The Effects of Different Rice Husk Loading and Size on The Properties of Standard Malaysian Rubber/ Rice Husk Composites

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Abstract. This research is focused on the rice husk (RH) that acts as natural filler in Standard Malaysian Rubber (SMR L). Two ranges of RH size were prepared, coarser size at 10-20 mm and fine size at 150-300 μm. While the RH loading was (10, 20, 30, 40 and 50 Phr). The effects of RH on the tensile properties, physical properties and cure characteristics of the compounds were examined. The results shown that the increasing of rice husk loading in SMR L/RH composites led to decrease in the cure (t 90) and scorch time (t 2) and increase in the minimum torque (ML) and maximum torque (MH). Despite the values of tensile strength decreased as RH content increase, the physical properties, such as hardness and crosslink density of the composites, particularly the composite with fine size filler were improved due to good filler-rubber interaction than coarser size filler.

1. Introduction
In general, natural fillers are produced from agriculture technology as it’s claimed as natural resources. It is subdivided from several factors such as origins; plants, animals or mineral. Stem, leaf, seed, wood and grass are examples of fillers from plant. These kinds of fillers contain cellulose and lignin. The filler structure contents are as they are composed of cellulose, hemicelluloses, waxes, lignin and water-soluble compounds [1]. The natural fillers do have differences in terms of properties as its properties are based on their own species. The exhibit properties are within a species that highly depending on area of growth, climate and age of the plant. Besides, it also varies depending on their
processing method used to breakdown to the fiber level. For other resource, which is mineral filler, it is naturally occurring fiber or slightly modified fiber procured from mineral.

There are some of natural filler’s advantages benefited compare to synthetic filler, such as high stiffness, biodegradable, low density, recyclable, renewable raw materials and their relatively low cost [2]. Moreover, natural fillers give less health problems for the people who work with the fillers. Natural filler also do no cause skin irritations and not suspected of causing lung cancer. Nevertheless, instead of advantages, it also has some disadvantages, such as high moisture sensitivity and variability of diameter and length. We need to dry it up to release the moisture as it will not definitely affect our natural fillers properties [3]. Rice husk is the largest waste produced from agricultural processing of grains. It has known as significant source of silica and to reduce the quantity of this squander material. RH can be burned in the open air. As the researcher begun to identify the benefits of using rice husk, it always been used in rubber properties research as a natural filler [4]. Santiagoo, R. et al. studied the properties of polypropylene/recycled acrylonitrile butadiene rubber/rice husk powder [5]. They found that the processing torque and tensile modulus increased with increasing rice husk content for all composites, which were attributed to the brittle nature of rice husk filler.

Our preliminary research investigated the effects of different size and content of rice husk on the tensile properties, physical properties and cure characteristics of Epoxidized Natural Rubber/ rice husk composites [6]. It is found that the addition of rice husk into rubber has improved the cure characteristics of the compounds by reducing the cure and scorch time. Other advantages, the physical properties (hardness and crosslink density) were improved as rice husk added. However, the value of elongation at break and tensile strength showed decline as rice husk added.

In our current work, we have investigated the influence of RH on the tensile properties, physical properties and cure characteristics of SMR L/ RH compounds at different RH loading and size.

2. Materials and methods

2.1. Materials and preparation of samples

Rice husks (RH) have been obtained from Bernas Malaysia Sdn. Bhd., Simpang Empat, Perlis. It was dried under sunlight exposure for 72 hours. Dried rice husk were ground and sieved by using Crusher model RT34 (Chyun Industrial Co. Ltd.) and lab sieves to obtain rice husk at 10-20 mm and 150- 300 μm respectively. Standard Malaysian Rubber (SMR L) was used as a raw material in this work, which is obtained from Rubber Research Institute of Malaysia (RRIM). While other chemicals, such as sulfur stearic acid, zinc oxide and N-cyclohexyl-2-benzothiazole sulfonamide (CBS) were purchased from Anchor-Chemical Co. (M) Ltd. All materials were prepared according to formulation as shown in Table 1. Based on ASTM D 3184-89, two-roll mill X-(S)-K/160-X-320 was used for ingredients mixing. Rheometer, model MDR-2000 was utilized for cure characteristics test based on ASTM D 2240-93. Hot-press machine was used to compression accordingly to respective cure time at 150 °C and under 30 tonne pressure.

2.2. Measurement of properties

Tensile test was done based on ASTM D412 using Universal testing machine, model Instron-5582. Hardness test was carried out using Durometer device, model Shore A. Toluene solvent has been used for swelling test purpose based on ASTM D3616. The data of swelling test were then used for crosslinking density calculation of the composites using Flory Rehner equations [7].

| Components       | Phr | R0  | R10 | R20 | R30 | R40 | R50 |
|------------------|-----|-----|-----|-----|-----|-----|-----|
| SMRL             | 100 | 100 | 100 | 100 | 100 | 100 | 100 |
| Sulfur           | 2   | 2   | 2   | 2   | 2   | 2   | 2   |
| Stearic Acid     | 2   | 2   | 2   | 2   | 2   | 2   | 2   |

Table 1. The ingredients of SMRL/RH composites.
3. Characterization

Figure 1 displays the morphology of raw RH with two different magnifications. Figure 1 (a) proved that the external surface of raw RH was rough and showing several aligned lumps. Meanwhile, Figure 1 (b) displayed the protuberance outer epidermis and silica. It also found that particles of RH were highly agglomerate, which could effect on some properties of the rubber compounds negatively [8].

![SEM image of raw RH at low (X100) and high (X2000) magnification.](image-url)

|       | 1  | 1  | 1  | 1  | 1  | 1  |
|-------|----|----|----|----|----|----|
| CBS   |    |    |    |    |    |    |
| ZnO   | 5  | 5  | 5  | 5  | 5  | 5  |
| *RH (FC &CS) | 0 | 10 | 20 | 30 | 40 | 50 |

4. Results and discussion

4.1. Cure characteristics

Table 2 shows scorch time $t_2$, cure time $t_{90}$, minimum torque $M_L$ and maximum torque $M_H$ respectively. It clearly seen that $t_2$ and $t_{90}$ (Figure 2) decreased as RH content increase. This was accredited with the presence of more crosslinking network in SMRL which was dominant matrix in
compound. As for fine RH loading incorporated, the large surface area of RH filler helps to interact more with rubber matrix and consequently reduce \( t_2 \) and \( t_{90} \) [9-11]. The increasing of rice husk loading in rubber compound reduced the macromolecular mobility in rubber compound, which in turn caused increase in the viscosity. Furthermore, the processability became more difficult. Thus, \( M_L \) increased. Fine RH filler has better processability since the smaller particle contributes to larger surface area and better interaction with matrix. \( M_H \) shows same trend of \( M_L \). The increasing of \( M_H \) as RH content increase contributed to crosslinking increasing. Fine RH filler was higher \( M_H \) than coarser size due to the bigger surface area of fine RH filler that contact with rubber matrix [12].

| RH | \( t_2 \) (min) | \( t_{90} \) (min) | \( M_L \) (dNm) | \( M_H \) (dNm) |
|----|----------------|------------------|----------------|----------------|
| R0 | Control        | 3.49             | 6.34           | 3.20           | 12.34          |
| R10| FS             | 2.28             | 6.25           | 5.56           | 22.28          |
|    | CS             | 2.41             | 6.75           | 6.00           | 36.16          |
| R20| FS             | 2.15             | 6.10           | 7.99           | 23.00          |
|    | CS             | 2.31             | 6.35           | 11.29          | 36.94          |
| R30| FS             | 0.59             | 5.24           | 11.04          | 25.62          |
|    | CS             | 0.67             | 5.79           | 12.41          | 37.41          |
| R40| FS             | 0.39             | 5.15           | 12.82          | 26.00          |
|    | CS             | 0.43             | 5.65           | 13.50          | 38.44          |
| R50| FS             | 0.28             | 5.06           | 13.44          | 40.21          |
|    | CS             | 0.32             | 5.45           | 13.99          | 28.8           |

**Figure 2.** The cure time (\( t_{90} \)) of SMR L/RH compounds

4.2. **Physical and tensile properties**

The values of physical properties (hardness and crosslink density) and tensile properties (tensile strength \( T_s \), elongation at break \( E_b \), and modulus at 100% elongation \( M_{100} \)) are listed in Table 3 respectively. It can be seen clearly \( T_s \) values decreased when RH increase due to the increasing of agglomeration of RH inside the matrix. Moreover, the miss-orientation of RH increased the difficulty of stress transmission from SMR L to RH. The inability of the coarse RH to support stress transferred from SMR L caused highly decreasing in \( T_s \) as compared with RH fine size [13,14].

\( E_b \) also decreases with increasing RH loading as expected. According to Premalal et al., as RH filler content is increases, a restriction to molecular motion of the macromolecules increase [15].
M100 is shown increasingly in trend and coarser RH has higher M100 than fine RH at same loading. Theoretically, the rigid filler in the rubber compound will significantly enhance the elastic stiffness and increase the modulus. As expected correlate with crosslinking density and hardness values, fine RH shows higher value than coarse RH at same loading. This was attributed to the better fine RH incorporation into SMR L matrix compared to coarse RH which in turn led to enhance the stiffness and rigidity of the rubber compound [16,17].

Table 3. The physical and tensile properties of SMR L/RH composites.

| RH | Ts (MPa) | Eb (%) | M100 (MPa) | Crosslink density X 10^-4 (Mole/cm^3) | Hardness (Shore A) |
|----|----------|--------|------------|--------------------------------------|-------------------|
| R0 | Control  | 18.24  | 1450       | 0.45                                 | 0.75              | 35.64             |
| R10 FS | 14.98  | 1305   | 0.50       | 0.82                                 | 41.75             |
| CS | 11.90    | 1179   | 0.60       | 0.76                                 | 39.5              |
| R20 FS | 12.65  | 1249   | 0.80       | 0.93                                 | 46.68             |
| CS | 8.12     | 1118   | 0.10       | 0.86                                 | 42.11             |
| R30 FS | 10.89  | 1098   | 1.00       | 0.97                                 | 50.93             |
| CS | 7.11     | 993    | 1.20       | 0.90                                 | 49.21             |
| R40 FS | 8.00   | 955    | 1.50       | 1.10                                 | 52.05             |
| CS | 5.20     | 931    | 1.90       | 0.94                                 | 51.49             |
| R50 FS | 6.10   | 853    | 1.90       | 1.36                                 | 53.58             |
| CS | 4.14     | 845    | 2.40       | 1.03                                 | 53.19             |

5. Summary
The increasing of rice husk loading led to reduce the cure and scorch time and increased the torque of the rubber compounds. The addition of RH improved the modulus, hardness and crosslink density of the rubber compounds. Elongation at break and tensile strength tend to decrease as rice husk content increased. Nevertheless, fine rice husk showed better properties as compared with coarse rice husk at all loading.

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