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Mechanical Properties of Lignite Fly Ash/Natural Rubber Composites: Effects of Filler Size, Loading and Silane Coupling Agent

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Abstract

Composites of lignite fly ash particle and natural rubber were prepared by conventional mixing and molding process. Mechanical properties of the natural rubber composites were studied. It was found that tensile strength, elongation at break, tear resistance and abrasion resistance decreased but hardness increased with increasing lignite fly ash loading. The tensile strength and tear resistance of composite of the small size fly ash (SF) filled the rubber are higher than the medium size (MF) and as-received (AF) one. The moduli at 300% and 500% elongation of SF-filled natural rubber composite increase with increasing fly ash loading. The silane coupling agent was found improving the tensile strength, tear resistance and modulus of the composites.

Keywords: natural rubber, lignite fly ash, mechanical properties, silane coupling agent

1. Introduction

Mechanical properties of natural rubber can be modified by mixing with different fillers such as carbon black, silica, calcium carbonate and clay. Recently, low cost available fillers both particulate and fibrous forms play an important role for improving properties of soft rubber comparatively to those high cost fillers. In this study, particulate lignite fly ash has been chosen to composite with natural rubber due to its containing hard and rigid potentially reinforcing compound of SiO_2, Al_2O_3 and Fe_2O_3. Lignite fly ash is a by-product obtained from lignite combustion process having a specific gravity of 2.00-2.60 and relatively sphere in shape.

Rice-husk ash was also interested to use as filler in a rubber because of its containing silica generally use as reinforcing fillers. Rice husk ash provides inferior mechanical properties (tensile strength, modulus, abrasion resistance, tear resistance and hardness) compared with neat silica and carbon black. However, its mechanical properties are comparable to those filled with inert fillers [1]. The addition of silane coupling agent showed little effect on the properties of the ash-filled vulcanizates [2-3]. Mechanical properties of bamboo fibre filled natural rubber composites such as tensile strength, tear resistance and elongation at break decreased but tensile modulus and hardness increased with increasing the fibre loading [4]. Moreover from the previous study [2, 5] it was found that the addition of a silane coupling agent improves the mechanical properties such as tensile strength, tensile modulus and tear resistance. The composite of oil palm wood flour and a natural rubber was also studied and found that the higher amount of the wood flour loading resulting in lower tensile strength, tear resistance and elongation at break in contrast with tensile modulus and hardness [6].

This research aims to study the effects of size and loading of lignite fly ash in the rubber and a silane coupling agent on mechanical properties such as tensile strength, moduli at 300% and 500% elongation, elongation at break, tear resistance, hardness and abrasion resistance.

2. Materials, equipments and method

2.1 Materials and chemicals

The natural rubber chosen in this research is the STR 5L. Lignite fly ash was collected and supplied by Mae-Moh Power Station (Lampang, Thailand). Chemicals used are sulfur, accelerator (MBT and DPG), activator (zinc oxide and stearic acid) and a silane coupling agent (Si-69). The formulation of rubber composite was shown in Table 1.

Table 1. The rubber composite formulation

Chemicals	Quantity (phr[a])
Natural rubber	100.0
Zinc oxide	5.0
Stearic acid	2.0
MBT[b]	0.5
DPG[c]	0.2
Sulfur	3.0
Lignite fly ash	0, 10, 30, 50, 70
Si-69[d]	3% w/w on filler

[a]parts per hundred rubbers
[b]2-mercaptobenzthiazole
[c]diphenyl guanidine
[d]bis [3-(triethoxysilyl) propyl] tetrasulfide

2.2 Testing equipments

(1) Universal testing machine (Model LR 10K, LLOYD)
(2) Durometer Shore A
(3) Akron abrasion machine

(4) Laser Particle Size Analyzer (Model LS230, Coulter)

2.3 Method

2.3.1 Lignite fly ash preparation

Three average sizes of lignite fly ash of as-received fly ash (AF), small-size fly ash (SF) and medium-size fly ash (MF) were used in this studied. The AF was sieved to MF on the 400 mesh sieves. The MF was mills to SF with a planetory ball mill for 6 hours. The laser particle size analyzer was used to analyze the average particle size of lignite fly ash as shown in Table 2.

Table 2. The average particle size of lignite fly ash

Lignite fly ash	Average particle size (μm)
AF	15.95
MF	4.98
SF	2.19

2.3.2 Rubber composite preparation

Two-roll mill was used to mix rubber compound from natural rubber, lignite fly ash and chemicals according to Table 1 for 20-25 minutes. Then the rubber compound was kept in a desiccator at room temperature for 16-24 hours. After that, it was molded at 150^{o}C for specimen preparation.

2.3.3 Tensile test

The tensile properties of rubber composite such as tensile strength, moduli at 300% and 500% elongation and elongation at break were tested according to ASTM D412-80 (Die C) with dumbbell-shaped samples and a universal testing machine at a speed of 500 mm/min.

2.3.4 Tear resistance test

Tear resistance was determined according to ASTM D624-81 (Die C) with angle-shaped samples. The universal testing machine was used for tests at a speed of 500 mm/min.

2.3.5 Hardness test

Hardness was determined according to ASTM D2240-81 with a Durometer Shore A.

2.3.6 Abrasion resistance test

Akron abrasion machine was used for abrasion resistance tests according to B.S. 903 Part A 9, Method C. The results are reported in term of the volume loss of the rubber composites.

3. Results and discussion

3.1 Tensile Properties

Figure 1 shows the relationship between filler size, loading and tensile strength of lignite fly ash filled natural rubber with and without the silane coupling agent. It can be seen that the tensile strength decreases with increasing fly ash loading. The decrease of tensile strength was due to low filler-rubber interactions (the lack of reactive function group on the ash surface such as hydroxyl group) [3, 7]. The tensile strength of SF-filled the natural rubber composite is higher than that of MF-filled and AF-filled the rubber due to the size effect, suggested that the surface area of SF to interact with the rubber surfaces was greater [7]. The result indicated that the presence of Si-69 caused an improvement to the filler-matrix interfacial bonding giving the tensile strength of the rubber composite with Si-69 is higher than the composite without Si-69.

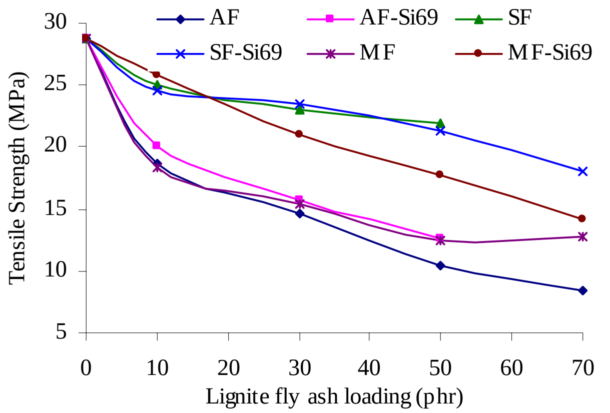

Figure 1. Effects of fly ash size, loading and a silane coupling agent on tensile strength

The effects of fly ash size, loading and a silane coupling agent on elongation at break are shown in figure 2. As expected, the elongation at break reduces with increasing lignite fly ash loading due to the brittleness of the composites increased [6]. The elongation at break of the natural rubber composite without Si-69 is higher than the natural rubber composite with Si-69. The presence of Si69 cause the ductility of composites decreases due to the enhancement in rigidity, so the composite break at lower elongation [8].

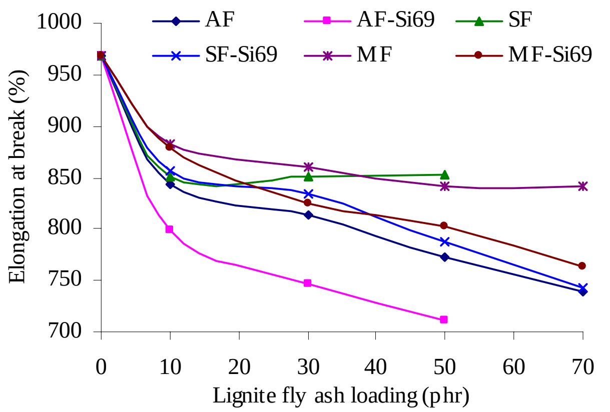

Figure 2. Effects of fly ash size, loading and a silane coupling agent on elongation at break

Figure 3 and 4 show the moduli at 300% and 500% elongation versus fly ash size and loading with and without a silane coupling agent. It can be seen that the moduli at 300% and 500% elongation of SF-filled the rubber composites increase with increasing filler loading, whereas the moduli of composites of AF-filled and MF-filled the rubber decrease with increasing filler loading. The addition of rigid and stiff particulate filler would increase the modulus of the rubber composites due to the introduction of restrictions to the molecular mobility of the rubber [8]. However, the inferior modulus of composites of AF-filled and MF-filled the rubber may be explained by the particle size which AF and MF have larger particle size and, therefore, smaller surface areas for interactions. The moduli at 300% and 500% elongation increase with the addition of Si-69 because the improvement of interaction between filler and rubber surfaces similar to the results of the previous studies [8-9].

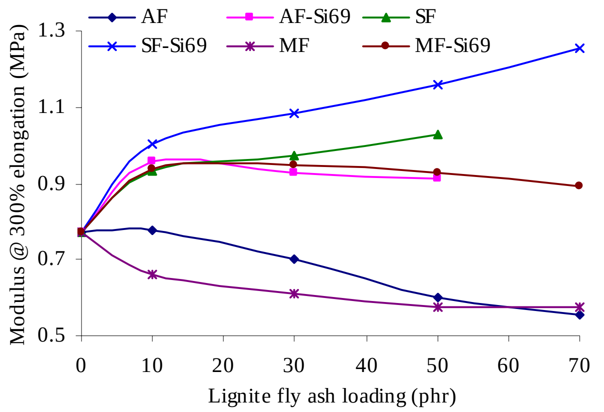

Figure 3. Effects of fly ash size, loading and a silane coupling agent on modulus at 300% elongation

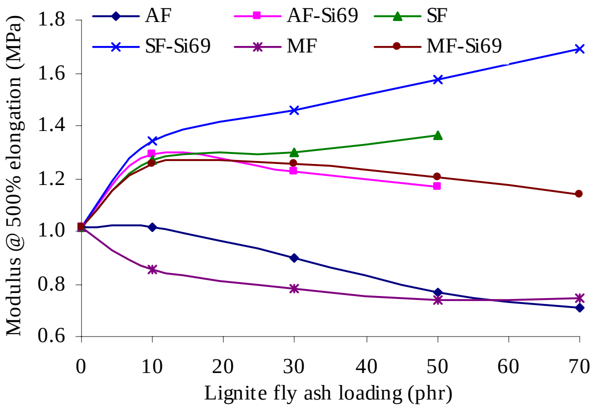

Figure 4. Effects of fly ash size, loading and a silane coupling agent on modulus at 500% elongation

3.2 Tear resistance

The relationship between filler size, loading and a silane coupling agent on the tear resistance of lignite fly ash filled natural rubber are shown in figure 5. The tear resistance of the vulcanizates tends to decrease with increasing fly ash loading due to the poor interface interaction between fly ash particles and the rubber phase [7]. The composite of SF-filled rubber has tear resistance higher than MF-filled and AF-filled natural rubber composite because the surface area of SF was greater than MF-filled and AF-filled rubber [7]. The tear resistance of the vulcanizates with Si-69 is higher than that without Si-69 because of the improvement in filler-rubber interactions, as discussed earlier [2, 8-9].

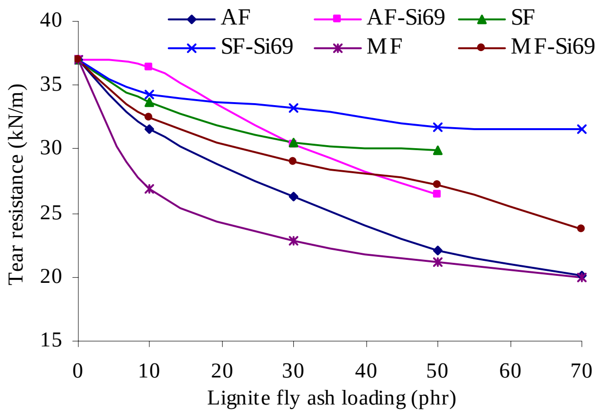

Figure 5. Effects of fly ash size, loading and a silane coupling agent on tear resistance

3.3 Hardness

The hardness of composites of lignite fly ash filled a natural rubber with and without Si-69 was shown in figure 6. It was observed that hardness increases as the fly ash loading increases. This result is expected due to the higher incorporating of fly ash particles, the lower the elasticity of the rubber chains, resulting in more rigid vulcanizates [1, 3]. The hardness of natural rubber composites with and without Si-69 were not difference because the silane coupling agent is liquid in nature and a small quantity was used [9].

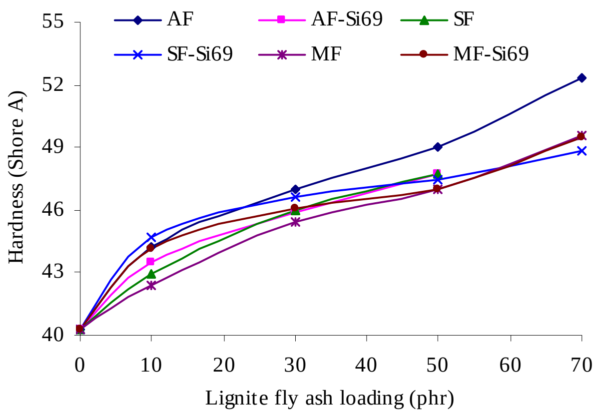

Figure 6. Effects of fly ash size, loading and a silane coupling agent on hardness

3.4 Abrasion resistance

Figure 7 shows the effects of fly ash size and loading on the abrasion resistance of composites of the rubber with and without Si-69. The results reported in term of the volume loss of the rubber composites. The greater the volume loss was, the lower the abrasion resistance. The abrasion resistance decreases with increasing lignite fly ash loading. The maximum abrasion resistance is found from the composite of MF-filled a rubber at 10 phr that it is higher than the vulcanizated rubber without lignite fly ash about 14%. The presence of Si-69 has no significant effect on the abrasion resistance.

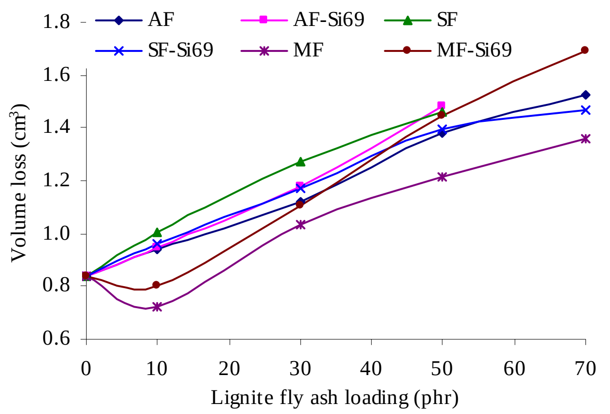

Figure 7. Effects of fly ash size, loading and a silane coupling agent on abrasion resistance

4. Conclusion

The results show that tensile strength, elongation at break, tear resistance and abrasion resistance decrease excepting hardness increases with increasing lignite fly ash loading in natural rubber composites. The SF-filled natural rubber composite has tensile strength and tear resistance better than The MF-filled and AF-filled natural rubber composite due to MF and AF have larger particle sizes and, therefore, smaller surface area than SF. The moduli at 300% and 500% elongation of composite of SF-filled natural rubber increase, whereas, these of MF-filled and AF-filled natural rubber decrease with increasing lignite fly ash loading. The presence of the silane coupling agent improves mechanical properties such as tensile strength, tear resistance and moduli at 300% and 500% elongation of the composites. However the filler particle size and the silane coupling agent have no significant effect on elongation at break, hardness and abrasion resistance.

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