EFFECT OF CHEMICAL PRE-TREATMENT ON THE CURE, MECHANICAL AND ABRASION PROPERTIES OF KENAF/NATURAL RUBBER GREEN COMPOSITES

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1. Introduction

Rubber materials have many advantages like low cost, excellent processibility, flexibility resilience, and elasticity over metals. However they also have drawbacks like recycling, rubber waste, incineration, etc., causing environmental problems. Hence many researches focusing on recyclable rubber and naturally available rubber increasingly interested in industries and academia [1]. Natural rubber, which is available from Hevea Brasiliensis, commonly called 'rubber tree', is referred to as cis-1,4-polyisoprene. Most of the raw rubbers can be obtained as latex through a tapping process from Malaysia, Thailand and India. Natural rubber is composed of hydrocarbons (89.3~92.4 wt%), protein (2.5~3.5 wt%), and other ingredients (4.1~8.2 wt%). It has good mechanical properties and bending resistance but less thermal stability and low resistance to ozone, acids and oils.

Recently, cellulose-based natural fibers like kenaf, jute, hemp, ramie, etc. are increasingly utilized in many academia and industries, particularly in the research areas using eco-friendly materials and applications for eco-friendly automobile parts and building materials since it has a variety of advantages over conventional glass fibers such as low cost, natural abundance, eco-friendliness, low density, acceptable mechanical properties, good abrasion resistance, etc. [2-5]

An introduction of eco-friendly natural fibers to natural rubber can play a role not only in reinforcing rubber but also in reducing the amount of carbon black used in rubber and tire applications. However many researches on natural fiber reinforced rubber composites have not been reported yet [6,7], compared to green composite areas using other thermosetting or thermoplastic resins. Kenaf fiber, which can be extracted from the bast of kenaf plant, is one of the most effectively CO₂ absorbing natural fibers during plantation. It has good mechanical properties so that it has been utilized as industrial ropes and reinforcement.

One of the main drawbacks of natural fibers is, in general, poor interfacial adhesion between natural fiber and polymer matrix. It has been expected that an enhancement of the interfacial adhesion is very important to improve relevant properties of resulting green composites. Therefore if the interfacial adhesion between kenaf and natural rubber can be enhanced, natural fibers may play a role in enhancing the properties of natural rubber [8]. Consequently, the objective of the present study is to improve the mechanical properties and abrasion resistance of green composites composed of natural rubber and kenaf fibers pre-treated with Chemlok 402.

2. Experimental

2.1 Materials

In the present study, natural rubber (NR) (SMR CV60, Malaysia) was used. Kenaf was supplied from Bangladesh. Carbon black (FEF grade) was used. Sulfur was used for vulcanization of NR and the compound. Vulcanization aid was zinc oxide. Diphenyl quanidine was used as accelerator. Chemlok 402 (Lord Korea, Korea) was used as pre-

treatment agent of kenaf. N2 oil was used as plasticizer.

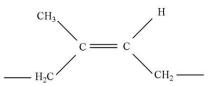


Fig. 1. Structure of natural rubber (cis-1,4-Polyisoprene).

2.2 Chemical Pre-treatment of Kenaf Fibers

Prior to pre-treatment, kenaf was sufficiently dried at 80°C for 24 h. Pre-treatment of kenaf fibers was performed with Chemlok 402 dissolved in toluene (50:50 vol%). Kenaf was soaked in the Chemlok 402/toluene solution for 10 min. The pre-treated kenaf was rested at room temperature for 60 min. Pre-baking process was conducted at 80°C for 10 min in order to avoid the removal of Chemlok 402 by friction during compounding. Then, pre-treated kenaf was rested at ambient temperature for 24 h before compounding. Pre-treated kenaf fibers were incorporated with NR at 0, 5, 10, 15 and 20 phr, respectively.

2.3 Processing and Vulcanization

The mastication of natural rubber was conducted by using a Banbury mixer in the temperature range of 100-110°C at 40 rpm for 1 min under a ram pressure of 100 psi. The compounding process with kenaf and other ingredients was done using a Banbury mixer after the mastication. The maximum temperature for compounding was 135°C at 40 rpm for 2 min under a ram pressure of 100 psi.

The sheet-type compound was rested between 15°C and 25°C for 24 h. Vulcanization of the compound was performed 160, based on the pre-test result obtained by an oscillating disk rheometer (ODR). The vulcanized kenaf/natural rubber green composites were cut to 180 mm×180 mm×2 mm for test specimens.

2.4 Characterization

The curing characteristics (maximum and minimum torques, ts₂, and Tc₉₀) of natural rubber and kenaf/natural rubber green composites were measured at 190°C according to ASTM D2084 by means of an oscillating disc rheometer (ODR, Myungji, Korea). Hardness test was performed using

a spring-type Shore hardness tester (Shore A type) (Kobunshi Keiki Co., LTD, Japan).

The tensile test was performed according to KS 6518 using a tensile tester (Zwick Z005, Swiss). Abrasion tests were conducted according to ASTM D1630 using a NBS-type abrasion tester (Yasuda, Japan) (Fig. 2) and the abrasion ratio was obtained by comparing the weights of the specimen and the reference lost after testing with 300 revolutions for each specimen. The downward force was 5 lb_f. The specimen dimensions were 25 mm \times 25 mm \times 6 mm. A scanning electron microscope was used to observe the fractured surfaces of composite specimens.

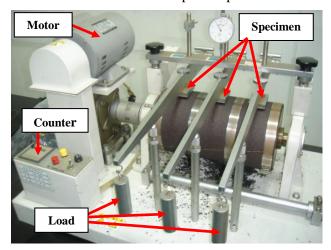


Fig. 2. NBS-type abrasion tester used in this work.

3. Results and Discussion

The maximum torque (T_{max}) monitored by the ODR during vulcanization was increased with increasing the kenaf fiber content in both untreated and treated kenaf/natural rubber green composites. The specimen treated with Chemlok 402 exhibited the greater T_{max} value than the untreated counterpart. As seen in Fig. 3, the difference in the T_{max} between the treated and untreated composites became greater when the kenaf content was higher than 10 phr. It was expected that the interfacial adhesion between the kenaf fibers and the natural rubber with the assistance of Chemlok 402, resulting from physical interaction or mechanical interlocking,

Fig. 4 shows the variation of tc_{90} as a function of kenaf fiber content for natural rubber and untreated and treated kenaf/natural rubber green composites. Here tc_{90} is defined as the time that 90% of vulcanization is accomplished. The result indicated

that the tc₉₀ was gradually reduced with increasing the kenaf content. This turns out that the vulcanization rate becomes faster when kenaf fibers were added with natural rubber. This is probably because kenaf fibers contributed to transferring heat to the rubber matrix during vulcanization. In addition, it may be expected that the presence of the reinforcing kenaf fibers increased the hardness of the material and as a result the torque was increased with kenaf, as seen in Fig. 3.

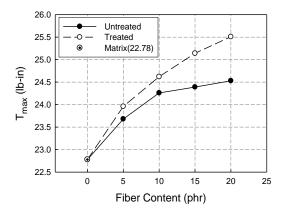


Fig.3. Variation of the maximum torque (T_{max}) measured for natural rubber and kenaf/NR green composites with various kenaf fiber contents. The vulcanization temperature was 190° C.

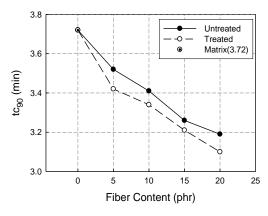


Fig. 4. Variation of the time to reach 90% cure (tc₉₀) measured for natural rubber and kenaf/natural rubber green composites with various kenaf fiber contents.

Hardness is a useful measure for determining the flexibility and rigidity of vulcanized rubber. Fig. 5 exhibits the variation of the hardness of kenaf/natural rubber green composites with varying the kenaf content. The result indicated that the

increased hardness with kenaf is due to the increased reinforcing effect by kenaf and the hardness can be further enhanced by treating kenaf fibers with Chemlok 402, reflecting the increased adhesion at the interfaces between the fibers and the rubber matrix.

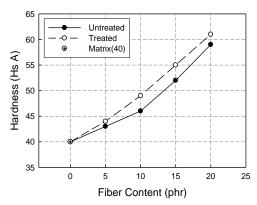


Fig. 5. Variation of the hardness of kenaf/natural rubber green composites as a function of kenaf content.

Fig. 6 depicts the tensile modulus of natural rubber and kenaf/natural rubber green composites as a function of kenaf content. With increasing kenaf, the modulus was gradually increased, as similarly found in the T_{max} and hardness results. The main reason for the increased modulus is that the incorporation of the crystalline kenaf fibers with a high aspect ratio into natural rubber increased the stiffness of the material.

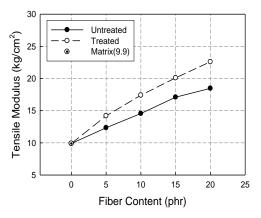


Fig. 6. Variation of the tensile modulus of kenaf/natural rubber green composites as a function of kenaf content.

The addition of kenaf fibers to natural rubber decreased the tensile strength of neat natural rubber

and the strength was further decreased with increasing the untreated kenaf and the treated kenaf to 15 wt%, as shown in Fig. 7. This can be explained by that cellulose-based kenaf fibers have a large number of cells in every single fiber and they may act as microstructural defects leading to the reduction of tensile strength. The tensile strength was slightly increased by the pre-treatment of kenaf with Chemlok 402 at 20 phr, indicating that the increased adhesion between the kenaf and the rubber matrix somewhat contributed to the mechanical strength of the composite at a higher kenaf content.

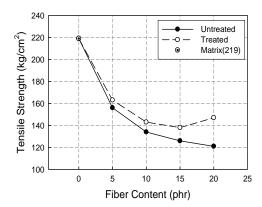


Fig. 7. Variation of the tensile strength of kenaf/natural rubber green composites as a function of kenaf content.

Fig. 8 shows the abrasion resistance of natural rubber and kenaf/natural rubber green composites. The abrasion ratio was increased with kenaf fiber, indicating that the addition of kenaf was rather decreased the abrasion resistance of natural rubber. However the resistance was enhanced by the pretreatment of Chemlok 402. The abrasion ratio of the composite greater than NR may be due to the brittleness of kenaf distributed in the natural rubber. Kenaf fibers have the greater modulus and stiffness than natural rubber, which is tough and ductile.

Fig. 9 presents the fractured surfaces of untreated and treated kenaf/natural rubber composites. It was clearly found that the untreated sample exhibited a pull-out fiber with the debonded region around the kenaf fiber, indicating a poor adhesion between the fiber and the matrix. On the other hand, the treated sample exhibited a shorter length of pull-out fiber and the better fiber-matrix adhesion. As a result, it may be said that the enhancement of the interfacial property between the kenaf fibers and the natural

rubber matrix contributed to influencing the vulcanization characteristics, hardness, mechanical properties, and abrasion resistance of kenaf/natural rubber green composites.

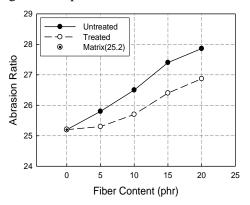


Fig. 8. Variation of the abrasion ratio of kenaf/natural rubber green composites as a function of kenaf content.

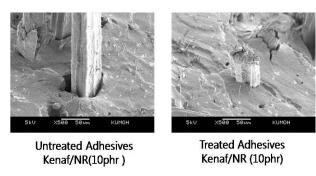


Fig. 9. SEM micrographs of the fracture surfaces observed with untreated and treated kenaf/natural rubber green composites (\times 500).

4. Conclusions

In this work, kenaf/natural rubber green composites were fabricated through mastication, compounding and vulcanization processes. It was concluded that with increasing chopped kenaf fiber content the ODR torque, tensile modulus and hardness of the composites were increased whereas the tc₉₀ and abrasion resistance were decreased. The ODR torque, mechanical properties, hardness and the abrasion resistance were enhanced by chemical pre-treatment with Chemlok 402 done to kenaf fibers prior to the composite process. The microscopic result of the fracture surfaces supported the test results.

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