



Porous magnetic materials based on EPDM rubber filled with carbonyl iron particles

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ARTICLE INFO

Keywords:

Magnetorheology
Porous magnetic materials
Smart materials
Sponge-like structure
Microstructure

ABSTRACT

This paper deals with a possibility to prepare porous magnetic elastomers whose mechanical behavior can be controlled by means of a magnetic field. The elastomers under investigation are based on ethylene propylene diene rubber (EPDM) filled with carbonyl iron particles. The foaming agent based on azodicarbonamide was used at 2 loading levels to control their porous character and the magneto-mechanical behavior was compared with non-porous magnetic EPDM system. The non-porous systems exhibited the highest magnetorheological efficiency even though the porous systems showed lower values of storage modulus in the inactive state. Porous systems also exhibited lower mechanical hysteresis when compared to non-porous one during single-strain measurements with increasing/decreasing magnetic field due to pores which increase a distance between particles and thus lower the strength of magnetic network created at high magnetic fields. Porous systems can be potentially utilized in applications in which conventional magnetorheological elastomers are too tough, thus, porous systems offer magnetorheological material with properties between magnetorheological fluids and solid elastomers.

1. Introduction

Magnetorheological (MR) systems are magnetic-responsive materials changing their rheological properties in a controlled way according to an applied external magnetic field. The MR systems are usually two phase structures of magnetic particles dispersed in non-magnetic medium. When a magnetic field is applied the magnetic particles are polarized and tend to join themselves along with a direction of the applied magnetic field [1], which leads to an increase in the rheological parameters of the systems [2–5]. Magnetorheological systems are referred to MR suspensions, gels, elastomers or foams depending on the matrix in which magnetic particles are dispersed. Among these, MR suspensions exhibit the highest difference between the rheological parameters in the presence of a magnetic field and in its absence since the position of the particles is not fixed in the matrix and the particles can create strong chain-like internal structures in the active state when the magnetic field is applied [4]. On the other hand, in the case of MR elastomers the polymer matrix is very tough resulting in not as significant increase in the rheological parameters as in the case of MR suspensions in the active state [6–8]. When elastomeric matrix with magnetic particles fixed inside, isotropic (magnetic particles are uniformly dispersed) or anisotropic (magnetic particles are aligned in the direction of magnetic field applied during curing process) magnetic

composites can be prepared. These systems then exhibit significantly different mechanical properties [9,10], thermal conductivity or radio-absorbing properties [11]. Porous MR elastomers (pMRE) can further provide magnetic-responsive material with the matrix toughness between MR suspensions and MREs, and enable to suit the properties of the system for intended applications. The pMREs can be involved in applications similar to MREs as dampers, vibration absorbers, actuators, and various cushions [4,11–15].

Magnetorheological foams with conventional MR suspensions entrained in either polymeric [16–18] or metallic foam [15,19] structure have been introduced. The MR suspensions are here filled in an absorbent matrix, thus, the systems are prepared separately, and the sponge-like materials are consequently immersed in such MR suspensions. Another approach how to involved foamed materials in MR systems was to introduce porous polystyrene onto the surface of carbonyl iron leading to outstanding stability of the system together with its sufficient MR performance [20]. However, there is only limited number of studies dealing with entrapping magnetic particles directly in the polymeric matrix and creating pMRE [12,21–23]. So far, the polymeric matrix from polyurethanes (PU) has been used for these systems due to their easy preparation from diisocyanates and polyols, where the pores are produced by reaction with a small amount of water. It was shown that mechanical behavior of pMREs can be controlled

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through application of an external magnetic field like in the case of MREs. The difference in mechanical behavior of isotropic, i.e. magnetic particles are distributed within the matrix randomly, and anisotropic, i.e. magnetic particles are distributed in a certain level and direction as the external magnetic field was applied during the matrix curing, pMRE under compression has also been investigated [21,22]. The particle content further significantly influences thermal stability [12,21] and pore size distribution in magnetic PU foams [23]. To conclude, MREs have recently attracted large scientific interest while pMREs have been studied rarely although their MR performance can fill the application space between MR suspensions and MREs. Hence, pMREs based on ethylene propylene diene rubber (EPDM) were prepared and their MR performance was compared to non-porous MR based on the same matrix in this study.

2. Experimental part

2.1. Materials

Carbonyl iron (CI) particles (grade CN, BASF company) with diameter $d_{50} = 6.5\text{--}8\text{ }\mu\text{m}$ (guaranteed by the supplier) were used as a magnetic filler in prepared MR systems. The various substances such as EPDM rubber, stearic acid, zinc oxide, polyethylene wax, MBTS accelerator (benzothiazyl disulfide), paraffin oil, and sulfur were obtained from MITAS a.s., the Czech Republic. A foaming agent based on activated azodicarbonamide TRACEL K 5/95 was supplied by Tramaco.

2.2. Preparation of MR systems

The composition of prepared pMREs based on EPDM (pEPDM) is summarized in Table 1. Briefly, the EPDM rubber was mixed using a roller mill (LabTech Engineering; Compuplast, LRMR-S-200/W; Czech Republic) at $70\text{ }^{\circ}\text{C}$ together with necessary substances such as stearic acid, zinc oxide, polyethylene wax, MRTS accelerator, paraffin oil, sulfur, and foaming agent in the case of pMREs. The prepared compound was left to relax for 24 h at $5\text{ }^{\circ}\text{C}$, and then, the CI was added using an internal mixer (Plastograph; Brabender W50 EHT PL, Germany) operating at $70\text{ }^{\circ}\text{C}$, 50 rpm for 5 min. The compound was then transformed to plate-like material using the roller mill, and the sample was then molded using a hydraulic press (FONTIJNE LabEcon 300, The Netherlands) operating at temperature $160\text{ }^{\circ}\text{C}$ and the pressure 150 kN. It should be mentioned that as-prepared MR systems were isotropic solid with uniform distribution of magnetic particles within the EPDM matrix. According to a different amount of the foaming agent loading (0, 4, 6 phr) and CI loading (150 phr, 250 phr), the samples are further labeled as pEPDM-0-150, where the numbers separated with a dash reflect amount of the foaming agent and loading of the particles, respectively. As a reference material, MRE prepared from EPDM rubber was prepared under the same conditions without the foaming agent.

Table 1
Composition of prepared pEPDM systems in parts per hundred rubber (phr).

Substance	phr
EPDM rubber	100
Stearic acid	3
Zinc oxide	5
Polyethylene wax	1
MRTS accelerator	0.6
Foaming agent	0/4/6
Paraffin oil	10
Sulfur	2.5
Carbonyl iron	150/250

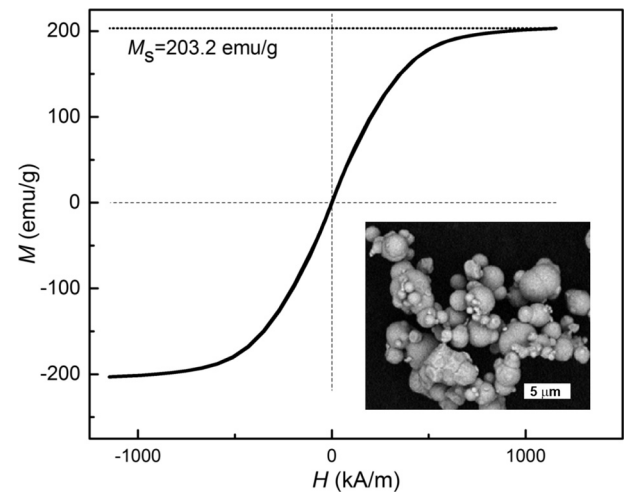


Fig. 1. Magnetization curve of used CI particles. The inset depicts their SEM image.

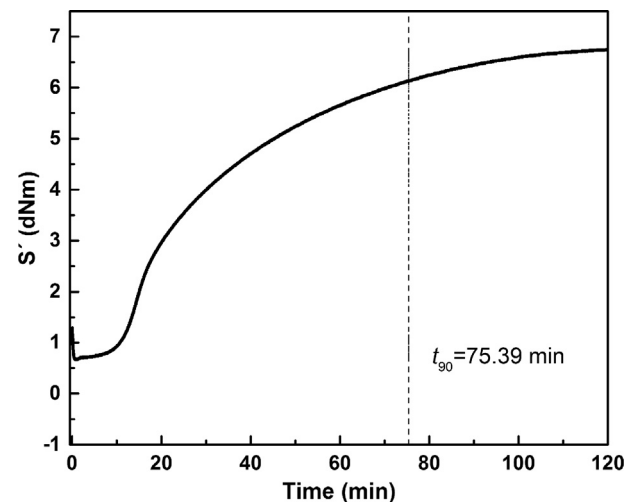


Fig. 2. Curing curve for used EPDM rubber at $160\text{ }^{\circ}\text{C}$ (quantity S' stands for the real part of torque).

2.3. Particle, MREs, and pEPDMs characterization

Scanning electron microscopy (SEM; Phenom Pro, Phenom-World, The Netherlands) was used to investigate the size and morphology of the magnetic particles as well to evaluate the structure of prepared pEPDM. A vibrational sample magnetometer (VSM; Model 7404, Lake Shore, USA) was used to evaluate magnetic properties of the used CI particles. Curing characteristics were determined using a rotorless curemeter MDR 3000 Basic (MonTech, Germany) at $160\text{ }^{\circ}\text{C}$ and 7% strain with frequency 1.667 Hz according to ISO 6502. The curing characteristics were determined only for system EPDM-0-150 as an orientation curing time, which was further used for all prepared systems.

2.4. Magnetorheological measurements

Rheological measurements of prepared MREs and pEPDMs in the absence and in the presence of a magnetic field were performed using a rotational rheometer (MCR 502 Anton Paar, Austria) with a plate-plate geometry and an external magnetic cell at $25\text{ }^{\circ}\text{C}$. Each measurement was done with defined normal force of 2 N. The utilization of foaming agent led to samples with different thickness (EPDM-0-150 1.55 mm, EPDM-0-250 1.62, pEPDM-4-150 3.06, pEPDM-4-250 2.89 mm, pEPDM-6-150 3.65 mm, pEPDM-6-250 3.47 mm) and the preload

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