Curing characteristics tensile and physical properties of rice straw filled standard Malaysian rubber

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Abstract. In this research, the effects of Rice Straw (RS) reinforced Standard Malaysian Rubber (SMRL) on curing characteristics, tensile properties and physical properties were investigated. All compounds were prepared using two roll mill at five different RS loading (10, 20, 30, 40, 50 phr). In addition, two different size of RS, fine size (FS) at 300 μm and coarse size (CS) at 10 mm were used. The properties such as cure characteristics, tensile properties and physical properties were determined. Results indicated that the fine size of RS filled SMRL contributed to the better properties such as tensile, hardness and crosslink density compare to coarser size of RS filled SMRL at same loading.

1. Introduction

Recently, the compounding of several materials with the crude rubber is commonly used in industries particularly in tires manufacturing and many studies on their processing, morphology and properties were conducted [1]. One of the most important materials that could added to the rubber compound are fillers, such as carbon black, which had been prove to be worthy filler as it gives higher tensile strength [2]. Despite rubber, particularly natural rubber (NR) have notable properties, such as high tensile strength, high flexibility and elasticity, poor conductivity, high abrasion resistance reinforcement, fillers are needed in rubber compounding to improve the properties of the elastomer itself. Paddy field has become one of Malaysia’s accommodations. Thus, the wastage of the rice straw from paddy may potentially become the next natural reinforcement filler. Researchers and scientists still attracted in making rice straw their main attention to study. Liu et al has studied on the flexural properties of rice straw and starch [3]. They stated that composite from straw had better interface and higher flexural properties. During harvest, the moisture content of straw is usually exceeded 60%. However, in dry weather straw can quickly dry to its equilibrium moisture content, which is around 10-12%. Thus, it is low in density and lightweight. In addition, rice straw has low protein but high ash content. The main sources of carbohydrate in rice straw are lignin, hemicellulose and cellulose. Silica
content that is high in rice straw does give effect on the economic [4]. In other research, rice straw has been reinforced with high-density polyethylene composite to find the effect of coupled compatibilizing and toughening treatment. This is due to the elastomer that alone fails to effectively improve the strength and modulus of final composites [5]. The addition of natural filler such as rice straw may reduce the cost of rubber manufacturing. In term of properties, rice straw may give good properties towards rubber compounding as good as synthetic filler used.

Our earliest work has studied the influences of different size and content of rice husk on the cure characteristics, tensile and physical properties of Epoxidized Natural Rubber/ rice husk Compounds [6]. Results showed that the addition of rice husk into rubber has enhanced the cure characteristics of the compounds by reducing the scorch and cure time. Furthermore, the physical properties (crosslinking density and hardness) were enhanced with rice husk addition. Nevertheless, the tensile strength and elongation at break showed deterioration with rice husk increasing.

In our current research, we have studied the effects of RS as a natural fiber on the cure characteristics, tensile and physical properties of SMR L/RS compounds at different RS size and content

2. Materials and methods

2.1. Materials

Standard Malaysian Rubber (SMR L) was used in this research as a raw material, which supplied by Rubber Research Institute of Malaysia (RRIM). While other chemicals, such as sulfur, zinc oxide, stearic acid and N-cyclohexyl-2-benzothiazole sulfonamide (CBS) was supplied by Anchor Chemical Co. (M) Ltd. Rice Straw was taken at Paddy field at Wang Ulu - Kangar - Perlis. Two different size of rice straw (300 μm and 10 mm) were prepared by using Crusher model RT34 (Chyun Industrial Co. Ltd.).

2.2. Preparation and testing of SMRL/RS compounds

The formulation of SMRL/RS compounds is shown in Table 1. Mixing and pre-blending of rubber with chemicals and additives was done using two-roll mill X (S) K - 160 X 320 based on ASTM D 3184-89 at room temperature. Cure characteristics were studied using Monsanto Moving Die Rheometer (MDR 2000) according to ASTM D 2240-93 at 160 C with about 4 g samples of the respective compounds. Electrically hot press machine was utilized to compression accordingly to respective cure time at 160 C and under a pressure of 30 tonne.

Hardness test was carried out for vulcanized radial shape samples that have 6 mm thickness using Shore-A Durometer (Asker, Kobunshi Keiki Co. Ltd). The tensile test was done using dumbbell-shaped mold cutting from the vulcanized rubber sheets of 2 mm thickness. 5 samples were tested for each rubber compound loading. Universal testing machine (Instron 5582) was used for tensile test according to ASTM D412 at room temperature. ASTM D3616 was used for swelling density. Samples were weighted before immerse in a glass containing 30 ml of toluene and kept in dark place, then weight again after 48 hours. The calculations that used for swelling data were Flory-Rehner equations to calculate the crosslink density of rubber compounds [7].

| Ingredients       | Phr |
|-------------------|-----|
| SMRL              | 100 | 100 | 100 | 100 | 100 | 100 |
| ZnO               | 5   | 5   | 5   | 5   | 5   | 5   |
| Stearic Acid      | 2   | 2   | 2   | 2   | 2   | 2   |
| CBS               | 1   | 1   | 1   | 1   | 1   | 1   |
| Sulfur            | 2   | 2   | 2   | 2   | 2   | 2   |
| *RS (FC &CS)      | 0   | 10  | 20  | 30  | 40  | 50  |

Table 1. The formulation of SMRL/RS compounds.
3. Characterization

Figure 1 shows the distribution of raw rice straw morphology in SEM. Low magnification displays that the RS surface is rough enough to entrapped rubber molecule during compounding. Moreover, it can be observed that there are some spaces between rice straw which indicates less compact distribution in RS molecules. This might result in uneven distribution of rice straw in rubber compounding. Thus, it might reduce some result such as tensile strength. From 2000x magnification, it is clearly observe that lignin was surrounded the rice straw particle as it shows glow at the end of the structure.

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

4. Results and discussion

4.1. Cure characteristics

The effect of rice Rice Straw (RS) loading and sizes on scorch time (t2), cure time (t90), minimum torque (Ml) and maximum torque (Mh) are shown in Table 1. Results indicate that t2 continuously decrease with increase in RS phr loading on SMRL vulcanizates. The decrease on t2 was due to the restriction of the mobility and deformability of the matrix with mechanical restrain indicating that SMRL vulcanize took less time to reach the onset time of vulcanization as rice straw loading increase. However, the RS coarse size was higher t90 than fine size at same loading, which means the tendency of rice straw filler with coarser size requires longer time to coalesce and form agglomerates between each other to distribute in the SMRL rubber. t90 was faster when RS loading increase in the compounding.

At same loading, the coarse size of RS need longer time to cure compare to finer size of rice straw loading. This is due to the poor interaction between the rubber matrix and bigger particle size of RS that relate poor dispersion during the compounding. ML, increased with increasing of RS loading due to increase in viscosity. This indicates that the processability of the blends becomes more difficult [8]. Ml of RS coarser size was higher than fine size. This might due to the coarser size gives viscosity and rigidity to the rubber vulcanized. Mh shows same trend of Ml. The increment of Ml relates to the increasing of stiffness of the rubber compound. This might due to the restriction of the chain mobility when RS added. However, the fine size of RS shows lower Mh than coarser size at same loading. This was attributed to better dispersion of RS finer size, which made better compounding process [9].
Table 2. The cure characteristic of SMR L/RS compounds.

| RS  | t2 (min) | t90 (min) | ML (dNm) | MH (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     |

4.2. Tensile and physical properties

The values of tensile strength (Ts), elongation at break (Eb), modulus at 100% elongation (M100), crosslink density and hardness of the compounds are listed in Table 3 respectively. It can be seen obviously that Ts decreased gradually with increasing RS loading, which might due to incompatibility between fiber and matrix [10,11]. Nevertheless, Ts for fine size was higher than coarser size at same loading. It is believe that the finer size is well distributed in the rubber compounding.

Eb decreased with increasing RS loading as well causing decrease the resilience and toughness, which in turn led to reduce the mobility of the chain inside rubber compound. Therefore, the compound became stiffer and harder. At same RS loading, fine size shows lower Eb than coarser size due to lower viscosity of rubber vulcanized and easily to be fractured. M100 values increased gradually as RS content increased. According to Sharifah, the increment of M100 values is directly proportional to the crosslink density [12].

The increases in M100 values were due to improvement in crosslinking density. These results indicate the fact that the incorporation of RS into the rubber matrix enhanced the stiffness of the composites [13,14]. Moreover, RS fine size was higher M100 than coarser size. This contributed to the high RS distribution that gives larger surface area to come in contact with rubber matrix. The values of hardness values also increased constantly. Theoretically, both M100 and hardness values increases as filler loading increase. These results indicate the fact that the incorporation of RS into the rubber matrix improved the stiffness and brittleness of the rubber compounds.

Figure 2 shows the swelling percentage of the rubber compounds at different RS loading and size. It can be clearly seen the coarse size of RS, showed higher swelling percentage as compared with fine size of RS. This was might be due to the tendency of RS coarse size to agglomerate, which in turn led to increase the voids of the rubber compounds causing toluene penetration. Therefore, swelling percentage increased. Though, the high RS loading, particularly (50 phr) reduced the voids of the rubber compounds, which led to reduce the toluene penetration through rubber compounds [13].
### Table 3. The tensile and physical properties of SMR L/RS compounds.

| RS | Ts (MPa) | Eb (%) | M100 (MPa) | Crosslink density X 10^-4 (Mole/cm^3) | Hardness (Shore A) |
|----|----------|--------|------------|--------------------------------------|-------------------|
| R0 | Control  | 18.4   | 1450       | 0.45                                 | 0.70              |
| R10| FS       | 15.1   | 1220       | 1.25                                 | 0.81              |
|    | CS       | 11.0   | 1350       | 1.20                                 | 0.77              |
| R20| FS       | 14.0   | 1175       | 1.45                                 | 0.98              |
|    | CS       | 8.8    | 1290       | 1.35                                 | 0.87              |
| R30| FS       | 12.2   | 1015       | 1.90                                 | 1.14              |
|    | CS       | 7.6    | 1160       | 1.85                                 | 1.05              |
| R40| FS       | 9.3    | 900        | 2.00                                 | 1.23              |
|    | CS       | 4.5    | 985        | 1.95                                 | 1.19              |
| R50| FS       | 5.9    | 800        | 2.10                                 | 1.34              |
|    | CS       | 3.0    | 825        | 2.00                                 | 1.27              |

**Figure 2.** Swelling percentage of SMR L / RS compounds

### 5. Summary

The addition of RS in SMRL has affected on the properties of the rubber compounds. From curing characteristics, it is shows that higher RS loading gave shorter scorch and cure time in SMRL compounds and increased the torque. On the other hand, it is shows that increment in RS content improved modulus, hardness and crosslink density but reduced the tensile strength and elongation at break. However, the fine size of RS gave better properties than coarser size at same loading.

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