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Polymer bonded magnetic composites based on Nd-Fe-B

Various types and contents of magnetic powder and polymer matrix induce different mechanical, magnetic, electrical and optical properties of final composite material. The strong influence of relatively small amounts of filler particles on the dynamic mechanical properties of polymers has significantly contributed to increased use of polymer materials in many commercial applications. The higher content of magnetic filler has direct influence on magnetic properties of composites, but also may change the rheology of polymer melt during process and, subsequently, impact the mechanical strength of bonded magnets. For better insight into viscoelastic behaviour of composites, beside experiments, a theory that explicitly takes the shape factor, particle distribution, particle-particle interactions as well as particle-polymer matrix interactions into account is required. The mathematical prediction of storage modulus behaviour is examined. The several proposed analytical models are tested versus experimental results. Some of applied models agree very well with experimental data, whilst others deviate significantly.

Key words: *Nd-Fe-B, Bonded Magnet, Composite, DMA*

1. INTRODUCTION

The magnetic composite materials based on Nd-Fe-B i.e. bonded magnets have been commonly used in various fields [1], such as micro-electromechanical system (MEMS) applications [2,3], automobile parts [4,5], electronics, communication and sensing elements [6]. Optimal mechanical properties and dimensional stability of final materials are difficult to obtain with commercial permanent (metallic) magnets due to their extreme brittleness and poor thermal stability that are exacerbated by high temperature, corrosive, and dynamic loading environments [7]. The mechanical properties of polymer bonded magnets depend strongly on properties of the polymer matrix, magnetic filler, and interfacial conditions between the components. In the other hand, the increasing amount of Nd-Fe-B powder in the bonded magnet plays a crucial role in determining magnetic properties. Advantages of the using bonded composite materials include their simple technology, possibility of forming their final properties, lowering manufacturing costs because of no costly finishing and lowering of material losses resulting from the possibility of forming any shape [8].

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The presented study is undertaken with the intention to understand the effect of different filler contents on the dynamic mechanical and magnetic properties of the Nd-Fe-B/epoxy magnetic composite materials. The filler content from 2.7 vol.% to 75 vol.% are employed in order to explain and compare behaviors of produced composites. The correlation between storage modulus (E') and Nd-Fe-B content in polymer matrix at the ambient temperature is widely discussed. Also, the experimental results are compared with several proposed mathematical models.

2. EXPERIMENTAL

Composites with varied content of Nd-Fe-B particles in epoxy matrix were produced by compression molding method under a pressure of 4MPa at room temperature, using a lab scale compression molding press. No external magnetic field is used during the cure.

The rapid quenched Nd-(Fe,Co)-B magnetic powder, brand named as NM-B, used in this study was supplied by Ningbo Haixin Co. Ltd, China with a particle size from 74 to 177 µm and following magnetic properties: H_{cb}=477.5 kA/m, H_{cj}=692.3 kA/m, B_r=0.82 T, $(BH)_{max}$=104.2 kJ/m^3. The chemical composition is: Nd 21-25 wt%, Co 3-5 wt%, B < 1.5 wt%, Zr 3-5 wt%, Fe balance. Thermosetting epoxy system that is a combination of liquid mixture of Bisphenol A and Bisphenol F resins and cross linking agent (hardener) which cures fully at room temperature was used as a polymer matrix. The cured pure epoxy resin has tensile strength ~ 58 MPa, elongation ~ 2.8%, compression strength ~ 96 MPa, flexural strength ~ 78 MPa and density ~ 1200 kg/m^3.

The structure and morphology of fracture surfaces for synthesized composite materials are observed by JEOL JSM-5800 Scanning Electron Microscope, with an accelerating voltage of 20 kV. In order to study dynamic mechanical behavior, TA Instruments Q800 Dynamic Mechanical Analyzer was used. Testing is done over a temperature range from 25 ºC to 100 ºC with a temperature ramp of 3 ºC/min. A universal material testing machine (Schenck TREBEL RM 100) is used for tensile measurements. The magnetic properties were obtained using Superconducting Quantum Interference Device (SQUID) magnetometer at ambient temperature (300 K). Maximum magnetic field strength was 5 T.

3. RESULTS AND DISCUSSION

Uniform particle distribution and good adhesion between Nd-Fe-B particles and a polymer matrix are essential for the quality of composites, especially at temperatures above the glass transition temperature of the polymer. SEM micrographs of fracture surface morphology of Nd-Fe-B/epoxy composites are presented in Figure 1.

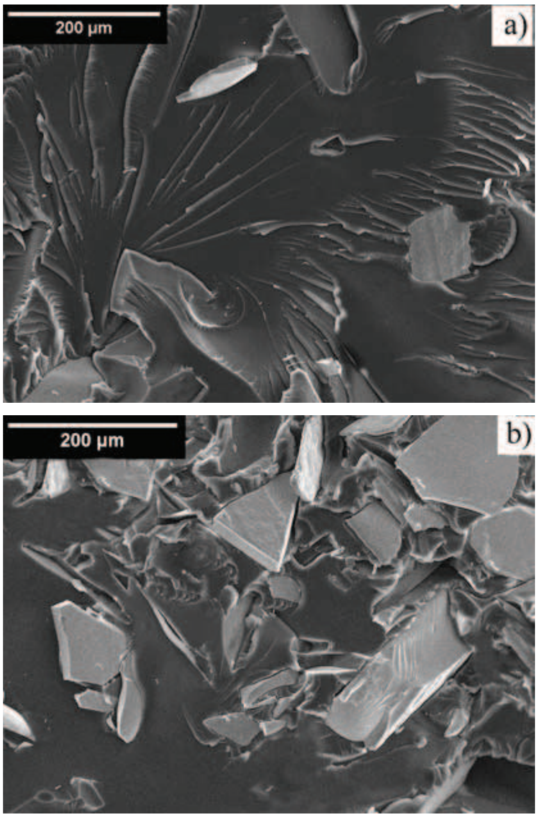

Figure 1 - SEM micrographs of fracture surface of composite material with a) 2.7 vol.% and b) 14 vol. % of Nd-Fe-B filler

The Nd–Fe–B particles are shown as light grey and the epoxy matrix is shown as dark. Although Nd–Fe–B particles are of variable size and shape, they seem to be attached rather well to the matrix.

The dynamic mechanical properties of the pure epoxy resin and the Nd-Fe-B/epoxy composite materials are examined as a function of temperature. In the glassy region (around 25 ºC), modulus of the pure polymer, modulus of the filler, and concentrations of both, as well as the adhesion factor between the filler and polymer, have direct influence on total dynamic modulus of composites [9-11]. The increasing content of Nd-Fe-B filler into epoxy matrix induces an increase the storage modulus of pure epoxy polymer, respectively. When comparing material properties, a material with a higher storage modulus would be stiffer and harder to deform than one with a lower E'.

In order to determine the in-plane tensile properties of the investigated composite materials, standard test tensile method is used [12]. The values of ultimate tensile stress, elongation and modulus of elasticity could be taken from obtained stress-strain diagrams for all investigated composites. Results suggest that with an increasing content of the Nd-Fe-B filler, the values of tensile stress and elongation are decrease [13]. In addition, composite materials become more brittle, and less ductile. Modules of elasticity obtained by tensile tests are increase with higher quantities of magnetic filler. This is crucial in analysis of possible use of the investigated magnetic composite materials as functional material (Fig. 2.). This means that materials with higher amounts of Nd-Fe-B filler, subject to equal stress levels (ballast), tolerate 2 to 3.5 times lower deformation.

The modulus of elasticity is a very important parameter for analysis of the composite materials behaviour under discontinuous load conditions. The values of elastic modulus, obtained by tensile tests, upswing with an increasing amount of Nd-Fe-B powder from 14 vol. % achieve 9.2 GPa. Within the narrow region, up to 4 vol. % content of Nd-Fe-B, where the modulus of elasticity is practically constant according to tensile tests, dynamic-mechanical analysis could be applied to acquire additional information's related to mechanical behaviour about transitions in polymer composites [14].

The values of storage modulus observed by DMA were compared with elastic modulus obtained by tensile tests (Figure 2.). In contrast to Deng, S. et al. [15] mechanical properties at temperatures higher than ambient are not compared with DMA results observed using two different clamps. It seems that observing the elastic modulus of composites by tensile and DMA tests at room temperature in the present study gives a better look at the increasing trend of elastic components of materials with increasing Nd-Fe-B filler content in the polymer matrix.

Ideally, in an attempt to reduce laboratory cost, one would like to make a prediction of a new material's behaviour by numerical simulation, with the primary goal being to accelerate trial and error experimental testing. The recent dramatic increase in computational power available for mathematical modelling and simulation raises the possibility that modern numerical methods can play a significant role in the analysis of heterogeneous microstructures. The several proposed analytical models are tested versus experimental data as it is illustrated in following text. Some of applied models agree very well with experimental data, whilst others deviate significantly.

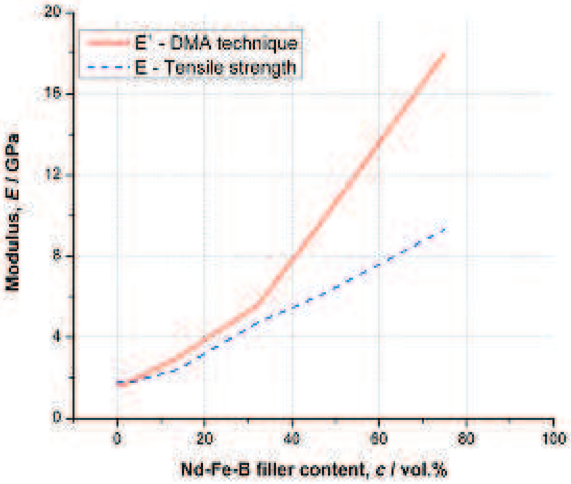

Figure 2 - Comparative view of the changes in the modulus of elasticity at 25 ^{O}C

There have been several attempts to derive formulas giving the apparent modulus due to a dispersion of particles in polymer [16]. The earliest of these attempts was by Smallwood using the analogy to Einstein's viscosity equation. Smallwood's estimate is only good at very low filler concentrations. A number of attempts have been made to incorporate interactions between neighbouring particles to allow prediction at higher volume fractions. Most of these models add one or more terms to a polynomial series expansion. One of the most cited model of this class is the Guth-Gold [17]. Later Guth extended the Guth-Gold model to include impact of particle shape on properties. Guth introduced a shape factor f (ratio of diameter to width of particle) and proposed a new equation [18]. Budiansky developed a model, for the special case of rigid particles in an incompressible matrix written as [19]. Ponte Castaneda has proposed a different self-consistent estimate for rigid particles in a neo-Hookean matrix [20]. Later Govindjee and Simo proposed the novel model, for the case of rigid particles in a neo-Hookean matrix written [21]. In addition, it is worth to mention is the empirical formula suggested by Brinkmann [22]. Major characteristics of all aforementioned theoretical models are: they neglect the impact of filler properties and assume that the medium wets the filler particles, and they do not chemically react with the filler surface. Mori-Tanaka proposed a model, which takes into consideration the impact of filler properties on overall composite properties [23].

The experimentally obtained values of storage modulus are compared with analytical models discussed above and presented in Figure 3. Predictions of models proposed by Budiansky, Ponte Castaneda and Govindjee-Simo give inadequate estimation so they are not included in Figure 3.

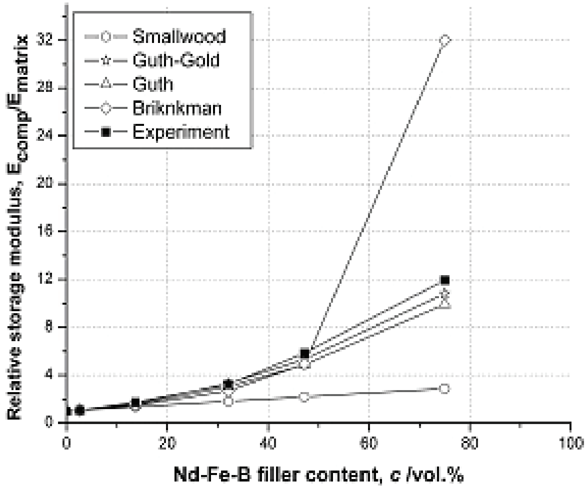

Figure 3 - Models predictions against experimental data

From Figure 3 one may notice that all models included in analysis give very good predictions of storage modulus at lower particles concentrations (till 14 %). This suggests that in this concentration range, the interactions between neighbouring particles have very low intensity. At higher concentrations, interactions become high intensity, this is why the Smallwood's model starts to show significant deviation from experimental results. Mori-Tanaka's model follows the trend of experimental results, but it gives poor predictions. Brinkman's model gives good predictions at high concentrations, but at very high concentrations of particles, it extensively overpredicts the storage modulus. The Guth and Guth-Gold models are in very good agreement with experimental results. The explanation for this behaviour lies in the fact that both models take into consideration interactions between neighbouring particles.

Magnetic properties of polymer bonded magnets are affected by the magnetic properties and weight (volume) ratio of the powder. The changes of remanence, coercivity and maximum energy product with an increasing content of Nd-Fe-B in the epoxy matrix are presented in Figure 4. The presented graph illustrates the upswing of three magnetic parameters of composite materials with increasing amounts of

Nd-Fe-B particles in the epoxy matrix. For example, the maximal energy product for composite with 75 vol. % Nd-Fe-B is around 8 MGOe, which is two times higher than for composite with the 47 vol. % Nd-Fe-B case. For composites with Nd-Fe-B content higher than 32 vol. %, *(BH)*$_{max}$ rapidly increase i.e. for the highly filled composites even a small addition of magnetic medium have a strong influence on magnetic properties of bonded magnets.

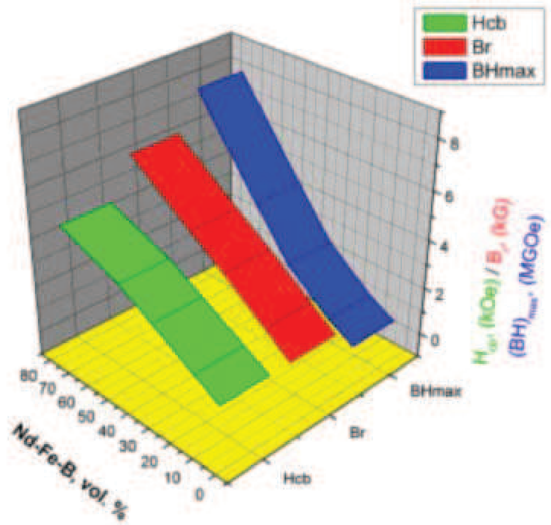

Figure 4 - Magnetic properties of the composites in the function of the Nd-Fe-B filler content

The maximum energy product $(BH)_{max}$ of Nd–Fe–B bonded magnets can be simulated by using a mathematical model, and choosing appropriate parameters for the magnetic texture and the magnetic coupling between micro-grains, one can increase the value of $(BH)_{max}$ [7, 24].

4. CONCLUSION

The mixture of Nd-Fe-B particles and epoxy resin is suitable for bonded magnet applications due to good adhesion, homogeneity, mechanical and magnetic properties. The tensile tests at ambient temperature show enhancement of modulus of elasticity with quantity of magnetic filler, which is a crucial parameter for analysis of composite materials behaviour. The increase of the amount of Nd-Fe-B filler into the epoxy increases the storage modulus of pure epoxy polymer. The values of the tensile stress and elongation of the composite material decrease while the modules of elasticity increases with the increase of the amount of the magnetic powder. This implies that the materials with the higher amounts of filler exposed to the same stress level, tolerate 2 to 3,5 times lower deformation. The presented results show increase in three magnetic parameters of composite material with the increase of the amount of the magnetic powder.

Considering the increasing interest in polymer composites and advanced analytical tools, the present study provides a useful basis and motivation for future experiments and theory development for the multifunctional components and commercially important polymer bonded magnets.

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IZVOD

POLIMERNI MAGNETNI KOMPOZITI NA BAZI Nd-Fe-B

Različite vrste primenjenih magnetnih prahova i polimerne matrice, kao i njihovi udeli, utiču na mehanička, magnetna, električna i optička svojstva finalnog kompozitnog materijala. Veliki uticaj relativno malih udela čestica punioca na dinamičko mehanička svojstva polimera značajno doprinosi većoj komercijalnoj upotrebi kompozitnih materijala. Veliki udeli magnetnog praha u mnogome utiču na magnetna svojstva kompozita, ali takođe mogu dovesti do reoloških promena u polimeru tokom procesa topljenja, što ima direktan uticaj na mehanička svojstva polimerom vezanih (bonded) magneta. Radi boljeg uvida u viskoelastična svojstva kompozita, osim eksperimenata, neophodno je uzeti u obzir i teorijska razmatranja koja eksplicitno objašnjavaju uticaj faktora oblika, raspodelu čestica, interakcije između čestica, kao i interakcije između čestica i polimera. U tom smislu vršeno je i matematičko predviđanje ponašanja modula uskladištene energije (E') kompozita. Nekoliko predloženih analitičkih modela su testirani i upoređeni sa eksperimentalnim rezultatima. Neki od primenjenih modela se veoma dobro slažu sa dobijenim eksperimentalnim rezultatima, dok neki značajno odstupaju.

Ključne reči: *Nd-Fe-B, Bonded Magnet, Kompozit, DMA*

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