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Note

The enhanced magnetodeformational effect in Galfenol/polyurethane nanocomposites by the arrangement of particle chains

Tatiana Kiseleva [a,*], Sergey Zholudev [a,b], Alla Novakova [a], Tatiana Grigoryeva [c]

[a] *Moscow M.V.Lomonosov State University, Department of Physics, Moscow, Russia*
[b] *Technological Institute for Superhard and Carbon Materials, Troitsk, Russia*
[c] *Institute of solid state chemistry and mechanochemistry RAS, Novosibirsk, Russia*

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ABSTRACT

The article focuses on the properties of the newly manufactured metal-polymer composites containing spatially oriented chains of Galfenol anisotropic particles in the polyurethane matrix. The magnetic and mechanical properties of the metal-polymer composite have been subject to experimental study that has shown the non-monotonic increase of the composite magnetodeformational response with particles concentration rise in polymer matrix. The explanation of this phenomenon was given on the basis of structural investigation of metallic particles chains direction and their density in the sample by means of TEM, SEM and 3D X-ray computed tomography. The maximum value of functional magnetodeformational characteristics has been obtained for the composition with 15% particles volume fraction with preferable spatial orientation in longest particle chains.

1. Introduction

Metall-polymer composites with magnetic particles dispersed in a polymer matrix represent a system with physical and chemical properties that are influenced by the powder fillers and polymer matrix characteristics as well as by the interaction at the particle-polymer interface [1–3]. These materials are considered as smart ones because their properties may be controlled by external stimuli such as magnetic fields, stresses, electric field, and temperature [4]. Furthermore, the polymer matrix enables the formation of practically any shapes of the material providing the correct transmission of load onto the reinforcing phase in the composite material. Fe-containing ferromagnetic particles are the most popular fillers for polymer composites for creating the magnetic sensors, actuators and dampers [5].

The ability to use the advantage of particular properties reinforcing metal and particular elastic properties of the polymer matrix is the most important motivation for the development of metal-polymer functional hybrids.

The mechanical performance of thermoplastic polyurethanes as composite counterparts has been demonstrated in a number of papers [6–10]. Very interesting results have been obtained when filler particles were made from magnetostrictive milled Terfenol-D alloy ($Fe_xTb_yD_{1-y}$ $x = 0.27–0.3$, $y = 1.9–2$ compositions), which allowed to apply the particular properties of the filler material [8–10]. The study has shown that there is a direct connection between magnetic properties, structural characteristics and distribution of particles sizes in polymer. These composites demonstrated considerable magnetodeformational effects (effects of dimension changes under the influence of magnetic field). The sophisticated synthesis of exact magnetostrictive composition in the Fe–Tb–Dy system makes one search for simpler magnetostrictive materials for these purposes. One of the promising magnetostrictive system is Fe–Ga (Galfenol) with composition close to Fe–Ga (18 at%) [11,12]. Amplification of the desired magnetostrictive effect in particles can be achieved if its phase structure has preferable magnetic direction, and by shape anisotropy of the particles as well. Magnetodeformational effect of the particles/polymer composite system can be favored by the successive particles orientation in the polymer matrix. Reducing the filler particle sizes results in a surface to volume ratio rise and changes the strength of the particle surface interaction activity to polymer molecules. But the magnetodeformational effect in metal-polymer composite filled by very small particles is not obvious. When constructing functional hybrid materials it has been recently advised to consider mechanochemical technique aimed at synthesising FeGa-particles with magnetostrictive composition [13,14]. Mechanical alloying

* Corresponding author. Fax: +7 (495)9328820.
E-mail address: Kiseleva.TYu@gmail.com (T. Kiseleva).

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of powdered metals produced a magnetostrictive nanocrystalline α-Fe (Ga)/Fe_3Ga phase composition of the particles with sizes ranging from several μkm to nm [15,16]. The Tarfenol-filled polyurethane [9] isotropic composite made up by 25% particles volume fraction has demonstrated the best magnetodeformational response. We have performed the preliminary study of polyurethane composite filled with 25% volume fraction of mechanochemically synthesized Galfenol particles magnetic and mechanical properties [13]. Conversion Electron Mossbauer spectroscopy and X-ray diffraction has not revealed explicit interaction between polymer and metal particles. Synthesized composite has clearly demonstrated a significant magnetodeformational effect, and it has shown that it may be raised threefold by application of magnetic field during composite polymerization.

The present work has been devoted to research effects of volume fraction and spatial arrangement of Galfenol particles in the polyurethane matrix on magnetodeformational effect and mechanical properties of the composites.

2. Experimental

2.1. Specimens' preparation

The preparation of the composite material comprised four steps:

(1) Mechanosynthesis of ferromagnetic particles (fp) with magnetostrictive FeGa phase composition. Synthesis was performed in AGO-2 planetary mill sealed under Ar. Specimens of powdered mixtures of 80Fe20 Ga composition (numbers are weight percentage) were subjected to mechanical activation during 2 h, using a 250 cm^3 milling drum and 200 g of 5 mm steel balls. The drum rotation speed was ~1000 rpm.
(2) Preparation of the liquid polymer substance. We applied modified polyurethane (PU).
(3) Filling the liquid polyurethane (PU) polymer with different fp volume fractions (x = 10, 15, 25 and 50%) and ultrasonic intermixing. The mixtures of PU and fp were prepared by ultrasound exposure at 18 kHz (Bandelin HD2200) for 3 min for homogeneous distribution of filler particles in liquid polymer.
(4) Fp stabilization with orientation in PU matrix. The prepared suspensions were transferred to a 20 × 10 × 4 mm Teflon rectangular mold. Final composite polymerization in the mold was performed under an applied magnetic field (0.5 T) in an electrically thermostatic furnace.

2.2. Samples characterization

Phase composition of synthesized fp particles has been analyzed by Mossbauer spectroscopy (MS) on MS1104 Em with Co57 (Rh) source in γ-ray transmission mode. For bulk composite samples spectra have been obtained in the backscattering mode with registration of resonant conversion electrons (CEMS) [17]. The spectra were fitted with Univem MS software [18], using spectrum fitting by a number of analytical functions describing the relative individual components of the processed spectrum.

Transmission electron microscopy (TEM) dark field particles images with electron diffraction have been obtained on LEO 912, and Scanning electron microscopy (SEM) images of powder composites on Quanta 3D FEI microscope.

A multidimensional X-ray computed tomography (CT) [19] for 3D-visualization of the composite microstructure was performed on an Empyrean PANalytical X-ray diffraction instrument equipped with a PIXcel3D detector using Cu(Kα)-radiation. Sample stages and optics have been optimized for phase determination, phase distribution and CT measurements. We analyzed the X-ray data using PANalytical High Score plus software and VG Studio MAX 2.1. by Volume Graphics [20].

We measured the mechanical properties of the composites via particles concentration in the direction of magnetic field applied at samples manufacturing on Perkin Elmer DMA 8000 in the temperature range of −100 to 200 °C, with the heating rate −5 °C/min, and the frequency of 1 Hz. We have carried out the operations in a single cantilever bend. The storage modulus and the mechanical damping of the composite was measured as a function of the temperature in the DMA test.

The magnetodeformational effect (changes in the length of the specimen under applied magnetic field) was analyzed in the determined direction of its maximum value on a laboratory set of magnetostriction measurements in applied field up to 12 kOe.

3. Experimental results

Flat particles (shown on Fig. 1(A)) obtained by mechanosynthesis as small as <2 μm represent microstructure composed of α-Fe (Ga) (80%) solid solution with small inclusions of Fe_3Ga (20%) phase derived from Mossbauer spectra analytical fitting [14] (Fig. 1(A) and (B)). The bulk polymerized PU/FeGa composite with direction of applied magnetic field is shown on Fig. 1(C). As-prepared samples exhibited remanent magnetization (RM) at room temperature acquired during the polymerization in external magnetic field. The Jr values have shown almost linear increase with the rise of filler concentration (Fig. 2(A)).

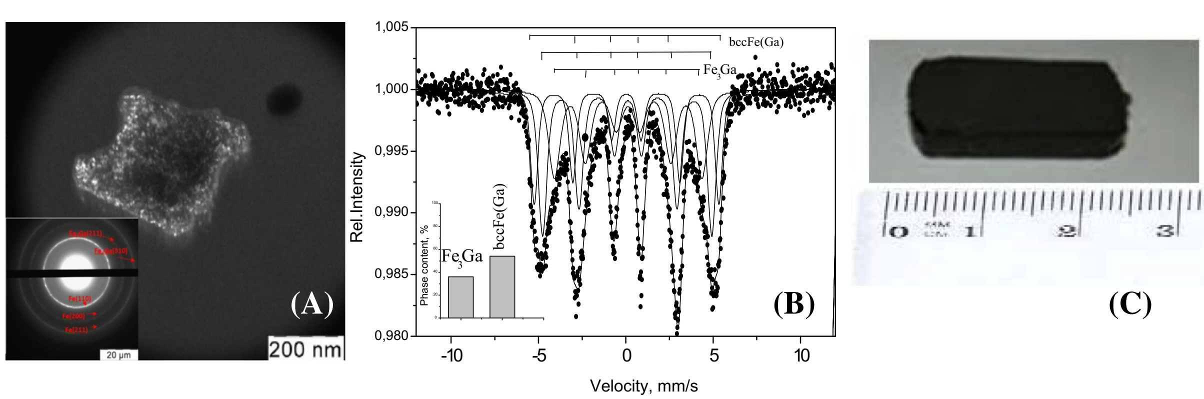

Fig. 1. Microstructure of synthesized FeGa-alloy particles: (A) dark field TEM image of the particle, (B) Mossbauer spectrum of synthesized particles with derived phase analysis (in inclusion), (C) real size of the FeGa/PU composite (x – the direction of applied magnetic field at manufacturing).