

## Viscoelastic Properties of NBR filled with Lignite Fly Ash

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### Abstract

Nitrile rubber (NBR) compounds containing various loadings of fly ash (FA) were prepared, and their viscoelastic properties and storage stability were investigated with the use of oscillatory shear test. Results obtained show that the addition of FA in NBR causes a slight decrease in storage stability, which is believed to be caused by molecular chain-scission induced by heavy metals in FA. With the use of Guth-Gold equation, it could be proposed that the presence of pseudo-network (as formed via FA-FA and FA-NBR interactions) as well as the ball bearing effect are responsible for the reinforcement in FA filled NBR compounds.

### 1. Introduction

Fly ash (FA) is a by-product from combustion of coal in thermal power plants. Extracted by mechanical collectors or electrostatic precipitators or combination of both, the disposal of FA makes such a serious environmental problem by contaminating the surrounding atmosphere and takes up space in landfill for its dumping. There are some attempts to decrease the amount of unusable FA, such as mixing fly ash with cement to achieve high compressive strength concrete [1] and making fly ash brick. In addition, the incorporating of FA particles in rubber can enhance

the shortcomings associated with rubber including its high shrinkage and low stiffness [2].

In this work, the viscoelastic properties and storage stability of NBR compounds filled with FA are studied. Also, a reinforcing effect provided by FA is investigated using the Guth-Gold equation.

### 2. Experimental

#### 2.1 Materials

NBR with acrylonitrile content of 35 % was used as a raw rubber. Fly ash (FA) from Mae Moa Power Station was used as filler.

#### 2.2 Preparation of rubber compounds

Rubber compounds were prepared by the use of 0.5-l Banbury internal mixer at set temperature of 50°C and rotor speed of 40 rpm for 13 minutes. The FA loading was varied from 0-200 phr. The rubber compounds were kept at room temperature in order to investigate the time dependent storage stability.

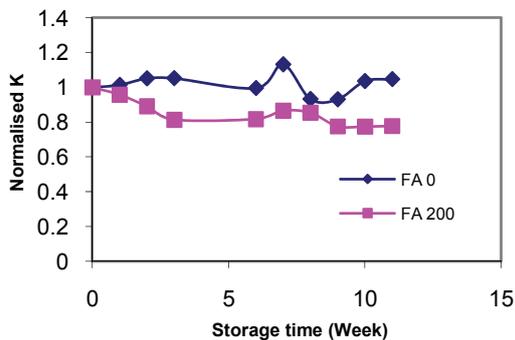
#### 2.3 Measurement of viscoelastic properties

Viscoelastic properties of NBR/FA composites were measured by the Rubber Process Analyzer (RPA2000) under strain and frequency sweep tests at test temperature of 70°C.

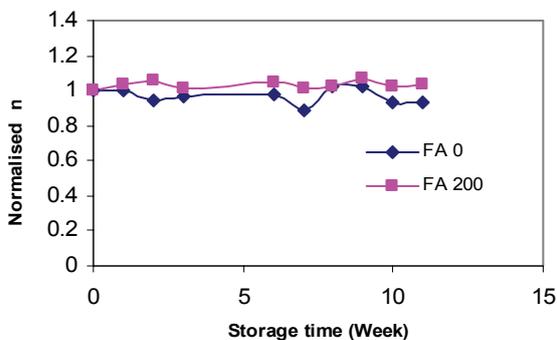
### 3. Results and discussions

The storage stability of FA filled NBR compounds were measured in terms of changes in consistency constant (K), pseudoplasticity (n) and storage modulus (G') as a function of storage time. In theory, minimal changes of these values imply good storage stability of rubber compounds.

Values of normalized consistency constant (K) and power law index (n) of compounds as a function of storage time are shown in Figures 1 and 2.



**Figure 1** Normalised values of consistency constant (K) as a function of storage time of NBR/FA compounds with FA loadings of 0 and 200 phr.

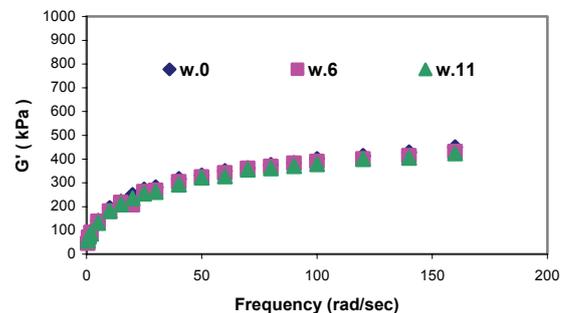


**Figure 2** Normalised values of power law index (n) as a function of storage time of NBR/FA compounds with FA loadings of 0 and 200 phr.

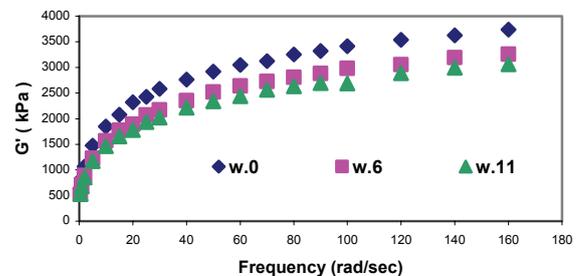
It can be seen that the values of normalised K of unfilled NBR do not change with storage time. On the contrary, the composites with 200 phr FA exhibit the initial drop of about 20 % in the first 3 weeks of storage. Then, the viscosity of filled compound

becomes stable. In the case of pseudoplasticity, it can be seen from Fig. 2 that there is no significantly change in normalised n of both unfilled and highly filled compounds. This means magnitude of shear-thinning behaviour is not affected by storage time.

Figures 3 and 4 reveal storage modulus (G') as a function of angular frequency of NBR compounds with 0 and 200 phr of FA loading. It is evident that G' of all blends increases with increasing frequency which is attributed to the shortened time available for molecular relaxation. Also, it is obvious that, at any given test frequency, the compounds without FA show no discrepancy in G', implying a good storage stability. By contrast, the compounds filled with FA exhibit significant drop in G' as storage time increases. This means the presence of FA is responsible for storage instability of FA filled compounds. It is believed that a decrease in G' is due to the molecular chain-scission.



**Figure 3** Plots of storage modulus (G') against angular frequency of unfilled NBR compound with storage time of 0, 6 and 11 weeks at strain of 1%.

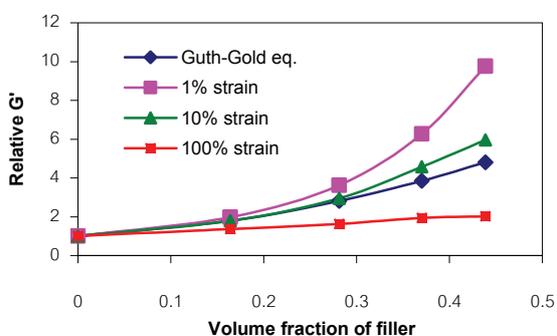


**Figure 4** Plots of storage modulus (G') against angular frequency of 200-phr FA filled NBR

compound with storage time of 0, 6 and 11 weeks at strain of 1%.

In general, it is believed that the presence of heavy metal including copper and manganese would act as strong oxidizing agents to NBR backbone leading to the molecular chain-scission [3].

In order to investigate the reinforcing effect provided by FA, the experimental values of relative modulus as a function of filler volume fraction are compared with the calculated values based on the Guth-Gold equation as illustrated in Figure 5.



**Figure 5** Plots of experimental and calculated values of relative modulus as a function of filler volume of NBR compounds under deformation strain of 1%, 10% and 100% at frequency of 1 rad/sec.

It can clearly be seen that the experimental values measured at small strain of 1% is greater than the theoretical values calculated from the Guth-Gold equation. This implies the presence of filler network among FA particles and/or NBR-FA interaction, which are sometimes known as the pseudo-network or pseudo-crosslink [4]. However, as strain increases to the moderate strain of 10%, values of experimental results are close to those of calculated values, indicating the disruption of such pseudo network. Surprisingly, at further strain of 100%, the experimental data are even lower than the theoretical ones. This is proposed to be due to the molecular

slippage promoted by the ball bearing effect of FA particles with spherical shape [5].

#### 4. Conclusions

Viscoelastic properties of NBR filled with FA are investigated in this work. It can be seen that the addition of FA in NBR causes a somewhat decrease in storage stability, which is believed to be caused by molecular chain scission induced by heavy metals in FA. With the use of Guth-Gold equation, it could be proposed that the presence of pseudo-network (as formed via FA-FA and FA-NBR interactions) associated with the ball bearing effect are responsible for the reinforcement in FA filled NBR compounds.

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