Polymer Testing 32 (2013) 38–44

Contents lists available at SciVerse ScienceDirect

Polymer Testing

journal homepage: www.elsevier.com/locate/polytest

Material properties

Synergistic effect of oil palm ash filled natural rubber compound at low filler loading

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A R T I C L E I N F O

Article history:
Received 22 June 2012
Accepted 7 September 2012

Keywords:
Oil palm ash
Natural rubber
Curing characteristics
Tensile properties
Morphological

A B S T R A C T

This work studied the possibility of utilizing oil palm ash (OPA) as filler to reinforce natural rubber (NR) compounds. The OPA loading used in this study ranged from 0 to 9 phr. The properties of low filler loading OPA filled natural rubber were investigated in terms of curing characteristics, tensile properties, rubber-filler interaction and morphology of tensile fractured surface. From the results, it was observed that the scorch and cure times decreased with the addition of OPA whereas maximum torque, tensile modulus (M_{100} and M_{300}) and hardness increased. With the addition of OPA, it was worthwhile to note that the tensile strength and elongation at break improved 16% and 7.4%, respectively, up to an optimum level of 1 phr. Beyond this point, there was a detrimental effect on the strength and flexibility due to agglomeration of hydrophilic OPA. The measurement of rubber-filler interaction by FT-IR and morphological studies of tensile fracture surface of oil palm ash filled natural rubber vulcanizates supported the result obtained from tensile properties.

1. Introduction

Malaysia is the world's largest producer and exporter of palm oil, thus resulting in a millions of tonnes of oil palm waste annually. Presently, the solid waste could be used as alternative fuel for steam generation in oil palm mill plants. Consequently, the by-product known as oil palm ash (OPA) is obtained. It was reported that 4 million tonnes of oil palm ash is produced annually [1] and it was forecast that the ash produced will increase in line with the increasing demand for palm oil. The ash produced during combustion at high temperature (~800 to 1000 °C) is normally disposed in landfill or dumped on an open field, causing severe environmental pollution [2]. Sombatsompop et al. [3] reported that the disposal of ash becomes expensive due to large areas of land needed. In addition, the utilization of ash at zero cost leads to reduction in compounding cost.

Many investigations on utilizing ash as filler in rubber compound had been studied and reported in the literature, such as rice husk ash [4,5], bagasse ash [6], coal fly ash [3], and oil palm ash [7]. It is known that the reinforcement efficiency of filler used in rubber compounding depends on the rubber-filler interaction in terms of particle size, particle dispersion and structure of the filler itself [8]. Foo and Hameed [1] reviewed that the morphology of OPA presented a spongy and porous structure in nature and the presence of pores suggests reinforcement occurred between OPA and rubber matrix. However, previous study [7] showed that the utilization of ash in rubber matrix caused a reduction in properties such as tensile strength, elongation at break, fatigue life and rubber-filler interaction for loading of ash ranging from 10 phr to 40 phr, and this was attributed to agglomeration of hydrophilic OPA.

Thus, the objective of this article is to study the potential of OPA filled natural rubber (NR) compounds at low filler loading. In this present work, the effect of low OPA loading on the curing characteristics, tensile properties, rubber-

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http://dx.doi.org/10.1016/j.polymertesting.2012.09.007

filler interaction and morphology of tensile fractured surface of NR/OPA compounds is reported.

2. Experimental

2.1. Materials

Natural rubber grade SMR L was used as the matrix, which was supplied by the Rubber Research Institute of Malaysia (RRIM). The oil palm ash (OPA) was collected from United Oil Palm Mill, Penang, Malaysia. OPA was sieved with 75 μm mesh sized and dried in a vacuum oven at 80 °C for 24 h to expel moisture. The pH, density and particle size of OPA was 10.13, 2.630 g/cm^3 and 41.09 μm, respectively. The main elemental composition (wt%) of OPA was analyzed by X-Ray fluorescence (XRF) spectrometry (model Rigaku RIX3000) as follows: SiO_2 (34.0), CaO (7.3), Al_2O_3 (3.9), MgO (3.8), K_2O (3.7), P_2O_5 (3.2), Fe_2O_3 (1.9). Other curing ingredients, zinc oxide, stearic acid, N-phenyl-p-phenylene diamine (IPPD), N-cyclohexyl-2-benzothiazole sulfenamide (CBS) and sulfur, were purchased from Bayer (M) Ltd. and used as received.

2.2. Preparation of OPA filled natural rubber compounds

The OPA loading used in this study was 0.5, 1, 3, 7, and 9 phr, and a semi-efficient sulphur vulcanization system was employed. The formulation used is shown in Table 1. Mixing of raw materials was done using a conventional laboratory sized two roll mill (model XK 160). The total mixing time was less than 20 min to avoid premature vulcanization by excess heat generated during compounding. Also, the same mixing sequence was kept constant for all the mixes. The curing characteristics such as scorch time (ts_2), cure time (tc_{90}) and maximum torque (M_H) of OPA filled natural rubber compound were determined by using a Monsanto Moving Die Rheometer (model MDR 2000) according to ISO 3417 at 150 °C, followed by compression molded at 150 °C according to their respective t_{c90} values. Molded sheets were conditioned in desiccators for 24 h prior to further testing.

2.3. Measurement of tensile properties

For tensile tests, five dumbbell shaped specimens were cut from the molded sheet with a thickness of about 2 mm and tensile test were conducted according to ISO 37 using a universal tensile testing machine (model Instron 3366) at a crosshead speed of 500 mm/min. Tensile strength, elongation at break and tensile modulus (M_{100} and M_{300}) data were evaluated from the stress-strain determinations, and average values from five repeated tests for each compound were recorded. Hardness was measured using a Shore A type durometer with the test conditions being in accordance with ISO 7619.

Table 1
Formulation of oil palm ash (OPA) filled natural rubber compounds.

Materials	Compound (phr[a])
NR (SMR L)	100
Zinc oxide	1.5
Stearic acid	1.5
IPPD	2.0
CBS	1.9
Sulfur	1.6
OPA	0, 0.5, 1, 3, 7, 9

[a] Parts per hundred parts of rubber.

2.4. Measurement of swelling and rubber-filler interaction

The swelling test was carried out according to ISO 1817. The cured specimens with dimensions of 30 mm × 5 mm × 2 mm were weighed using an electrical balance, followed by immersion in toluene for 72 h at room temperature (25 °C) in a dark environment. After the conditioned period, the swollen specimens were taken out and weighed again. The specimens then were dried in an oven at 70 °C until constant weight was obtained. The Lorenz and Park equation [9] was applied in order to study the rubber-filler interaction, the weight of toluene uptake per gram of rubber hydrocarbon (Q) was calculated as given in Equations (1) and (2) below:-

$$\frac{Q_f}{Q_g} = ae^{-z} + b \quad (1)$$

$$Q = \frac{Swollen\ weight\ (W_s) - Dried\ weight\ (W_d)}{Initial\ weight\ (W_i) - (100/Formula\ weight)} \quad (2)$$

where subscript *f* and *g* refer to filled and gum vulcanizates, respectively. z is the ratio by weight of the filler to the rubber hydrocarbon in the vulcanizates, whereas a and b are constants. The lower the Q_f/Q_g values, the higher the extent of rubber-filler interaction.

2.5. Fourier transform infra-red (FT-IR) spectroscopy

FTIR was used to obtain some qualitative information about the functional groups and chemical characteristics of the OPA, and possible interaction between OPA and natural rubber. FTIR spectra were obtained and recorded using a Perkin Elmer Spectrometer in the range of 550 cm^{-1} to 4000 cm^{-1} at 4 cm^{-1}. For each spectrum, 16 scans were co-added. For OPA, potassium bromide (KBr) and OPA were ground to form a pellet that was used to obtain the infrared spectra of OPA where KBr is an inert, infrared transparent material which acts as a support and diluent for the OPA.

2.6. Scanning electron microscopy

Besides observing filler morphology, tensile fractured surfaces of OPA filled natural rubber compound were scanned with a scanning electron microscope (FESEM: model Zeiss Supra 35 VP). The fractured specimens were mounted on aluminium stubs and sputter-coated with a thin layer of gold-palladium in order to avoid electrostatic charge and poor resolution during examination. The SEM