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Butadiene acrylonitrile rubber loaded fast extrusion furnace black as a compressive strain and pressure sensors

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Abstract

Electrically conductive acrylonitrile butadiene rubber (NBR) compounds filled with different concentrations of fast extrusion furnace black (FEF) were experimentally investigated. The percolation concentration of the investigated composites was found to be 65 phr. Of all composites examined, sample N70, which apparently belonged to the region of percolation phase transition, was found most sensitive to compressive strain. The electrical conductivity of this sample was changed by more than 50% upon a 16% compression. Sample N70 showed a good reproducibility after preconditioning rather than before preconditioning. The piezo-resistive effects practically are thermally stable within the interval of 297–308 K. The working range of load pressure, for sample N70, from 5 to 25 kg and the working voltage from 30 to 150 V.

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1. Introduction

Conductive rubbers are widely used instead of metallic conductors because they posses the obvious advantage of flexibility and ability to absorb mechanical shocks [1]. In the last few years the black filled conductive rubbers have wide applications, such as electromagnetic shielding materials [2], and sensors for measurement of vehicle weights to collect toll tax on roads [3]. The choice of rubber and filler type is basically related to this program, where in fact it is already recognized that carbon black plays a dominant role in determining the electrical and mechanical properties of polymer-based composites [4]. For this reason fast extrusion furnace (FEF) carbon black is used in the present work because it has a large particle size particle diameter and low tendency to form aggregates and in turn very low hysteresis compared with the other types of carbon black [5].

Butadiene acrylonitrile rubber (NBR) is also used as a matrix because it has good electrical properties rather than other types of rubber, where it has a polar $C \equiv N$ groups, and also due to its thermal stability up to $80 \,^{\circ}$ C [6].

2. Experimental work

2.1. Materials and processing

Acrylonitrile butadiene rubber (NBR) (density 0.98 g/cm³ and acrylonitrile content 26%) was used as polymer matrix. Fast extrusion furnace (FEF) carbon black (38 nm particle size diameter; dibutyl phthalate absorption number (DBPA) 121 cm³/100 g; 45 m²/g surface area) was used as reinforcing filler. Other compounding ingredients like zinc oxide and stearic acid (activators), Dibenz thiazyl disulphide (MBTS) semi-ultra accelerator (vulcanization time 30 min at temperature 150 °C), phenyl-β-naphthyl-amine (PBN) antioxidant (melting point 105 °C), dioctyle phthalate (DOP) plasticizer and sulfur (vulcanizing agent) were used. These materials were supplied by Bayer Company (USA). These materials were compounded according to the recipe listed in Table 1. For the compounding a home made two-roll mixing mill (length 0.3 m, radius 0.15 m, speed of slow roll 18 rpm and gear ratio 1.4) was used. The mixing occurred for 40 min at a temperature of 25 °C. The compounded rubbers were compression molded into cylinders of 1×10^{-4} m² area and 0.01 m in height. The vulcanization were conducted under a heating press (KARL KOLB, Germany) at a pressure of $P = 0.40 \,\mathrm{MPa}$. The optimum conditions of temperature and time were T = 150 °C and t = 30 min. The vulcanized

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Table 1
Ingredients of the investigated NBR rubber composites

	Sample ingredients (phr) ^a									
	N10	N20	N30	N40	N50	N60	N70	N80	N90	N100
NBR	100	100	100	100	100	100	100	100	100	100
Stearic acid	2	2	2	2	2	2	2	2	2	2
Zinc oxide	5	5	5	5	5	5	5	5	5	5
FEF	10	20	30	40	50	60	70	80	90	100
DOP	10	10	10	10	10	10	10	10	10	10
MBTS	2	2	2	2	2	2	2	2	2	2
PBN	1	1	1	1	1	1	1	1	1	1
Sulfur	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5

^a Part per 100 parts of rubber by weight.

samples were shelf aged for 48 h before test. The mixing time and vulcanization conditions were fixed for all samples.

2.2. Measurements

Brass electrodes were attached to the parallel faces of the samples during vulcanization. The samples are subjected to compressive strain and pressure by using a material tester machine (AMETEK, USA). A digital force gauge (Hunter Spring ACCU Force II, 0.01N Resolution, USA) connected to a microprocessor was used to measure compression force. The machine has the properties of varying both amplitude and frequency. In electrical measurements a digital electrometer (616 Keithly, USA) and RLC bridge (3531 Z-Hitester, Hioki, Japan).

3. Results and discussions

3.1. Effect of FEF black volume fraction on electrical conductivity

The percolation threshold is a basic characteristic of a conductive composite; in which the percolation threshold defines the composition range for studying the effect of compression on conductivity (σ). Fig. 1 shows the variation of conductivity with carbon black volume fraction. A typical S-shaped curve is observed that separates three regions: insulating, transition, and conductive. The model that is most often used to quantify the changes in the transition and conductive regions is the so-called statistical percolation model [7]. Proposed by Kirkpatrick [8] and Zallen [9], this model predicts the electrical conductivity of an insulator–conductor binary mixture by assuming random positions of the filler particles. The result is a power-law variation of the conductivity (σ):

$$\sigma = \sigma_0 \left(\frac{\phi - \phi_c}{1 - \phi_c} \right)^t$$

where σ_0 is the conductivity of unfilled rubber, ϕ the volume fraction of filler, ϕ_c percolation threshold and t is a universal exponent that is close to 2 for a 3D dispersion [10]. The two parameters fit are represented in Fig. 1 by the solid line and gives $\phi_c = 0.65$ and t = 2.03. The value of the exponent t is consistent with the model prediction.

3.2. Electrical properties under uniaxial compression

The relative conductivity $[\sigma(\varepsilon)/\sigma(0)]$, where ε is the compressive strain, as a function of compressive strain for all rubber samples is displayed in Fig. 2. This figure indicates that for samples below the percolation concentration, the conductivity increases with compressive strain up to 8%. For samples above the percolation concentration, the conductivity decreases with compressive strain up to 16%. It is also found that sample N70 is the most sensitive to the variation of compressive strain.

One may explain such behavior as follow; since the compression of the composite sample causes its deformation the intrinsic structure of the material changes. In other words when a compressive strain acts on polymer composites that is filled with conducting particles the change in their electric conductivity during deformation results from particle displacement which affects the conducting paths in the material. When the filler content is below the percolation threshold and conducting particles are separated by an insulating layer of the polymer matrix, the electric current can flow between neighboring particles only by

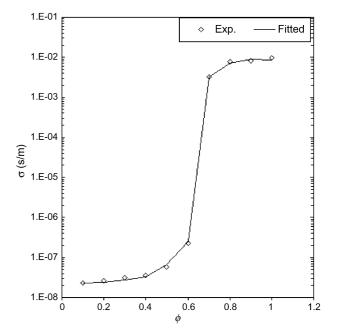


Fig. 1. The conductivity (σ) vs. carbon black volume fraction (ϕ) .

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