Reinforcement of natural rubber using silica and zeolite mixed fillers

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Introduction

Among the main ingredients added in the compounding process of Natural Rubber (NR), fillers play a major role. The purpose of adding fillers into the rubber matrix is 2-fold: to reduce the production cost or to give reinforcement. (Blackley, 1997). A reinforcing filler would increase the mechanical properties such as tensile strength, elongation at break, and tear resistance of the rubber vulcanizate. Increasing the area of contact between rubber matrix and filler particles, and increasing the degree of bonding between the two phases seem to be the most important factors in providing the strong reinforcement effect (Treloar, 2005). The current research is focused on a combination of two fillers, namely silica and zeolite, to provide a reinforcing effect on NR. Silica is in general widely used for reinforcement of NR. On the other hand, zeolites were considered due to their unique porous structure which would enhance the area of contact between the phases.

Methodology

Materials

Double Centrifuged Natural Rubber Latex (DCL) with 60% dry rubber content (stabilized with ammonia) provided by Dipped Product Industries in Sri Lanka was used in the preparation of the samples. Mineral samples, Silica and Zeolite were purchased from Glorchem, Colombo. The rubber additives, namely, zinc oxide, sulphur, antioxidant, dispersing agent, Zincmercaptobenzothiozole (ZMBT), Zincdiethyldithiocarbamate (ZDDC), Diphenylguanidine (DPG), etc) were of commercial grades.

Procedure

The samples were prepared using NR latex compound formulation used for glove manufacture. The mineral samples were ground using “TEMA mill” and were sieved to get the particle sizes in the range of (45 – 60) µm. A stabilized natural rubber was obtained by adding potassium laurate (20%) and KOH (10%, 2.50 g) to natural rubber latex (60%, 167 mL). ZnO (1.00 g), Zincdiethyldithiocarbamate (0.20 g), Zincmercaptobenzothiozole (1.00 g), Diphenylguanidine (1.20 g), antioxidant (1.50 g), dispersing agent (0.20 g) and Sulphur (1.40 g) were ground together using a mortar and a pestle. The ground mixture was dissolved in distilled water (8.30 mL) using a magnetic stirrer to obtain the dispersion. The stabilized NR solution and the dispersion were mixed together and distilled water was added to dilute the total dry rubber content of the solution up to 40%. This sample was labelled as “NR” and was used as the control sample.

The amount of mineral needed to make up 1%, 2%, 3%, 4% and 5% of the final weight of the sample was calculated and added to the dispersion. In this research, two series of samples were prepared: first series was prepared by adding silica as the mineral and was labeled “Sil” and the

second series was prepared by adding a 1:1 mixture of Silica and Zeolite as the mineral and was labeled "Mix". Calculated amounts of sieved minerals were added to both series. The solutions (NR, Sil and Mix) were then filtered and poured into small glass tanks, covered and left overnight to dry. The samples were then cured for 3.5 min at 120 °C using the hot box oven (Gallencamp). The entire procedure was triplicated.

The mechanical property analysis was based on tensile and tear strength. The samples with optimum mineral percentages showing the best tensile and tear properties were subjected to the structure determination by Fourier Transformed Infrared (FTIR) analysis and thermal properties by thermogravimetric analysis (TGA).

Results and Discussion

The variation of tensile strength as the type and the amount of mineral filler in NR samples are varied is given in Figure 01.

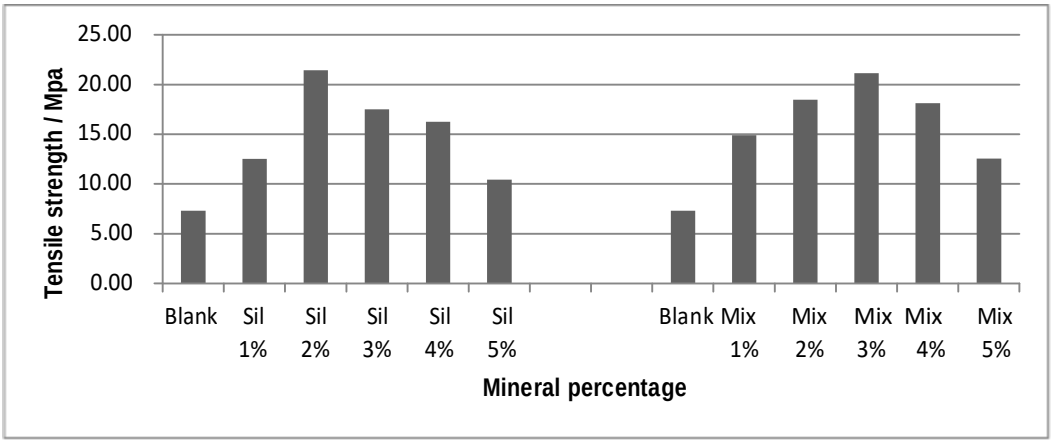

Figure 01. The variation of tensile Strength with varying the type and amount of mineral filler

According to the results, the tensile strength is increased with increasing mineral percentage up to a certain level in both silica and mixed fillers and further increase of mineral percentage has caused the tensile strength to decrease. The maximum tensile strength value for silica loading samples (Sil) was observed at 2% and the relevant value for mixed mineral loading samples (Mix) was observed at 3%. Both type of samples had shown a nearly 200% increment in tensile strength at its optimum load of mineral in the sample, compared to the Blank sample. Similar observations were made for tear strength analysis as well. Enhanced tensile and tear properties of NR at low loads of filler can be explained by polymer–filler interactions and filler–filler interactions within the polymer. However, the expected reinforcement in the filled samples with mixed minerals was not very promising. Porous structure of zeolites combined with reinforcing properties of silica, was expected to give a marked increase in mechanical properties. The ratio of silica and zeolite used, 1:1, may not have produced sufficient polymer-filler interactions. However, when the filler content is higher than 4%, the filler would tend to agglomerate leading to the observed reduction of tensile strength.

The Fourier-Transformed Infra-Red (FTIR) spectra confirmed the incorporation of fillers showing peaks at corresponding frequencies and cleavage of bonding in the filled samples due to the insertion of silica and zeolite into the NR matrix (White, *et al*, 2001).

TGA results of selected 5 samples are given in Table 01. The temperature at which 3% sample weight loss occurs ($T_{3\%}$) is considered as the degradation onset temperature. The temperature at which the highest weight loss rate occurs is set as the degradation temperature (T_{max}).

Sample	Degradation onset Temperature / °C ($T_{3\%}$)	Degradation Temperature / °C (T_{max})	Percentage Weight loss at 200 °C
Blank	180	200	3.0
2% Sil	210	225	2.8
3% Sil	215	223	2.0
2% Mix	210	226	2.7
3% Mix	225	235	2.1

Conclusions

Both tensile and tear properties of NR are improved at low loads of filler. However, when the filler content is higher than 4%, the tensile and tear properties are reduced. The expected reinforcement in the filled samples with mixed minerals of silica and zeolites was not very promising. The ratio of silica and zeolite used, 1:1, may not have produced sufficient polymer - filler interactions. Further research must be carried out by varying the ratio of silica and zeolites to determine the optimum ratio which would provide the greatest reinforcement. However, the thermal properties of all the filled samples are significantly increased.

Acknowledgement

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