Viscoelastic Properties of Fly Ash/Natural Rubber Compounds: Effect of Fly Ash Loading

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Abstract

Fly ash (FA) as a by-product of power station plants is known to consist of silicon dioxide similar to precipitated silica. The use of FA as filler in natural rubber (NR) was of interest in order to reinforce and/or reduce product cost. In this article, viscoelastic properties of FA filled NR composites with various FA loadings were investigated with the utilization of oscillatory shear flow. It is found that the addition of FA to NR increases storage modulus (G') as a dilution effect. Moreover, there is an unexpected increase in magnitude of viscous response with increasing FA loading. The explanation is proposed in terms of ball-bearing effect of FA with spherical shape associated with the occurrence of molecular degradation induced by inorganic constituents particularly manganese, iron and copper in non-rubber component of NR as well as the small amount of heavy metals including iron, cupper in FA. An isoprene rubber (IR) containing no non-rubber component was used to validate the proposed explanation.

1. Introduction

Silica is widely used as reinforcing filler to improve mechanical properties of vulcanizates, particularly hardness and resistances to tension, tear and abrasion. Although silica is still well accepted among rubber technologists, the use of some other fillers from the natural resources as alternative reinforcing fillers in NR has been carried out to replace silica in rubber compounds [1]. Such fillers include clay, lignin, black rice husk ash (BRHA) and white rice husk ash (WRHA), and cellulose fiber.

FA is a relatively inexpensive by-product of coal-fired power plants, and its usages have the benefit of decreasing environmental problems. Furthermore, FA has been used in industry as a consequence of such advantages as low cost, smooth spherical surface and good processability of the filled materials [2,3].

The present work aims to focus mainly on viscoelastic properties of FA filled NR composites measured from oscillatory rheometers. The viscoelastic properties of NR compounds filled with various FA loadings will be discussed and compared with FA filled synthetic cis-1,4-polyisoprene (IR) compounds containing no natural non-rubber substances.

2. Experimental

2.1 Materials

Raw rubbers used in this work were natural rubber or NR (STR 5L) and a synthetic cis-1,4-polyisoprene (IR). Fly ash (FA) originated from coals typical of those burned in electric power plant (Mae Moa Power Station) was used as a filler.

2.2 Preparation of rubber compounds

Rubber compounds were prepared using a laboratory-sized two roll mill (LRM150, Labtech) at the set temperature of 60 ^oC for 13 minutes. The loading of FA compounds was varied from 0 to 200 phr.

2.3 Characterization of FA

The chemical composition of FA was determined using the X-ray fluorescence spectrometer (XRF) (S4 Pioneer BrukerAXS). A scanning electron microscope (SEM) (LEO 1455 VP) was used to examine the morphology of FA particles.

2.4 Measurement of viscoelastic properties

Viscoelastic properties of compounds under oscillatory shear flow were measured using the Rubber Process Analyzer (RPA2000). The test temperature used for rheometers was of 70 ^oC.

3. Results and discussion

The chemical composition of FA as determined by the XRF is shown in Table 1. It is evident that the largest amount of composition in FA is of silicate (SiO_2) or silica**.** Figure 1 depicts SEM micrograph of FA particles used in the present study. It is evident that FA particles are spherical in shape with a broad size distribution.

Viscoelastic results are exhibited in Figures 2-4. Clearly, G' increases with increasing FA loading which could be due to the hydrodynamic effect and/or filler network formation [4]. As shown in Figure 3, there is an unexpected increase in damping factor of compounds with increasing FA loading which is believed to be the results of a viscous response promotion via molecular slippage particularly surface between FA and NR matrix. Such slippage is caused by: spherical shape of FA (or the ball-bearing effect) (i), and molecular degradation of NR matrix which might be accelerated by the presence of non-rubber component and/or of heavy metal such as iron and copper in FA as illustrated in Table 1 (ii) [5].

Table 1. Chemical compositions of FA used in the present study

Compositions	Content (%)	Compositions	Content (%)
SiO_2 (silica)	37.6	BaO	0.130
Al_2O_3	22.4	SrO	0.123
CaO	15.1	MnO	0.101
Fe_2O_3	14.5	ZrO_2	0.0297
K_2O	2.97	Cr_2O_3	0.0240
SO_3	2.71	Rb_2O	0.0193
Na_2O	1.33	CuO	0.0187
TiO_2	0.447	NiO	0.0121
P_2O_5	0.171	Y_2O_3	0.00431

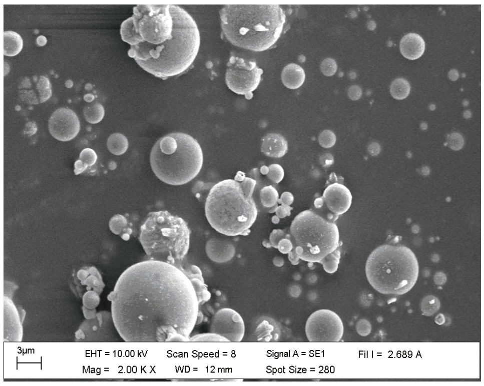

Figure 1 SEM micrograph of FA particles used in the present study

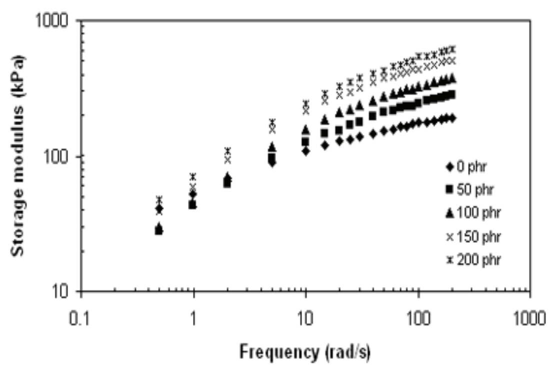

Figure 2 Storage modulus (G′) as a function of frequency at 7 % strain of NR compounds filled with various FA loadings

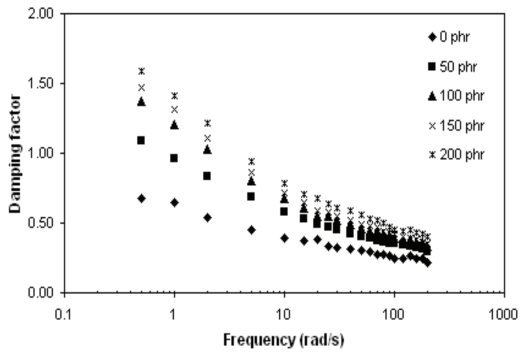

Figure 3 Damping factor as a function of frequency of NR compounds filled with various FA loadings

In the case of IR having no non-rubber substances, Figure 4 shows that, with increasing FA loading, the magnitude of discrepancy in damping factor of compounds with different FA loadings is much less apparent than that of NR compounds (see Figure 3). This implies the lower viscous response in IR compounds than NR compounds at any given FA loading. Thus, it might be summarized that the non-rubber substances presented in NR play role in such increase in magnitude of viscous behavior to some extent.

An existence of small magnitude of change in viscous response observed in IR compounds filled with FA might be due to the molecular slippage via the ball-bearing effect of FA and/or the occurrence of molecular degradation of rubber matrix by a thermo-oxidative process, as mentioned previously.

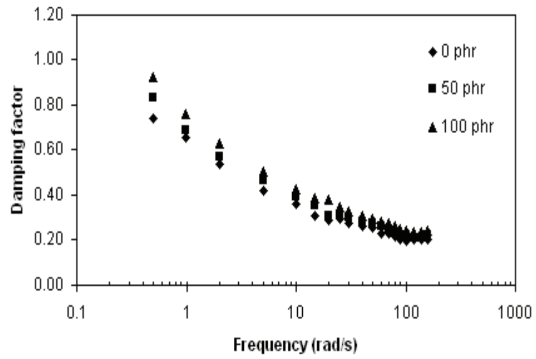

Figure 4. Damping factor as a function of frequency of IR compounds filled with various FA loadings

It is likely that a small amount of heavy metals in FA including iron or copper as shown in Table 1 could act as good oxidizing agents, which are believed to accelerate the degradation of rubber molecules [5].

4. Conclusions

The FA filled NR compounds are investigated in this article, and the following conclusions can be drawn. The addition of FA gives simultaneously increases in G′ and damping factor. The explanation is proposed in terms of ball-bearing effect of FA associated with molecular degradation induced by inorganic constituents particularly manganese, iron and copper in non-rubber component of NR as well as the small amount of heavy metals including iron and copper in FA. The isoprene rubber (IR) containing no non-rubber component was used to validate the explanation.

Acknowledgments

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