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Effect of particle diameter and packing density on heating of metallic iron particles in alternating magnetic field

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Abstract. The heat losses originated from the electro-magnetic absorption in mechanically packed metallic iron nano- and micro-sized magnetic particles (MPs) and in magnetic epoxy-based composites with embedded MPs were studied by Calvet microcalorimetry. Nano-sized MPs with numerical average diameter 40 nm were synthesized by electrical explosion of wire method; micro-sized MPs with numerical average diameter 2 μm were synthesized by flame decomposition of the iron pentacarbonyl. The specific loss power (SLP) of re-magnetization of press-packed powdered samples and epoxy composites in magnetic field 1750 A/m at 214 kHz was measured as a function of the volume fraction of MPs in the sample. The results showed up that SLP depended both on particle size and on volume fraction, which meant the significant effect of magnetic interaction among particles on the heat losses in electromagnetic absorption.

1. Introduction

Modern polymer composites are multicomponent systems containing a polymer matrix and powder filler, which impart different functional properties. Composite materials based on epoxy resins are of particular interest, since composites of this class have mechanical strength, chemical resistance, high dielectric properties after curing, good adhesion to metals. Epoxy composites are promising materials for use in the aerospace industry. Magnetic-filled polymer composites containing magnetically soft particles are widely used to manufacture magnetic screens to absorb electromagnetic radiation of various frequencies and coatings to protect instruments and sensors that are sensitive to electromagnetic radiation. Currently, there is a number of works devoted to the electromagnetic absorption of polymer composites based on thermoplastic polymers with embedded magnetic particles. [1-9].

Absorption of electromagnetic radiation often means the conversion of electromagnetic energy into heat. Therefore, a direct measurement of the thermal power emitted by composites with magnetic filling during the absorption of electromagnetic fields can be an accurate quantitative estimation of the value of the absorbed emission. Calorimetry is a direct experimental method of measurement of heat effects in various chemical and physicochemical processes.

In our recent article [10] we used Calvet microcalorimetry to measure heat evolution due to electromagnetic absorption in polymer composites related to hyperthermia, which is a novel developing method in cancer curing therapy. The majority of works on the magnetic hyperthermia deal with the systems of non interacting ferromagnetic particles suspended in a liquid carrier [11].

These results can be applied only to the systems with very low concentration of the particles. Meanwhile, it was shown [10] that magnetic interaction between the particles can play a significant role in the heating effect. Essentially, interaction among magnetic particles is a function of the average distance between them, which in turn is related to the volume fraction of particles in the sample or their packing density.

The objective of the present research was to study the effect of packing density and particle diameter on the heat evolution in alternating magnetic field. We have tested nano-sized and micro-sized metallic iron particles using Calvet microcalorimetry for the direct measurement of the heat effects associated with electromagnetic absorption.

## 2. Experimental

2.1. Materials

The experimental study was performed on two different samples of magnetic particles (MPs), further on denoted as nano-Fe and micro-Fe. Nano-Fe MPs (10–200 nm in diameter) were synthesized by the electric explosion of wire (EEW) method. The EEW method is based on the evaporation of a part of a metal wire by a high-voltage discharge with the subsequent condensation of particles in the vapor phase. Details of the experimental setup of EEW can be found in ref. [10]. Micro-Fe MPs (0.5–4.5 μm in diameter) were a commercial product purchased from Sintez-CIP, Dzerzhinsk, RF, synthesized by flame decomposition of the iron pentacarbonyl.

Figure 1 presents TEM images of nano-Fe and micro-Fe MPs elaborated in the study together with number averaged particle size distribution (PSD) plots obtained by the graphical analysis of multiple TEM images (Figure 1, insets). Nano-Fe MPs are spherical, while carbonyl Fe particles are unevenly shaped. The size difference between samples is significant. Carbonyl Fe median PSD value is around 2 μm, whereas nano-Fe particles are relatively small with median value of the PSD around 40 nm, while also being more uniform, as carbonyl Fe particles has a wider distribution with considerable number of particles larger than 4 μm.
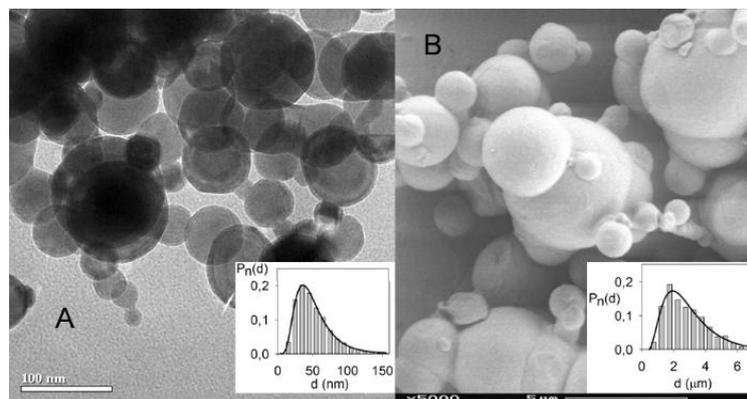
Figure 1. TEM images of magnetic particles used in the present work.. A – Fe nanoparticles synthesized by EEW method B – carbonyl Fe, a commercial sample The insets give PSD of the particles obtained by the graphical analysis of TEM images.

Hysteresis loops for nano-Fe and micro-Fe and the enlarged view of the hysteresis in low fields are given in Figure 2.
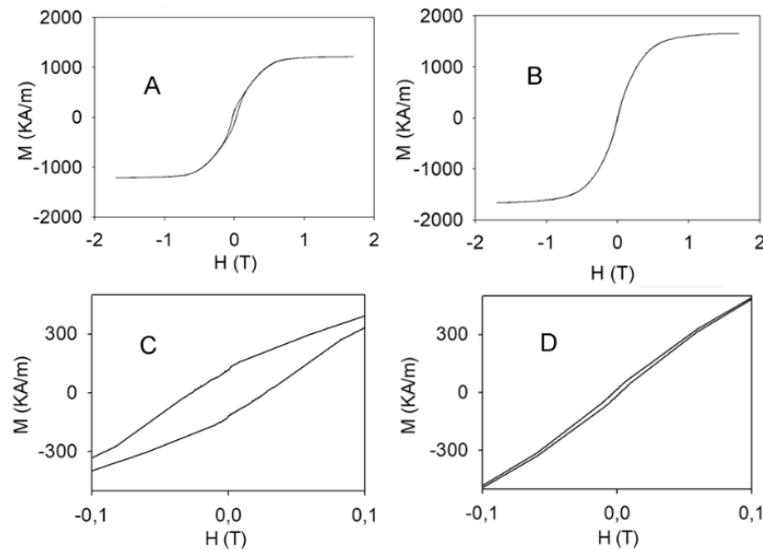
Figure 2. Hysteresis loops of magnetic particles used in the present work. A – nano-Fe, synthesized by EEW method B – micro-Fe, a commercial sample. Enlarged view of the hysteresis loops in low fields. C – nano-Fe, D – micro-Fe.

The shape of the loops indicates that the samples are magnetically soft ferromagnetic materials. Coercivity was 0,026 T for nano-Fe, 0,005 T for micro-Fe, while residual magnetization occurred to be 120 kA/m for nano-Fe, 24 kA/m for micro-Fe.
XRD images are given on the Figure 3.
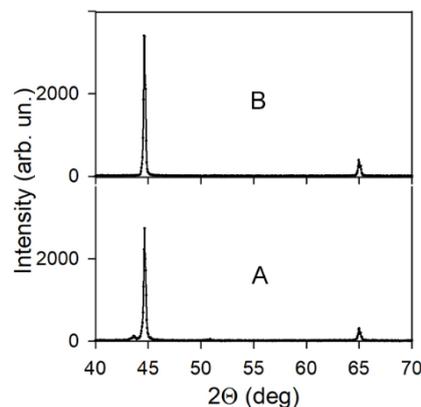
Figure 3. XRD of magnetic particles used in the present work.. A – nano-Fe synthesized by EEW method B – micro-Fe, a commercial sample.

XRD images show that the crystalline structure in both cases refer to α-Fe cubic lattice.
Samples of metal powder with varying packing density were prepared by compressing of powdered material in glass tube vials, with diameter of 4 mm. Length of metal powder part was close to 20 mm for all samples. The average packing density was characterized by the volume fraction of Fe particles in a vial, which was evaluated based on the weight of the sample and the geometrical dimensions of a press-packed sample cylinder. Resulting volume fraction of packed metal particles varied from 0,12 for nano-Fe to 0,5 for micro-Fe.

Epoxy-diphenylolpropane resin KDA (Chimex Ltd., St. Petersburg, Russian Federation) was used as a polymer matrix for the preparation of magnetic composites with particle content from 2 to 26% by volume. First, the resin was mixed with a hardener - three (ethyl) -tetra (amine) (Epital, Moscow, Russia) in a ratio of 6:1 by weight. Thereafter, the weighed amounts of powdered samples were mixed with the liquid epoxy resin composition at temperature of 25 ° C for 10 minutes to obtain a homogeneous mixture. The filled composition was then placed in a cylindrical polyethylene mold with a diameter of 4 mm and cured for 2 hours at 70 ° C. Resulting sample length was ~40 mm.

Bulk Fe wire 0,5 mm in diameter was used as a reference material with 100% metal content. It was cut into 3 mm pieces and was embedded in epoxy resin to fit the dimensional properties of other samples. It was the same wire that was used to synthesize nano-Fe by the EEW method.

2.2. Methods

Images of transmission electron microscopy (TEM) were obtained using a JEOL JEM2100 microscope operated at 200 kV. TEM samples were applied to carbon coated copper grids from an isopropanol suspension homogenized by ultrasonic treatment. Magnetic hysteresis loops were measured at 300 K using a VSM magnetometer.

X-ray diffraction (XRD) was performed on a BrukerD8 Discover instrument using copper emitting with a graphite monochromator on a diffracted beam. Processing was performed using the TOPAS 3.0 program with Ritveld parameter refinement.

Calvet microcalorimetric method was used to study the specific power losses of magnetically filled polymer composites with varying uniform magnetic fields. The commercial microcalorimeter was modified using solenoids with a number of turns of 12,000 per meter, which were placed in the calorimeter cells and connected in series to an alternating current source of up to 146 mA, as shown in Figure 4(a). The peak current in the circuit was controlled by a DS203 oscilloscope with a standard resistance of 1 ohm, connected in series to the coils. The field strength was 1750 A /m. The frequency of the sound wave generator was 214 kHz. The sensitivity of calorimetric cells was 10^{-6} J / sec.
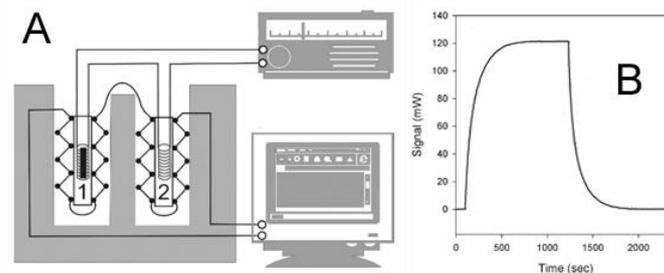
Figure 4. Experimental setup for hyperthermia measurements. A – scheme of the laboratory installation; 1 – working cell with magnetic coil and a specimen of epoxy composite; 2 – empty reference cell with magnetic coil. B – typical time plot of calorimeter signal in hyperthermia experiment – epoxy composite with 70% of nano-Fe particles at 214 kHz.

Figure 4 (b) shows the example of a heat release curve registered by calorimetry. Before to the measurement the baseline was monitored for at least 20 min. Then, as the wave generator was turned on, the calorimetric signal increased due to the heat evolution in the cell. Because of the differential scheme of the connection of thermocouples in the calorimeter the signal corresponds to the difference of the heating power occurred in the cell with sample and in the control cell. During this phase the signal eventually came to saturation after exponential growth. After it was monitored for at least 10 min, the wave generator was turned off, and the attenuation of the signal to the initial baseline was recorded to establish the reproducibility of the initial conditions. The difference between baseline signal and saturation signal is called specific loss power (SLP) and represents the re-magnetization

heating intensity. The absolute experimental error for measurements was less than 5 mW, or around 12 mW/g for the specific heating power in an experiment.

3. Experimental

The calorimetric method was used to obtain the direct heating power of re-magnetization in alternating magnetic field for samples of micro-Fe, nano-Fe with different compression ratio, epoxy composites with different volume content of particles and a wire sample. SLP data for all the samples are compiled in Table 1.

Table 1. Specific power losses of samples with different volume fraction of Fe

Sample	vol fract	SLP,mW/g
Nano-Fe powder	0.12	270
	0.18	303.7
	0.22	334
	0.25	371.3
Nano-Fe+KDA composite	0.02	222.1
	0.04	241.7
	0.06	239.1
	0.09	276.3
	0.19	330.1
	0.26	397.7
Micro-Fe powder	0.50	827
Micro-Fe+KDA composite	0.02	440.6
	0.06	390.3
	0.13	587.1
	0.26	816.1
Fe wire	1.00	4239

Selected data from Table 1 are shown on Figure 5.
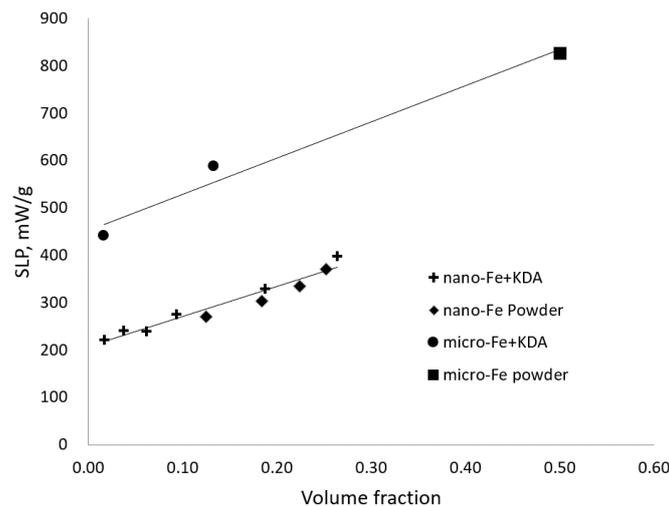
Figure 5. Specific thermal power losses per 1 g of Fe in remagnetization of different magnetic samples in the alternating field 214 kHz, 1750 A/m, depending on the volume fraction of Fe.

It is evident from Figure 5 that specific thermal power losses per 1 g of Fe linearly increase with volume fraction of MPs in the sample both for nano-Fe and micro-Fe. SLP of press-packed MPs and epoxy composites with MPs fit the same dependence both in case of nano-MPs and micro-MPs. The regression equations are y = 635x+206 for nano-Fe and y = 764x+452 for micro-Fe, where « y » is SLP in mW/g and « x » is the volume fraction of MPs. The intercept of linear regression is an extrapolation for the SLP value without interparticle interaction, as if it would be a single particle. The dependency between SLP and volume fraction is strong, as the slope of linear regression is higher than the intercept for both trends. That leads to a conclusion that the magnetic influence is of greater importance than the heating in the particle itself. The slopes of dependencies are fairly close to each other – it means that the influence of interparticle interaction is quite universal for different particle size. Meanwhile, micro-Fe MPs show up substantially higher power losses than nano-Fe MPs

To understand the dependence between particle size and SLP, the solid Fe wire with 100% volume fraction of Fe was taken into consideration as a reference system. SLP of Fe wire was 4239 mW/g (Table 1). Due to geometrical restrictions it is not possible to obtain ensemble of particles with 100% volume fraction of Fe even for multimodal distributed particles. Meanwhile, it is possible to estimate the apparent value for SLP for 100% volume fraction of MPs by the extrapolation of the trends in Figure 5 to 100% volume fraction. Such extrapolation gives apparent values 1216 mW/g for micro-Fe and 841 mW/g for nano-Fe at 100% volume fraction. Comparison of this estimation with the experimental data on solid Fe wire gives a way to explain the dependence between particle size and SLP.

Specific loss power in magnetic particles occurs due to relaxation of their magnetic moment. Nano-Fe samples consist of two major subgroups of particles. Larger ones (with diameter higher than 50 nm) have a complex vortex-like structure [12] instead of domains. Still, this structure has borders between areas with different magnetization, that emit heat while moving during re-magnetization process [13]. Smaller particles (with diameter less than 50 nm) are single domain, and their re-magnetization is governed by Neel effect [10], which appears to be low at the frequency of 214 kHz. Solid wire emits substantially more energy in the re-magnetization process because of uninterrupted Eddy currents inside. This type of heat dissipation needs a long conductive route, compared to particle diameter, which might inconsistently occur in micro-Fe MPs aggregates [11], The main source of SLP in case of multi-domain micro-Fe is, however, the relaxation of magnetic moment that happens due to magnetic domain shift. All these effects are frequency and size dependent, and for the present systems Neel relaxation effect and Eddy current heating effect contributions are low. As a result, nano-Fe MP's power dissipation mechanism is likely vortex structure border sweeping for the particles that are larger than 50 nm, while for multi-domain micro-Fe MPs re-magnetization process is governed by sweeping of domain walls, which apparently gives higher SLP values.

4. Conclusions

Calvet microcalorimetry method was used to study re-magnetization loss power in magnetic filled polymer composites nano-Fe+KDA, micro-Fe+KDA and in mechanically packed powders of nano-Fe and micro-Fe in an alternating magnetic field 214 KHz, 1750 A/m. The heat dissipation power varied from 200 to 400 mW/g for nano-Fe powder and composites, reaching a maximum value of 400 mW/g at a maximum volume fraction of Fe of 26%. For micro-Fe power varied from 450 mW/g to 816 mW/g with a maximum value at a maximum volume fraction of 50%. A linear dependence of the heat power on the volume fraction of iron was found. It was shown that specific loss power is dependent on particle size and packing ratio, but is not dependent on the existence of polymer matrix. This indicates a significant effect of the magnetic interaction of particles on heat losses during electromagnetic absorption. The effect associated with magnetic interaction is comparable in order of magnitude with the thermal effects of the magnetization reversal of individual particles.

Acknowledgments

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