Electromagnetic Properties of Composite Materials Based on Nd-Compound Modified Fe-Si-Al Alloy

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Abstract. The investigation of microstructure, optical and electromagnetic properties of powder prepared from Nd-compound doped Al6Si9Fe85 (Alsifer) alloy. The result of energy-dispersive X-ray spectroscopy is an evidence that the complementary Nd-containing Nd-Fe phase is on the surface of Alsifer particles. The magnetic particles distribution histogram in powder after mechanical treatment was plotted and analyzed. The electromagnetic properties (components magnetic permeability and dielectric permittivity) for produced composites with Nd-compound doped Alsifer powder with 50-90% (wt.) was studied. The nonlinear concentration dependence of magnetic and dielectric permittivity versus concentration Nd-compound doped Alsifer particles was observed. The magnetic resonance in composites was detected that shifted in low frequencies with increased magnetic filler concentration. The resonance field, absorption linewidth, g-factor and microwave absorption ability of Nd-compound doped Alsifer powder were determined from FMR experiment.

1. Introduction

The composite materials with magnetic fillers are frequently used in construction of electromagnetic shields, for microwave emission protection, for electromagnetic compatibility of electronic technical devices, and as component of microwave absorption materials [1–6]. The electromagnetic parameters which are magnetic ($\mu = \mu' + i\mu''$) and dielectric ($\varepsilon = \varepsilon' + i\varepsilon''$) permittivity’s are major parameters for estimation of ability of dispersed composite materials for microwave absorption. The magnetic soft materials are suitable for high values of magnetic permittivity for composites in ultra-high frequency (UHF) range. The alloy Fe-Si-Al family is one of the main ferromagnetic soft magnetic materials used as magnetic powder fillers for composite materials [7–12]. Doping or modification of AlSiFe alloy is way to improve electromagnetic characteristic to microwave electromagnetic waves range [13–18].

In this work, for the powder of a commercial Al6Si9Fe85 alloy modified by the addition of Nd compounds, the microstructural, optical, and electromagnetic properties were investigated. The magnetic and dielectric properties for manufactured composite materials with different concentrations of the investigated magnetic powder filler in paraffin matrix were studied depending on the frequency of the applied electromagnetic field.

2. Materials and measurement

The commercially available highly permeable ferromagnetic Nd-compound modified FeSiAl alloy (China, Chengdu Huarui Industrial Materials Co., Ltd.) was used in this study. The alloy sample was previously mechanically ground, followed by grinding in a ceramic mortar for 1 hour at room
temperature in ethanol. Next, the obtained powder was transferred to filter paper and dried in a vacuum oven for 2 hours at 40 °C.

Micrographs of the Nd-compound modified AlSiFe alloy powder were obtained using a “JEOL JSM – 7500F” scanning electron microscope by mean of the back-reflected electrons detection mode (COMPO). The energy dispersive analysis (EDA) method was used to study the elemental composition of the test sample using a “INCA X-Sight” energy dispersive X-ray spectrometer.

The optical diffuse reflectance spectra of the investigated alloy powder were measured on a “Hitachi U-3900” spectrophotometer with a two-channel integrating sphere in the range 300–900 nm.

The powder EPR spectrum was obtained at room temperature (20 °C) on “JEOL JES FA-300” EPR spectrometer at a frequency of 9.14 GHz with a modulation frequency of 100 kHz.

To determine the magnetic and dielectric properties of the studied Nd-compound modified AlSiFe alloy, AlSiFe/paraffin composites were prepared by means of hot pressing in special brass fixture with a concentration of magnetic filler of 50–90% in increments of 10% by weight. The studied composite materials were made in the form of a toroid with dimensions of an inner diameter of 3.05 mm and an outer diameter of 7 mm. The scattering parameters (S-parameters) of the prepared composites in a coaxial cell were measured using a “Deepace KC901V” vector network analyzer in the transmission line mode in 0.015–7 GHz range.

3. Results

Figure 1 shows the EDA spectra for a particle of the studied magnetic powder. According to the obtained results, it follows that the main fraction of particles contains Fe, Si and Al chemical elements (Figure 1, a) that correspond to the AlSiFe magnetic alloy under study. However, a detailed study of the surface of the powder in the detection mode of back-reflected electrons showed the microparticles whose contrast shade was significantly different from the main powder under study. The additional elemental analysis of the selected particle was carried out, according to the results of which it follows that on the surface of the studied Fe-Si-Al alloy an Nd and Fe elements are present. The fractions of Nd and Fe elements corresponds to Nd2Fe14B chemical compound.

Figure 1. Energy dispersive analysis spectrum with a selected analysis zone for (a) Fe-Si-Al powder and (b) Nd-compound particles.

Microphotograph of the obtained powder of Nd2Fe14B modified AlSiFe alloy is presented on figure 2a, a. As can be seen from the figure, a large fraction of the particles of the sample under study represents an irregular fragmented and stone-like shape. Based on the obtained photographs the histograms of the particle size distribution of the powder under investigation were calculated depending on their percentage using the “ImageJ” program (figure 2b). The obtained data revealed that the ground sample has a wide particle size distribution in the range from 10 to 115 microns. The main fraction of particles is in the range of 11–46 microns, the total percentage of which is more than 60% of the total.

Figure 3 shows the optical diffuse reflectance spectrum for ground Nd2Fe14B modified AlSiFe alloy in the range 300–900 nm. The magnetic powder has a light reflection of not more than 20% in investigated spectral range. There is a gradual decrease in light reflection from 17% at λ = 780 nm to 12.7% at λ = 380 nm.
The value of the reflectance minimum for the AlSiFe modified Nd micropowder under study is in the range of 358–418 nm, not exceeding 13%.

The values of magnetic permeability ($\mu = \mu' + i\mu''$) and dielectric permittivity ($\varepsilon = \varepsilon' + i\varepsilon''$) in a complex form for fabricated composite materials based on the studied powder of Nd$_2$Fe$_{14}$B modified AlSiFe alloy were calculated from experimentally measured S-parameters values according to the Nicholson-Ross-Weir algorithm [19–21].

The values of real and imaginary parts of magnetic permeability ($\mu'$, $\mu''$) in the studied frequency range are presented in Figure 4. As can be seen from the data in Figure 4, a, in the low-frequency region an increase in $\mu'$ is observed with increasing filler concentration ($\mu'_{50\%}=1.90$, $\mu'_{60\%}=2.41$, $\mu'_{70\%}=2.83$, $\mu'_{80\%}=3.40$, $\mu'_{90\%}=4.71$ at 20 MHz). A sharp decrease in $\mu'$ value is observed at a frequency of $\approx 1$ GHz, for all studied composite materials as the frequency of electromagnetic radiation increases. Based on the data of Fig. 4b, for $\mu''$ values there is a peak in magnetic losses, which corresponds to the resonant frequency. A shift of the resonance to the low-frequency region was observed with an increase in the filler concentration ($f_{r,50\%}=1.65$ ГГц, $f_{r,60\%}=1.55$ ГГц, $f_{r,70\%}=1.1$ ГГц, $f_{r,80\%}=0.56$ ГГц, $f_{r,90\%}=0.19$ ГГц).

In addition, it should be noted that the authors of [22] investigated the magnetic properties of composite materials based on ferromagnetic alloys. The analysis of the data of [22] revealed that magnetic resonance is also observed for composite materials based on plate-like AlSiFe particles in the UHF region. As can be seen from the high-frequency spectra obtained in [22], the $\mu'$ and $\mu''$ values are significantly higher than for our values for the composite materials investigated in this work. The main reason may be the high uniformity of the structure of the plate-like particles of AlSiFe alloy used in [22], which entails a uniform distribution of magnetic particles inclusions in the composite material [23].
The frequency dependence of magnetic permeability components for composite based on Nd$_2$Fe$_{14}$B modified AlSiFe in paraffin is shown in Figure 4. The dependences of the dielectric constant parts ($\varepsilon'$ and $\varepsilon''$) for the investigated composites are shown in Figure 5 (a, b). According to the experimental data, it can be seen that with an increase in the magnetic filler concentration, both $\varepsilon'$ and $\varepsilon''$ increase for all the samples under study. It can be seen that the $\varepsilon'$ values remain almost unchanged in the entire investigated frequency range for composite materials with Nd$_2$Fe$_{14}$B modified AlSiFe filler concentrations of 50–80% ($\varepsilon'_50% \approx 3.6$, $\varepsilon'_60% \approx 4.5$, $\varepsilon'_70% \approx 5.95$, $\varepsilon'_80% \approx 9.8$). However, for investigated composite material with a concentration 90% of the studied magnetic powder a decrease in $\varepsilon'$ is observed from 24.3 to 20.8. Based on the analysis of dielectric permittivity parts data ($\varepsilon'$ and $\varepsilon''$) in the studied frequency range, we can conclude that there are no relaxation processes for the manufactured composite materials.

The EPR spectrum of the studied Nd$_2$Fe$_{14}$B modified AlSiFe powder is shown in Figure 6. The resonance field $H_r$, the absorption line width $\Delta H$ (from peak to peak), and the intensity of the first derivative of the absorption signal $I$ were determined from the EPR spectrum. The EPR spectrum has the following parameters: $H_r = 115.56$ mT, $\Delta H = 138.18$ mT, $I = 1662$ a.u. The $g$-factor value was calculated by the formula (1):

$$g = \frac{h \nu}{\mu_B \cdot H_r}$$

where $h$ – Planck constant ($6.626 \times 10^{-34}$ J·s), $\nu$ – EPR spectrometer electromagnetic frequency (9.14 GHz), $\mu_B$ – Bohr magneton ($9.274 \times 10^{-24}$ J/T), $H_r$ – resonance field (115.56 mT). The $g$-factor calculated by formula (1) has a value $g = 5.65$. 
Figure 6. EPR spectrum of the studied Nd$_2$Fe$_{14}$B modified AlSiFe powder.

According to the study [24], the g-factor for Fe compounds can vary from 4 to 6. Based on this, we can conclude that the EPR signal for the Nd$_2$Fe$_{14}$B modified AlSiFe powder under study gives Fe ions in materials structure. It is also worth noting that composite films based on AlSiFe powder with various concentrations in epoxy resin were studied in [25]. By analyzing the FMR spectra of the magnetic AlSiFe-based composite films from [25], we can conclude that magnetic phase A [25] has a similar EPR spectrum shape as the Nd$_2$Fe$_{14}$B modified AlSiFe powder in present study.

4. Conclusion

As a result of the study the data on the microstructure, chemical composition, optical diffuse reflectance properties of the Nd$_2$Fe$_{14}$B modified AlSiFe alloy the powder after mechanical machining were obtained. The analysis of the calculated magnetic permeability for composites with Nd$_2$Fe$_{14}$B modified AlSiFe alloy powder as magnetic filler showed rather high values in the studied frequency range. Magnetic resonance in frequency range 0.2–3 GHz was detected for Nd$_2$Fe$_{14}$B modified AlSiFe powder-based composite materials under study. Data analysis showed the dependence of the microwave absorption resonance frequency on the concentration of the investigated AlSiFe powder filler. Based on the obtained electromagnetic characteristics, it can be concluded that studied Nd$_2$Fe$_{14}$B modified AlSiFe powder filler is a promising material for development of microwave absorbing materials in the low frequency range of 0.2–2 GHz.

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