available in the nature [3]. In the past few years, several studies were conducted on the
natural fibers using as competently as possible to produce high quality fiber-reinforced polymer
(thermoplastic, thermoset and rubber) composites for a wide range of applications [4-8]. Natural fibers
are currently used as an alternative material to conventional fibers, such as carbon, glass, aramid and
other due to advantages of natural fiber compared to conventional fibers as mention above.
Nonetheless, the declining of compatibility of polymer composites as natural fiber added is the major
constraint in the polymer industry.
Compatibilization is defined as a process that uses to reduce the high interfacial tension between
the thermoplastic and elastomeric phases of the polymer components [9]. Based on researches that
found in the literature, the compatibilization effects can increase the interfacial adhesion, which in turn
lead to improve the mechanical properties of the polymer components. This process could be achieved
either by adding a new component, called a compatibilizer, or by improving the polymer components
interaction mechanically or chemically. One of the most effective techniques that used widely for
compatibilization is adding a third polymer into the polymer components to be partially miscible in
early stage [10].
Trans-polyoctylene rubber (TOR) was used widely as a compatibilizer in incompatible polymer
blends, particularly the heterogeneous rubber blends. TOR introduces a good processability in the
rubber processing with a temperature range (100-150 °C). Furthermore, TOR can present a good
collapse resistance below the melting temperature because of the recrystallization [11]. Several
researchers studied the compatibilization effect of TOR on the properties of polymeric materials such
as rubber blends. Bizi et al. have studied the TOR effects on the properties of NR/SBR blends. Results
showed that the physical properties of the rubber blends have clearly improved [10]. Our previous
researches scrutinized the TOR influence on the Properties different rubber compounds. The results of
these researches proved that most of the rubber blends properties were improved as TOR added [8].
The aim of the current research is to evaluate the impact of different TOR content as a
compatibilizer on the cure characteristics, tensile properties and morphology of the epoxidized natural
rubber / sawdust flour compounds

2. Experimental

2.1. Materials
Epoxidized natural rubber with 25mol% epoxidation (ENR-25) was obtained from Rubber Research
Institute of Malaysia (RRIM). Whereas other materials such as zinc oxide, stearic acid, Trans-
Polyoctylene Rubber sulfur and N-cyclohexyl-2-benzothiazole sulfonamide (CBS) were obtained from
Anchor Chemical Co. (M) Ltd. Sawdust (SD) was collected from Perlis Sawmill Sdn. Bhd. The
formulation of the rubber compounds of this study is shown in Table 1.

| Table 1. The formulation of ENR-25/SD/TOR compounds at different TOR content. |
|---------------------------------|---|---|---|---|---|---|
| Ingredients                     | Phr |
| ENR-25                          | 100 | 100 | 100 | 100 | 100 | 100 |
| Sulfur                          | 2   | 2   | 2   | 2   | 2   | 2   |
| CBS                             | 1   | 1   | 1   | 1   | 1   | 1   |
| Stearic Acid                    | 2   | 2   | 2   | 2   | 2   | 2   |
| ZnO                             | 5   | 5   | 5   | 5   | 5   | 5   |
| SD                              | 5   | 5   | 5   | 5   | 5   | 5   |
| TOR                             | 0   | 2   | 4   | 6   | 8   | 10  |
2.2. Sample Preparation and analysis

After Sawdust cleaning and drying Crusher model RT34 (Chyun Industrial Co. Ltd.) was utilized for sawdust grinding. Lab sieves were used to obtain SD flour at 100 - 200 μm size range. ENR-25 was mixed together with the other ingredients using two-roll mill X (S K - 160 X 320) based on D 3184-89 and then rubber compounds were put inside freezer for 48 hours to avoid further crosslink. After freezing, hot press machine was used to cure and mold the rubber compounds with 160°C temperature and under 30 tonne pressure. The cure characteristics test was carried out using Rheometer (MDR 2000) according to ASTM D 2240-93. A dumbbell-shaped mold was used to obtain tensile test samples. According to ASTM D412, tensile test was conducted using universal testing machine (Instron 5582). The tensile fracture surface of samples that obtained from tensile test was analyzed using scanning electron microscopy (SEM) model JEOL JFC6460LA. Before SEM analysis, palladium layer (1.5 - 3 nm) was coated samples using sputter coater machine to increase the image resolution and also to avoid the electrostatic charging during samples analyzing.

3. Results and discussion

3.1. Cure characteristics

The results of scorch time $t_2$, cure time $t_{90}$, minimum torque $M_L$ and maximum torque $M_H$ of the rubber compounds with different TOR content are shown in Table 2 respectively. It can be seen that the value of $t_2$ and $t_{90}$ become shorter as TOR content increased in the rubber compounds. This could be attributed to the nature of TOR that acts as a compatibilizer in the rubber compounds. Additionally, TOR also could behave as an unsaturated rubber, which in turn lead to increasing of bonding between TOR itself and elastomer in short time. The increasing of TOR content decreased the viscosity and improved the processability of the rubber compounds due to the plasticizer effect of TOR [12,13]. Therefore, $M_L$ decreased. By contrast, the value of $M_H$ presented an opposite direction, which increased as TOR content increased in the rubber compounds. The increasing of TOR content has increased the crosslinks between the elastomer and TOR. Thus, $M_H$ value improved.

| TOR | $t_2$ (min) | $t_{90}$ (min) | $M_L$ (dNm) | $M_H$ (dNm) |
|-----|-------------|----------------|-------------|-------------|
| R0  | 1.81        | 9.22           | 8.30        | 22.5        |
| R02 | 1.23        | 7.71           | 6.10        | 26.15       |
| R04 | 1.15        | 6.89           | 5.90        | 28.00       |
| R06 | 0.99        | 6.02           | 5.12        | 28.95       |
| R08 | 0.84        | 5.54           | 4.55        | 31.40       |
| R10 | 0.57        | 4.21           | 3.88        | 35.10       |

3.2. Tensile properties

The results of tensile properties, such as tensile strength (Ts), modulus (M100) and elongation at break (Eb) of the rubber compounds at different TOR content are listed in Table 3 respectively. It’s obviously seen that the Ts values have increased linearly with the increasing of TOR content in the rubber compounds. The addition of TOR, particularly high TOR content has increased the dispersion of SD in the rubber matrix, which in turn improved the interfacial adhesion of the rubber compounds [14]. M100 values shown same direction of Ts value. The addition of TOR, particularly high TOR content into the rubber compounds has increased the incorporation between SD and ENR-25 matrix, which in turn gave a positive impact on the modulus value. By contrast, Eb shows decline as TOR content increased in the rubber compounds. The addition of TOR, particularly high TOR content hindered the flow and mobility, which means the flexibility of the rubber compounds has reduced.

| TOR | Ts (MPa) | M100 (dN/m) | Eb (%) |
|-----|----------|-------------|--------|
| R0  | 12.3     | 5.12        | 50.2   |
| R02 | 13.4     | 5.23        | 49.1   |
| R04 | 14.5     | 5.34        | 48.2   |
| R06 | 15.6     | 5.45        | 47.3   |
| R08 | 16.7     | 5.56        | 46.4   |
| R10 | 17.8     | 5.67        | 45.5   |
3.3. Morphology

The SEM micrographs of tensile fracture surface of ENR-25/SD/TOR at 2 phr and 10 phr TOR content are shown in Figure 1 (a) and (b) respectively. It’s clearly seen both two samples, low (2 phr) and high (10 phr) TOR content shows several tearing line on the tensile fracture surface of the samples indicating that the samples needed more energy to break the matrix [15,16]. The high TOR content sample shows strong SD bonding in SD ENR-25 due to the TOR effect that increased compatibility between SD in rubber matrix. However, the low TOR content shows less SD bonding in ENR-25 matrix due to the relatively poor compatibility between SD and ENR-25 compared to high TOR content, which caused breakages and pull out of the SD and few holes occurred.

![SEM micrograph](image_url)

(a)

Table 3. The variation of tensile properties of ENR-25/SD/TOR compounds.

| TOR  | Ts (MPa) | Eb (%) | M100 (MPa) |
|------|----------|--------|------------|
| R0   | 12.4     | 0.55   | 1245       |
| R02  | 14.12    | 0.75   | 1012       |
| R04  | 14.90    | 0.85   | 989        |
| R06  | 15.25    | 0.90   | 901        |
| R08  | 16.30    | 1.00   | 814        |
| R10  | 18.95    | 1.25   | 694        |
4. Conclusion
Due to the nature of TOR that can act as an unsaturated rubber and also as a compatibilizer in the rubber compounds, the curing characteristics have improved as TOR added into rubber compounds. Furthermore, the addition of TOR, particularly high TOR content has increased the dispersion of SD in the rubber matrix, which in turn improved the interfacial adhesion of the rubber compounds. Therefore, the tensile properties have improved. This matter was proven in SEM section.

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