Magnetodeformational Anisotropy of FeGa/PU Hybrid Nanocomposite Via Particle Concentration And Spatial Orientation

Tatiana Yu. Kiseleva[1,a*], Sergey I. Zholudev[1,b], Alla A. Novakova[1,c], Tatiana S. Gendler[2,d], Igor A. Il'inykh[3,e], Alexandra I. Smarzhevskaya[1,f], Yuriy Anufriev[1,g], Tatiana F.Grigorieva[4,h]

[1] M.V. Lomonosov Moscow State University, Faculty of Physics, Leninskie Gory, Moscow, Russia

[2]Shmidt Institute of the Physics of the Earth, RAS, Gruzinskaya str., Moscow, Russia

[3]National University of Science and Technology (MISIS), Leninskiy av., Moscow, Russia

[3]Institute of solid state chemistry and mechanochemistry RAS, Novosibirsk,Kutateladze str, Russia

[a]Kiseleva.TYu@gmail.com,[b]S.I.zholudev@gmail.com,novakova.alla@gmail.com,[d]gendler06@mail.ru, [e]ilinyh.igor@gmail.com, [f]smarzhevskaya@physics.msu.ru,[g]hf_hf@newmail.ru, [h]grig@nsc.ras.ru

Keywords: Nanocomposites, FeGa, magnetic anisotropy

Abstract. The present work has been undertaken to research effects of structure, morphology, volume fraction, spatial arrangement of magnetostrictive intermetallic FeGa alloy particles dispersed in modified polyurethane matrix. Correlation of composite magnetic behavior with structure and mechanical properties has been obtained by measurements of magnetostriction, remanent magnetization anisotropy, SEM, and dynamical mechanical analysis. Anisotropic chain structures of magnetic particles within the polymer with different interparticle interactions were observed. The increase of the magnetostrictive response with tailor-made magnetic anisotropy induced by magnetic particles volume fraction has been demonstrated.

Introduction

Composites with magnetic fillers 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]. The ability of taking the advantage of particular properties of particles as the constituent materials and their orientation in the matrix is the most important motivation for the development of magnetically anisotropic metal-polymer functional hybrids. The present work has been undertaken to research effects of structure, morphology, volume fraction and spatial arrangement of magnetostrictive mechanosynthesized intermetallic FeGa alloy particles in polyurethane matrix. The choice of FeGa alloy composition was made in view of its famous magnetostrictive behavior increase at specific phase composition [2].

Experimental

The preparation technique of the composite material comprises three steps: 1) mechanosyntnesis of ferromagnetic particles (fp) with magnetostrictive FeGa phase composition; 2) filling of the liquid polyurethane (PU) polymer with different fp volume fractions (x=15÷50%) and ultrasonic intermixing; 3) fp stabilization in PU matrix during its polymerization in two regimes, without and with an applied magnetic field of 0.5 T, in an electric furnace. The random oriented samples will be labelled further as **RO** and magnetically oriented samples - as **MO** correspondingly. Composites xFeGa/PU samples were obtained as rectangular pieces with dimensions of 20 × 10 ×4 mm. Peculiarities of sample preparation may be found in papers [3-5].

Scanning electron microscopy (SEM) pictures of composite have been obtained on Quanta 3D FEI microscope. Anisotropy of remanent magnetization acquired during the composite

preparation in external magnetic field was measured using magnetometer (JR-6, AGICO) in three orthogonal directions in shielded space. Mechanical properties and magnetostriction were measured using Perkin Elmer DMA 8000 and laboratory set of magnetostriction measurements correspondingly.

Results and discussion

As was reported recently [3] squashed flat particles of < 2 μm sizes can be subsequently obtained by mechanosynthesis of Fe:Ga powder mixtures in high energy planetary ball mill. This results in magnetostrictive bccFe(Ga)/Fe_3Ga phase composition. Fig.1a) taken from [4] shows the behaviour of magnetostriction in external magnetic field for compacted FeGa particles, **RO** and **MO** composites with 25% fp volume concentration as well as values in maximal applied field for FeGa single- and polycrystal [2]. The main results of preliminary investigations [3-5] are the following: 1) maximal values of magnetostriction (dλ/λ) for compacted particles and **RO** composite with 25% Fe-Ga particles are close and three-fold lower than that of **MO** composite with the same fp concentration; 2) MO maximal magnetostriction value in turn is significantly lower compared to values typical for single crystalline and polycrystalline FeGa alloy samples; 3) the testing of PU/particles composite by Mossbauer spectroscopy and XRD did not reveal explicit interaction between polymer and metal particles. So the attempts of modifying MO composite magnetic properties by choosing the concentration of ferromagnetic particles were the aim of this study.

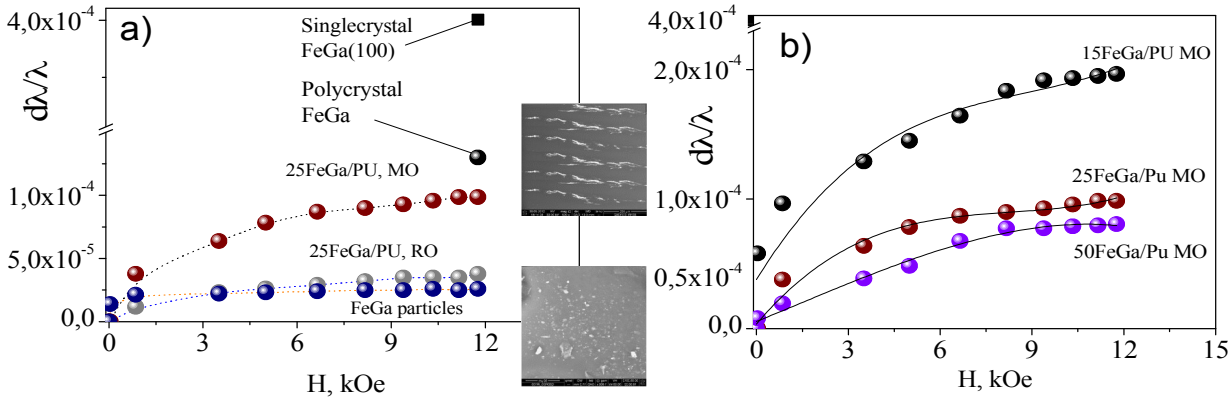


Figure 1. Magnetostriction behavior in external magnetic field. (**a**): blue balls- compacted FeGa particles, grey balls- **RO** FeGa/PU composite (25% fp), dark red bolls- **MO** FeGa/PU composite (25% fp); black square and ball- magnetostriction values for FeGa single and polycrystal correspondingly at 12 kOe [2]; SEM images for MO and RO composites are shown in insets (**b**): *x*FeGa/PU, **MO** magnetostriction in dependence on applied field for different particles *x* (fp volume fraction).

As Fig. 1(b) shows, the magnetostriction behavior at variable applied magnetic field demonstrates the inverse dependence on concentration of Fe-Ga particles in polymer matrix. Maximal magnetostriction values achieved at 12 kOe decrease exponentially with the increase of fp volume concentration (Fig.2 (a)).

As prepared samples exhibited at room temperature remanent magnetization (RM) acquired during the polymerization in external magnetic field. The RM values showed linear increase with the rise of filler concentration. X-Y axes on the surface of measured scraps were chosen accidentally because the last were not oriented relative to the magnetic field applied at polymerization. So the values obtained give no correct information about in plain RM anisotropy. Nevertheless RMx/RMy or vice versa values varied from 8 to 16 with concentration increase suggesting the chain structure of fp particles evidently in external field direction. Z-axe was chosen along each sample thickness. Plain anisotropy of remanent magnetization (ARM) revealed the exponential decrease with the fp concentration rise (Fig. 2(a)), what correlates with magnetostriction behavior. The mechanical composite properties such as Young module E and glass temperature Tg at the same time showed expected monotonic increase via particle concentration (Fig.2(b)). It is worth noting that adding fillers to a neat polymer melt changes its rheology influencing both the way of the curing processes and the properties of the ultimate product. Key factors are filler size, shape, concentration and extension of any interactions between the particles [1]. As a result of polymerization procedure performed in external magnetic field highly strong magnetostriction was observed for greatly anisotropic composite with low concentration of ferromagnetic particles in polymer.

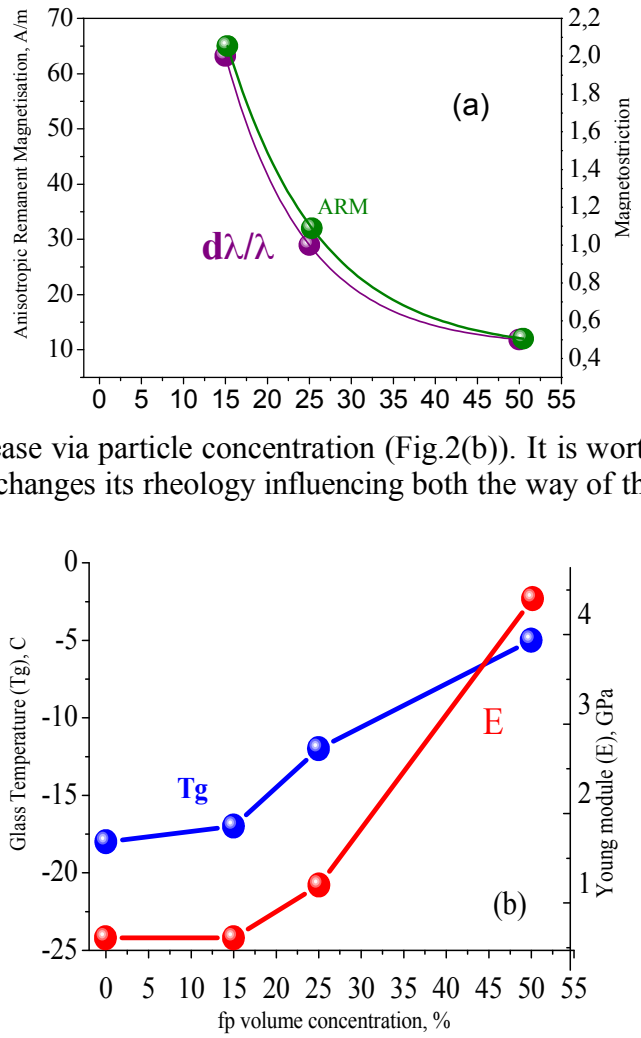
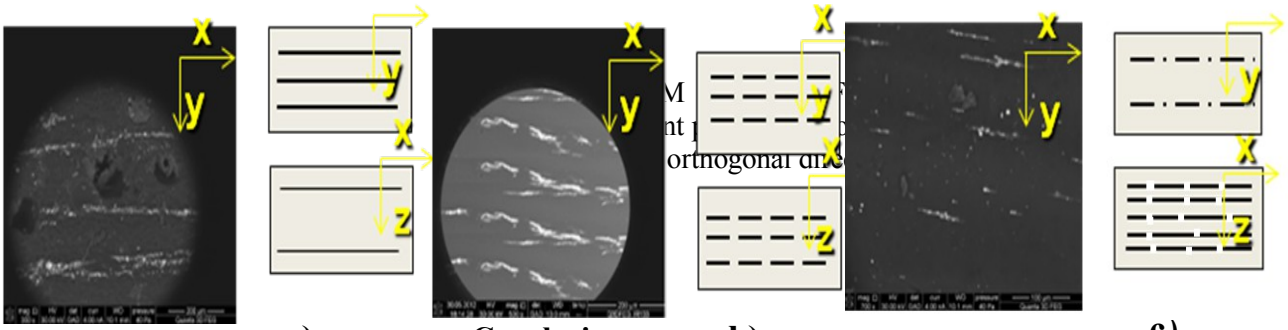


Figure 2. Maximal magnetostriction values and Volume ARM (a) and Young module E, Glass temperature Tg (b) via fp volume concentration

Explanation of this effect was found in SEM pictures obtained from different directions of the composite samples. (Fig.3). Well-structured chains (strips) of FeGa particles within the composite strained in external magnetic field direction have been determined from the images. Particles self-organization in a polyurethane matrix during the polymerization in external magnetic field results in their spatial anisotropic stabilization along the magnetic field lines similar to described earlier [4]. The longest chains revealed the composite containing 15% of particles, the most broken chains were observed for 50% volume fraction of particles. Moreover large particles are covered by chains of small particles [5]. Non-uniform arrangement of the particles along the lines arises [6] from the combination of magnetic forces and gravity such as polymer rheology. The increasing of fp volume fraction in composite followed by strong ferromagnetic interparticle interaction leads to formation of transversal columnar structure [7] in direction perpendicular to external magnetic field (Fig.3, c). This fact evidently is affected in ARM and magnetostriction decrease.

Magnetostrictive properties of composites are conditioned by a number of factors. The sample appears to possess its own demagnetizing field, depending on sample shape and the amount of magnetic material inside. This causes the elongation of the sample (positive magnetostriction). Another factor is related to the inhomogeneity of the local fields, the contribution of which depends on the short-range order in the subsystem of magnetic particles in a polymer matrix. Furthermore, the magnetostriction value should depend on the presence of interparticles spatial correlations for the

given sample size and their concentration in polymer matrix. The decisive factor according to [8] is the ratio between the number of isolated particles and particles aggregated into chained clusters. The influence of the particle chains to overall deformation value is the more significant the greater their length. It should be noted that in this work we used particles of material that itself belongs to the class of magnetostrictive materials. The presence of elastic stresses in the particles due to mechano-chemical process of their preparation is a reinforcing factor in increasing their magnetostrictive effect. Recently it was reported [9] that intensive mechanical treatment of the bcc-iron particles in a planetary ball mill leads to the formation of particles with strain inducing magnetic anisotropy. It was shown that iron particles are crushed and flattened out in the elongated plate with the axis of easy magnetization oriented along the plane of flattening. We consider that this effect may affect the particles magnetostriction value by additional stress influence on the FeGa particle structure. Polymerization of polyurethane filled by FeGa particles in applied magnetic field leads to their alignment along the axis of easy magnetization.

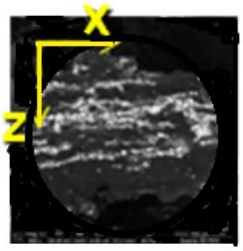


M posites with
nt % , c,d) 50%
orthogonal dir

a) b) c)

Conclusions

Summarizing the present results, the correlation of composite magnetostrictive properties with the anisotropic arrangement of Fe-Ga particles in polymer has been observed. The maximum values of functional characteristics were obtained for composition with 15% particles volume fraction with preferable spatial orientation of longest particles chains in field direction. Particles phase composition, lattice distortion and stresses obtained mechanochemically possess the increase the own particles magnetostriction. Directed stabilization of such particles into associated chains enhances the magnetodeformational anisotropy of the composite.



d)

Acknowledgements. *Support by RFBR Grant 13-00-838 and Grant 14.Z50.31.0017 of the Government of the Russian Federation are acknowledged*

References

1.Metal-polymer nanocomposites ed. By L.Nicolais, Willey&Sons, 320, (2005)
2. J.Atulasimha, A.B. Flatau*: Smart Mater. Struct.*, V.20. P.043001 (2011)
3.T.Grigoryeva, T. Kiseleva et al. *Physics of Metals and Metallography* 113,575-582 (2012).
4. T.Kiseleva, S.Zholudev, A.Novakova et al. *Tech. Phys. Letters*. (2013) 39 1110
5. S.I.Zholudev , T.Yu.Kiseleva *Hyperfine Interactions* 226, 375-382 (2014).
6. A.Novakova,V. Lanchinskaya, T.Gendler et al *JMMM* 13, 256(2003).
7.S.Abramchuk ,E. Kramarenko, G.Stepanov, et al. *Polymer Adv.Techn*. 18, 883-890 (2007)
8. Y.L.Raikher, O.V. Stolbov *Computational Continuum Mechanics*. 2,2, 85-95 (2009)
9.A.A.Novakova, O.V.Agladze, T.Yu.Kiseleva *J. of Surf. Investig*. 16,12, 1863-1867 (2001).