

## Magnetic and dynamic mechanical properties of barium ferrite–natural rubber composites

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### Abstract

High coercivity barium ferrite ( $\text{BaFe}_{12}\text{O}_{19}$ ) powders have been prepared by a coprecipitation method. After removing fine powders, they were incorporated into a natural rubber matrix with different loading levels up to 120 phr (part per hundred rubber). The variations in characteristics of rubber–ferrite composites (RFCs) were evaluated as a function of ferrite loading. The results showed that the coercivity is improved and the saturation magnetization is linearly dependent on the mass fraction of the filler, while the tensile strength, strain at break and modules are highly influenced by the size, shape and volume fraction of ferrite particles. The present RFCs showed a typical characteristic of low density ( $2.2 \text{ g/cm}^3$ ), high stored energy ( $\text{BH}_{\text{max}} = 1.18 \text{ MG Oe}$ ) and flexibility (resilience = 0.844) at 120 phr of ferrite, which are relatively high when compared to previous works.

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### 1. Introduction

The advantages of polymer bonded magnets (PBMs) over their metallic and ceramic counterparts include low weight and cost, resistance to corrosion, ease of machining and forming, and capability of high production rates [1]. Each materials has unique properties that make it more suitable for selected applications than other magnet and mechanical option such as, magnetic strength, resistance to demagnetization, density, physical strength, and flexibility [2]. The impregnation of magnetic fillers in the matrix imparts magnetic properties and modifies the physical properties of the matrix considerably. The mechanical properties of PBMs depend strongly on properties of polymer matrix, magnetic fillers, and interfacial condition between the components [3]. Also

the surface area of the filler is the primary factor to control the mean distance between particles. The higher the surface area, the shorter the interaggregate distance would be [4].

Ferrites remain one of the best magnetic materials ever discovered and cannot be easily replaced by any other magnetic material because they are inexpensive, stable and have a wide range of technological applications [5,6]. So they are still widely used wherever the product cost is a major consideration over magnetic performance although they have less magnetic strength than rare earth magnets. Recently strontium ferrite ( $\text{SrFe}_{12}\text{O}_{19}$ ) has received a wide attention as a permanent magnet than barium ferrite ( $\text{BaFe}_{12}\text{O}_{19}$ ) although they are relatively closed in magnetic properties [7], and in addition the latter has a high chemical stability and low price than the former. The consideration that  $\text{SrFe}_{12}\text{O}_{19}$  is superior to  $\text{BaFe}_{12}\text{O}_{19}$  may be owing to the accompaniment of little successes for the production of barium ferrite powders in a good form [8].

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Table 1  
Formulation of the mixes for preparing RFCs

Ingredient	Phr (part per hundred rubber)
Rubber (ADS1-XI)	100
Stearic acid	1
Zinc oxide	5
Accelerators (CBS)	0.6
Antioxidant (AW)	1
Sulfur	1.5
Ferrite	10 30 60 90 120

CBS: *N*-cyclohexyle benzothiazyl sulphenamide; AW: 6-ethoxy-1,2-dihydro-2,2,4-trimethylquinoline.

In this work an attempt has been made to prepare barium ferrite powders suitable for magnetic application in the form of a barium ferrite–natural rubber composite by a simple method. Our aim is to produce a low weight, low cost, highly flexible bonded magnet with a suitable stored energy, which is one of the most important magnetic properties of the composite. This will be achieved by selecting the recipe and high coercivity fillers with a low surface energy. We report in this article some magnetic and mechanical properties of RFCs prepared from coprecipitated barium ferrite powders with a selected size distribution and natural rubber.

## 2. Experimental procedure

Barium ferrite powders having a coercive force 5.2 kOe, magnetization 67 emu/g and 45–200  $\mu\text{m}$  particle size were prepared by a coprecipitation method. Detailed procedures were reported elsewhere [8]. After pre-characterization they were mixed with a natural rubber ADS (air dried sheet) by various loadings up to 120 phr to form rubber–ferrite composites (RFCs). The recipe used for this study is given in Table 1. The composites were prepared in a two-roll mixing mill and after homogenization they were cured and molded into thin sheets of 1–2.5 mm in thickness at 150 °C using a hydraulic press according to ASTM D-15. Scanning electron micrographs (SEM) for RFCs were obtained from a fractured and polished surface under liquid nitrogen. The resulting surface was coated with carbon prior to the SEM investigation.

Magnetic measurements for the ferrite powders and RFCs were carried out in a SQUED magnetometer up to 50 kOe at room temperature. Tensile stress and shore hardness were measured according to ASTM D-412 and ASTM D-2240, respectively. The variation of storage modulus, loss modulus and damping factor for RFCs were studied at 10 Hz using dynamic mechanical analyzer (DMS-2000).

## 3. Results and discussion

### 3.1. Magnetic properties of rubber–ferrite composites

Hexagonal barium ferrite powders were prepared by a coprecipitation method and before mixing with rubber fine powders less than 45  $\mu\text{m}$  were removed by sieving so as to disaggregate the powders. Fig. 1 shows the microstructures for ferrite powders in a free state (a) and RFCs with the maximum ferrite loading 120 phr (b). In Fig. 1(a) it is clear that the ferrite particles have smooth surfaces and rather irregular shape. Fig. 2 shows the hysteresis loops of the ferrites before and after removing fine powders less than 45  $\mu\text{m}$ . The final powders recorded 67 emu/g saturation magnetization and 5.2 kOe coercive force, which can be attributed to the increase in the porosity of powders. Fig. 3 shows the variation of the magnetic properties for RFCs as a function of ferrite loading. It can be observed that the magnetization increases with increasing ferrite content. From these curves the values of saturation magnetization ( $M_s$ ) and coercivity ( $H_c$ ) for the samples with different loading were determined and plotted in Fig. 4 against the mass fraction of the filler. The figure indicates that  $H_c$  is almost independent of the mass fraction at lower ferrite loading in contrast with the previous works [9–12], and that only the major effect of loading is to increase the remanence magnetization. The saturation in  $H_c$  may be resulting from (1) the desegregation of the filler caused by removing the fine powders prior to mixing, (2) the high degree of mixing which is a critical factor in controlling the magnetic and mechanical properties of polymers bonded magnets [13], and (3) the fact that the present magnetic particles as a filler are relatively large in size, randomly oriented, uniformly dispersed

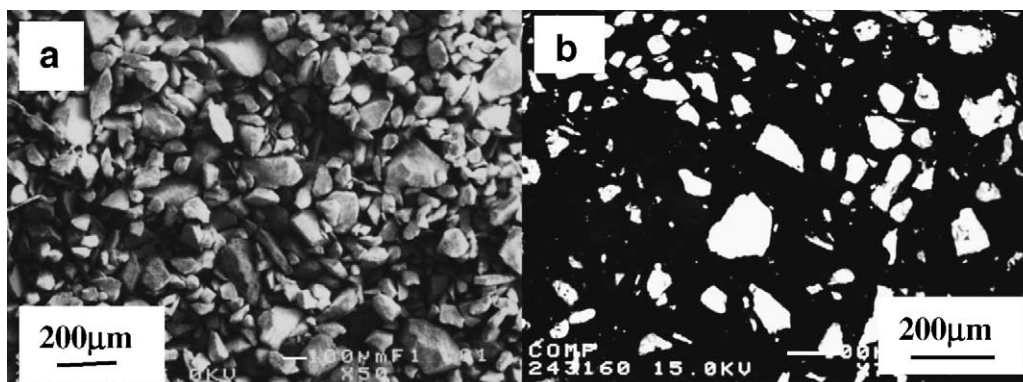


Fig. 1. Scanning electron micrographs for (a) ferrite powders, (b) RFC at 120 phr ferrite loading.

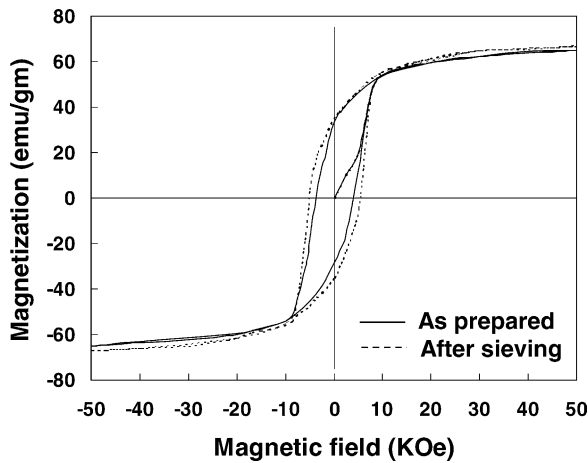


Fig. 2. Hysteresis loops for as prepared and sieved barium ferrite powders.

and isolated from each other as seen in Fig. 1(b). The letter will lead to reduce the surface energy of the particles, which control the mean distance between particles during a mixing process. So there is no tendency to form agglomerates in the RFCs even at a relatively high loading level. At a critical loading about 100 phr the coercivity starts to decrease slightly due to the increasing of magnetic interaction between the ferrite particles as the mean interparticle distance decreases.

The specific saturation magnetization,  $M_{\text{rfc}}$ , of RFCs is found to be linearly dependent on the mass fraction of ferrite and to obey a general relation as seen in Fig. 4:

$$M_{\text{rfc}} = M_f W_f,$$

where  $M_f$  and  $W_f$  are the saturation magnetization and weight fraction of the filler, respectively.

As shown in Fig. 5 both of the maximum energy product  $(BH)_{\text{max}}$  and the density of RFCs increase linearly with increasing ferrite loading. The recorded  $(BH)_{\text{max}}$  (1.18 MG Oe) for our bonded magnet with a density of 2.2 g/cm<sup>3</sup> is higher than those given in literatures [9,11] due to the good selection

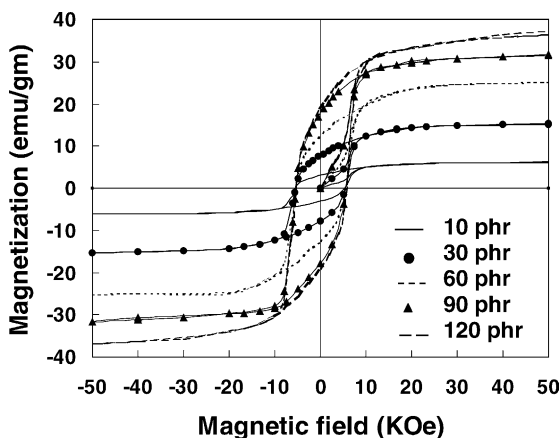


Fig. 3. Hysteresis loops for RFCs as a function of ferrite loading.

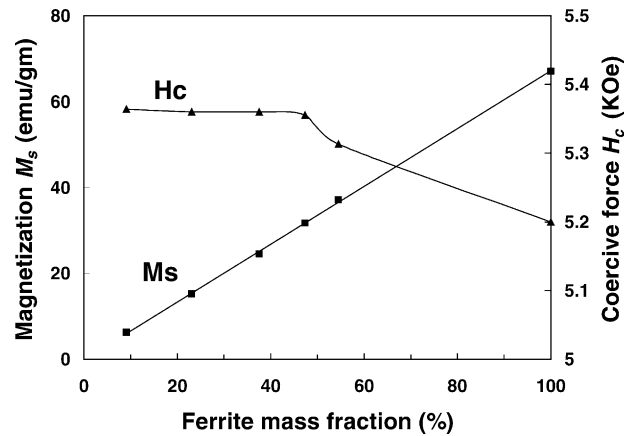


Fig. 4. The variation of specific saturation magnetization ( $M_s$ ) and coercive force ( $H_c$ ) as a function of the mass fraction of ferrite filler.

of the magnetic powders and the composites characteristics as mentioned above.

### 3.2. Mechanical and dynamic properties

The mechanical properties of PBMs are strongly influenced by the size, shape, type, loading and dispersion of the magnetic filler, as well as the matrix properties and the interfacial adhesion between the filler and the polymer matrix. In the present work, the filler particles are relatively large in size (45–200  $\mu\text{m}$ ) and having a granular shape and untreated smooth surfaces. Therefore, it is expected that the filler has low wettability and adhesion with the rubber matrix [14].

The variation of tensile strength and elongation at break with filler loading in RFCs are presented in Fig. 6. The tensile strength of these composites decreases with increasing filler loading. This is because the interaction between the ferrite and rubber matrix is weak and the influence of the interaction increases by increasing filler loading. The elongation at break also recorded the same behavior.

Fig. 7 shows the change in the real ( $E'$ ) and imaginary ( $E''$ ) part of storage modules against the filler fraction. The modules values were found to decrease after attaining maximum values and again increase at higher filler loading. In contrast the values of the damping factor  $\delta$  and shore hardness tend to increase with increasing filler content as shown in Fig. 8. The attenuation in modules is an evidence of lower interaction between filler–filler and filler–rubber matrix.

There are numerous cases where the fillers increase the damping and consequently decrease the modulus [15]; (1) particle–particle friction where particles touch each one another as in weak agglomerate, (2) particle–polymer friction where there is essentially no adhesion at the interface, and (3) excess damping in the polymer near the interface because of the induced thermal stress or changes in polymer confirmation or morphology. According to Fig. 1(b) the particles in our RFCs are isolated from each other so that the attenuation in the modulus may be resulted from the particle–polymer friction. At a low concentration of the filler there is a little

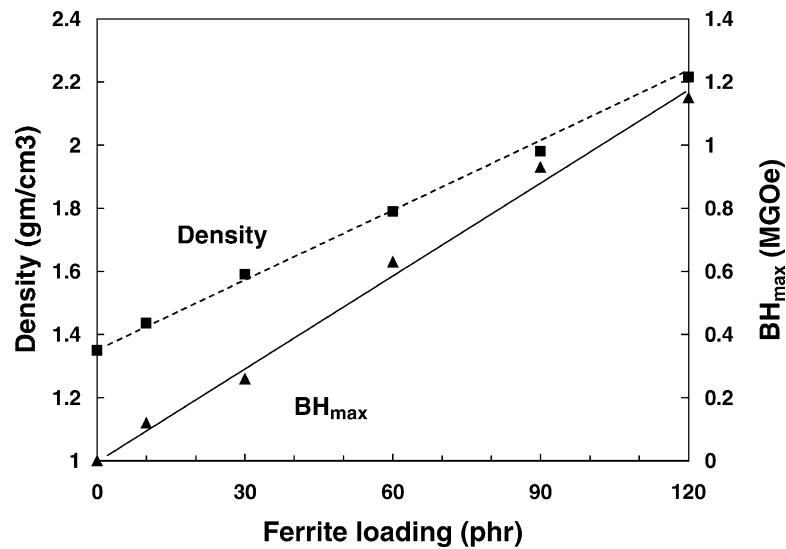


Fig. 5. Variation of the maximum energy product (BH)<sub>max</sub> and composite density with ferrite loading.

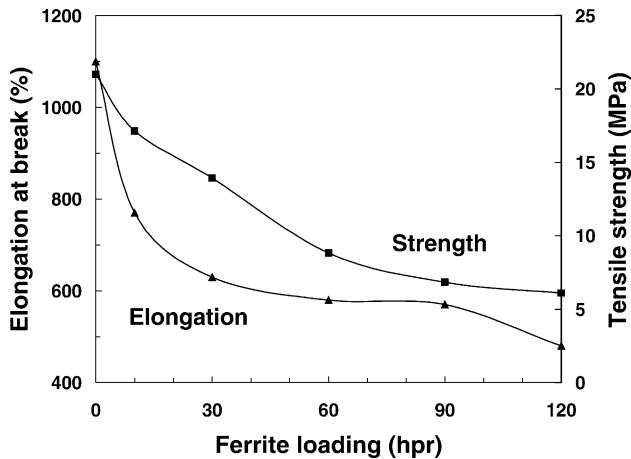


Fig. 6. Variation of tensile and elongation at break as a function of ferrite loading.

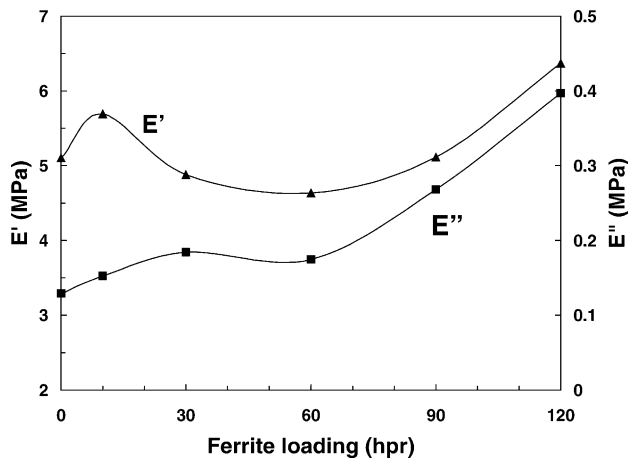


Fig. 7. The variation of storage ( $E'$ ) and loss ( $E''$ ) modulus with ferrite loading.

friction and the modulus increases due to the incorporated rigid particles in the rubber matrix. By increasing the filler content the polymer–particle friction and their relative slipping will increase causing the increase of damping factor and decrease in modules. It is also expected that due to the particle morphology, thermal energy released from the friction and slipping increase the damping in the polymer near the interface. By increasing the filler content furthermore the composite hardness reaches a high level and the particle–particle interaction increases as mentioned before in the last section. Consequently the modulus starts to increase again with increasing filler content.

The composite resilience,  $R$ , was estimated according to the formula [15]:

$$R = \exp(-p \tan \delta),$$

where  $\delta$  is the damping factor. The variation of composite resilience with ferrite loading is indicated in Fig. 9. Clearly

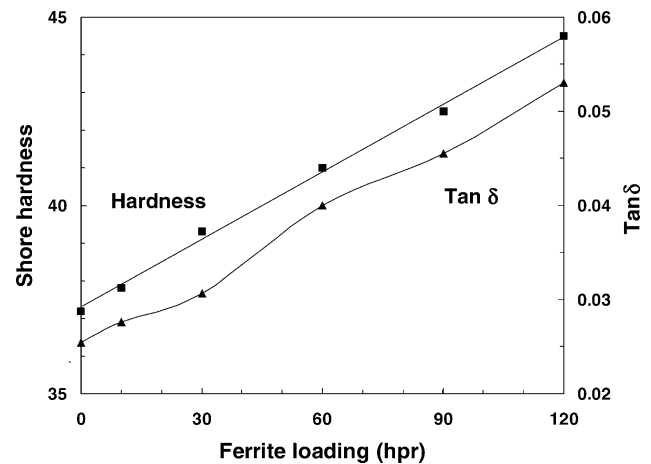


Fig. 8. The variation of shore hardness and  $\tan \delta$  with ferrite loading.

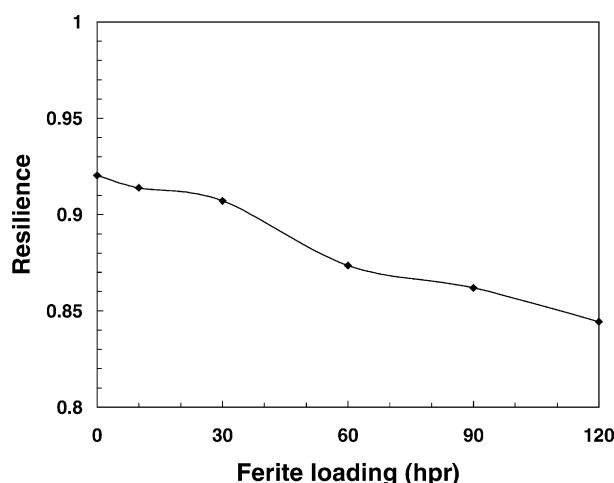


Fig. 9. The variation of resilience with ferrite loading.

Table 2

Rubber–ferrite bonded magnet parameters at the maximum loading of ferrite (120 phr) in this work

Parameters	Value
Coercive force (kOe)	5.18
Remnant magnetization (G)	528
Saturation magnetization (G)	1173
Maximum energy (MG Oe)	1.18
Density (g/cm <sup>3</sup> )	2.215
Tensile strength (MPa)	6
Hardness	44.5
Resilience	0.844

we see the resilience decreases with increasing ferrite loading and the order of change is parallel to that of the modules. The recorded values of resilience are still high when compared to similar works [9,11].

For comparison with the results in previous works the final characteristics of our RFCs with a ferrite loading of 120 phr (about 54% mass fractions) are given in Table 2. It is apparent that the present values are high for the magnetic and reasonable for the mechanical properties if we compare to literatures.

#### 4. Conclusion

Barium ferrite powders having the particle size in a range 45–200  $\mu\text{m}$  were incorporated into a natural rubber with different loading levels up to 120 phr to obtain RFCs. By selecting disaggregated, high coercivity and low surface energy powders, we successfully produced the RFCs having a rather low density, high coercivity, large stored energy and high flexibility even at relatively low mass fraction of the filler. The dynamic properties are more strongly dependent on the particles and particle–matrix characteristics than the magnetic and the mechanical properties. For further improve-

ment detailed future works are needed to study the effects of size, shape and surface roughness of fillers on the magnetic and dynamic properties of barium ferrite–rubber composites.

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