Cure Characteristics and Physical Properties of SMR L/EPDM Blends: Effects of Blend Ratios

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Abstract. The effects of different blend ratios of Standard Malaysian Rubber (SMR L)/Ethylene Propylene Diene Monomer (EPDM) blend with different ratios on the cure characteristics (cure time, scorch time, minimum and maximum torque) and physical properties (swelling test, crosslink density and hardness test) were studied. The blends of (SMR L)/ (EPDM) (100/0, 90/10, 80/20, 70/30, 60/40, 50/50, 40/60, 30/70, 20/80, 10/90 and 0/100 phr) were prepared by using laboratory two-roll mill. The result showed that the ratio 80/20 showed the best properties compared to the other blend ratios. The blend at this ratio exhibited less scorch time, cure time and swelling percentage, and showed highest crosslink density.

1. Introduction

The blending of two or more types of polymers is a useful technique for the preparation and development of materials with superior properties to those of individual constituents [1]. The purpose of the rubber blending is to improve the physical and mechanical properties, as well as modifying the processing characteristics and reducing the cost of the final product [2].

Standard Malaysian Rubber (SMR L) is a natural rubber (NR), which it is sticky and colored rubber. Recently, SMR L has produced in large quantities. In addition to other properties such as natural product, low cost and easy to use, SMR L can be a good candidate to be used in various applications. Chemically, SMR L has the formula that based on cis-1,4-polyisoprene, which is characterized by good elastic properties, resilience and damping behavior. On other hand, SMR L possesses poor chemical resistance and processing ability. The unique mechanical properties of SMR L are due to the highly stereoregular microstructure resulting from its high molecular weight [3].

Ethylene-propylene-diene monomer (EPDM) rubber is produced by the copolymerization of ethylene and propylene in the presence of non-conjugated diene. The saturated backbone of EPDM resulted from the high mechanical, dynamic and electrical properties. Furthermore, EPDM enjoys many noble properties such as resistance to aging. Heat and oxidation and low temperature flexibility with high chemical and swelling resistance. These inherent properties make EPDM the preferred elastomers for outdoor application such as automotive sealing systems. Wire and cable insulation, building profiles, roofing sheets and under-the-bonnet application [4].
Blends of natural rubber (NR) and ethylene propylene diene monomer (EPDM) have gained much attention over the past three decades. That was according to the good physical properties with adequately high aging and ozone resistances [5]. However, this works studied the effects of various blend ratios of standard Malaysian rubber (SMR L) as a type of natural rubber (NR) and ethylene propylene diene monomer (EPDM), on the cure characteristics and physical properties of these blends.

2. Experimental

2.1 Materials

Standard Malaysian natural rubber grade L (SMR L) and ethylene propylene diene monomer (EPDM) rubber were supplied by Rubber Research Institute Malaysia (RRIM), wood and saw dust that used as natural fillers were supplied by Saad perabot, perlis, whereas CaCO3, synthetic filler was purchased from Anchor Chemical Co. (M) Ltd. The other chemicals that used in the compounding process such as zinc oxide and steric acid (S. A.) as activators, N-cyclohexyl-2-benzothiazole sulphonamide (CBS) as accelerator, butylated hydroxytoluene (BHT) as antioxidant and sulphur as vulcanizing agent were supplied by Anchor Chemical Co. (M) Ltd.

2.2 Formulation of SMR L/EPDM compounds with different fillers

SMR L/EPDM was used with 11 different ratios and blended using two roll mill model X(S)K-160 X 320. The weight of compound was 250g per loading. The formulation was in per hundred rubbers (phr) and as stated in Table 1. 100/0 and 0/100 were the control compounding for this study. The incorporation of blend ration SMR L/EPDM compound is shown in Table 1.

| Materials | Blend ratios (phr) |
|-----------|-------------------|
|           | 100/0 90/10 80/20 70/30 60/40 50/50 40/60 30/70 20/80 10/90 0/100 |
| SMR L     | 100     90     80     70     60     50     40     30     20     10     0  |
| EPDM      | 0       10     20     30     40     50     60     70     80     90     100  |
| Sulphur   | 3       3      3      3      3      3      3      3      3      3      3  |
| CBS       | 1       1      1      1      1      1      1      1      1      1      1  |
| ZnO       | 5       5      5      5      5      5      5      5      5      5      5  |
| S. A.     | 1       1      1      1      1      1      1      1      1      1      1  |
| BHT       | 1       1      1      1      1      1      1      1      1      1      1  |

2.3 Cure Characteristic test

After compounding process, rubber compounds were taken out from freezer and defrozen at room temperature for 30 minutes for the purpose of curing characteristic test. Curing characteristic test was done by using Rheometer Model HT-M2000. Rheometer was heated until achieve the desired curing temperature which is 160 °C. The test was run for 20 minutes. 4.0g of uncured compound rubber was weighed and placed on the heated rotor. and the heated top die cavity was immediately brought down on to the lower dies thus filling the cavity. The machine plots a graph of torque verses time. Curing characteristic such as minimum torque ML maximum torque MH. Cure time (t90) and scorch time (t2) Were obtained.

2.4 Hardness test

Hardness of the samples was measured according to ASTM D-2240-81 using a Shore A type durometer which employed a calibrated spring to provide the indenting force. Since the hardness reading decreased with time after establishment firm contact between the indentor and the sample, the recording immediately after establishment to firm contact were taken. The measured values of hardness were obtained at three different points distributed over test piece. Three test pieces were used and their average value was determined.
2.5 Swelling test and Crosslink density
A cured test piece of above dimension was weighed using an electronic balance before swollen in toluene. After that, all parts of test piece were fully immersed in toluene. After 48 hours, test piece was taken out, wiped, weighed again rapidly. Swelling bottle that contains test sample and toluene need to be closed neatly because of rapid evaporation of toluene. Precautions need to be taken into account during handling toluene because this solvent is hazardous. Swelling percentage was calculated by the equation 1.
Swelling percentage = \( \frac{(M_2-M_1)}{M_1} \times 100 \) (1)
Which M1 is the initial mass of the tested piece (g) and M2 is the mass of it after swollen in toluene.

3 Results and Discussions

3.1 Cure Characteristics
Table 2 shows the scorch time (t2), cure time (t90), Minimum Torque (ML) and Maximum Torque (MH) obtained for SMR L/EPDM with different blend ratios.

3.1.1 Scorch time (t2).
The results indicated that the t2 of SMR L /EPDM blends decreased until reached the ratio 80/20 phr and increased gradually. The optimum t2 was exhibited by 80/20 phr blend ratio. Lower t2 means that the compound starts to crosslink faster when the SMR L dominant than EPDM at this ratio. This is because SMR L is highly unsaturated, non-polar rubber and it is chemically non-reactive while the EPDM is highly saturated, polar rubber and chemically reactive. It can keep in elastic state at a wide temperature range. During vulcanization, SMR L possesses a higher crosslinking rate and curatives. It also has a great potential to diffuse toward the SMR L phase [6].

3.1.2 Cure time (t90) and cure rate index CRI.
From the results in Table 2, it can be seen that similarly to scorch time t2, also t90 continuously decreased until 80/20 phr then increased over80/20 phr with the increasing of the concentration of EPDM content. The augmentation in t90 of SMR L /EPDM could be attributed to the low vulcanization efficiency of EPDM for the sulphur system because EPDM has comparatively low diene content. The result shows an increasing from in the values of CRI from100/0 phr until 80/20 phr then decreased to 0/100 phr. The cure rate index indicated that the rate of vulcanization SMR L/EPDM decreased slightly with increasing levels of EPDM. It is proven that the effect of cure incompatibility and the migration of the accelerator indicated to a decrease in the number of reactive sites on the rubber molecules used for the cross-linking reactions [7].

| Blend ratios | Scorch time t2 (min:s) | Cure time t90 (min:s) | Cure rate index CRI (min⁻¹) | Minimum torque ML (dNm) | Maximum torque MH (dNm) |
|--------------|------------------------|-----------------------|-----------------------------|-------------------------|-------------------------|
| 100/0        | 2.10                   | 12.00                 | 10                          | 2.52                    | 12.52                   |
| 90/10        | 1.33                   | 9.13                  | 12                          | 2.82                    | 13.95                   |
| 80/20        | 1.23                   | 4.06                  | 36                          | 3.07                    | 14.81                   |
| 70/30        | 1.34                   | 5.02                  | 29                          | 3.31                    | 16.27                   |
| 60/40        | 1.40                   | 5.33                  | 27                          | 3.55                    | 17.36                   |
| 50/50        | 1.47                   | 6.02                  | 24                          | 3.82                    | 18.91                   |
| 40/60        | 1.52                   | 6.46                  | 20                          | 4.05                    | 19.94                   |
| 30/70        | 1.56                   | 7.32                  | 16                          | 4.33                    | 21.1                    |
| 20/80        | 2.01                   | 9.42                  | 12                          | 4.58                    | 23.63                   |
| 10/90        | 2.06                   | 11.15                 | 9                           | 4.83                    | 25.21                   |
| 0/100        | 2.32                   | 14.52                 | 6                           | 5.06                    | 28.83                   |
3.1.3 Minimum torque (ML) and maximum torque (MH).
Minimum and maximum torque of SMR L/EPDM with different ratios are shown in Table 2. Minimum torque increased as increasing of weight ratios of EPDM. This result indicated that EPDM had a negative impact on the processibility of the blends. This is according to the low diene content. In addition to presence of crosslinked EPDM and other factors. The same situation of maximum torque MH, it has increased gradually with the increasing of EPDM content up to the ratio 0/100, which means completely EPDM. From both ML and MH values, it can be noticed that the presence of EPDM makes the processibility of rubber compounds more difficult compared to SMR L rubber.

3.4 Hardness test
From the results in Table 3, it can be seen the effect of hardness test on SMR L/EPDM blends with different ratios. As can be observed, the trend of hardness test of SMR L/EPDM blends increasing with the increasing of EPDM content. As more EPDM were added and thus incorporated in SMR L, the flexibility and elasticity of the rubber chain were decreased. It can be seen that the SMR L/EPDM became more rigid vulcanized rubber thus the hardness was increased. It was proven that the increment of hardness value of SMR L/EPDM blends due to the present of various additives and precursors of EPDM [8]. SMR L/EPDM blend at 80/20 phr was selected as the optimum hardness value. SMR L was more dominant than EPDM. It also indicated that the incorporation of EPDM in the SMR L/EPDM blend at 80/20 phr enhanced the crosslink density properties of the blend. This is because its stiffness and became more rigid of EPDM when added at low concentration.

3.5 Swelling percentages
The swelling percentages were determined by using toluene as a solvent at room temperature until equilibrium swelling was reached. As can be interpreted that the swelling percentages decreased gradually until 80/20 phr and increased back up to 0/100 phr. More content of EPDM was a reason to increase the swelling percentage of SMR L/EPDM. The optimum value of swelling percentage that observed in SMR L/EPDM was at ratio 80/20 phr which exhibited the lowest value compared to other ratios. It is widely accepted that the swelling is directly correlated to the crosslink density of a network chain [5,9,10], with less solvent uptake or penetration into the blends indicating higher crosslink density.

| Blend ratios (SMR L/EPDM) | Swelling percentages (%) | Hardness test (Shore A) |
|---------------------------|--------------------------|-------------------------|
| 100/0                     | 313                      | 38.4                    |
| 90/10                     | 195                      | 42.8                    |
| 80/20                     | 272                      | 43.4                    |
| 70/30                     | 195                      | 46.8                    |
| 60/40                     | 307                      | 47.6                    |
| 50/50                     | 318                      | 48.4                    |
| 40/60                     | 335                      | 49.3                    |
| 30/70                     | 352                      | 50.2                    |
| 20/80                     | 364                      | 52.3                    |
| 10/90                     | 375                      | 54.5                    |
| 0/100                     | 388                      | 57.1                    |

4. Conclusion
The increasing content of EPDM in SMR L/EPDM blends made a significant alteration on the curing characteristics and also physical properties. The optimum ratios showed by SMR L/EPDM at 80/20 phr. However, the usage of EPDM at 20 phr showed lower cure and scorch time compare to other different ratios and as well as it has increased the minimum torque and maximum torque. The physical
properties such as swelling percentage and hardness test of SMR L/EPDM at 80/20 phr showed the better properties compared to other blends.

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