High frequency properties of ferrite/Fe-Si-Al alloy soft magnetic composites

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Abstract
The inclusion of Fe-Si-Al alloy particles in NiCuZn ferrite matrix was investigated with regard to the high frequency electromagnetic properties (complex permeability and permittivity). The resultant composites of relatively low density exhibit a shift of the permeability spectra to higher frequencies and an increase of dielectric polarization, which finally favour the electromagnetic wave attenuation at microwave frequencies. Thus, wider band return loss peaks are attained at frequencies above 6 GHz by thinner composite materials.

Keywords: Ceramic-alloy composites, Fe-Si-Al, NiCuZn ferrites, complex permeability, microwave attenuation

1 Introduction

The need for shielding population, facilities and equipment against electromagnetic waves of increasingly high frequency has driven the extensive research on materials with the proper electromagnetic behaviour. In this direction, polymer composites with magnetic or conductive fillers predominate in the respective studies and applications due to the offered flexibility in design and tuneability of performance [1-3]. Among various types of inclusions, soft magnetic oxides (NiZn or MnZn spinel ferrites) or Fe-rich alloys are widely and effectively used either separately or in conjunction for the preparation of microwave absorbing composites in the above 1 GHz region [4-7]. It is important to note that, the dispersion of the constitutive electromagnetic properties in composite materials allows the employment of spinel ferrites and ferromagnetic alloys in the microwave regime, despite their conventional competence at lower frequencies.

Although the ceramic-metallic composite materials (commonly referred to as “cermets”) are expected to share some of the advantageous characteristics of polymer composites, there is a deficiency of respective studies. Thus, the different approach of the present investigation lies in the...
synergy of ferrimagnetic and metallic materials, by the incorporation of Fe-based alloy particles in a ferrite ceramic matrix, in order to form rigid soft magnetic composites as efficient electromagnetic absorbers.

2 Sample preparation and experimental procedure

The selected soft magnetic composites for this investigation basically comprise a Cu-substituted NiZn ferrite and Fe-Si-Al alloy powder. In specific, the ferrite powder (F) with nominal composition $\text{Ni}_{0.3}\text{Zn}_{0.6}\text{Cu}_{0.1}\text{Fe}_2\text{O}_4$ was prepared with the conventional solid-state reactions technique, which involved the wet mixing of the precursor oxides/carbontes for 3 hours, prefireing at 1000°C for 2 hours in air and ball milling for 3 hours. The milled powders were then annealed at 1100°C for 4 hours before the final milling process for 3 hours. Regarding the used Fe-Si-Al alloy (A), it is a commercial sendust grade with the composition Fe$_3$Si$_{0.7}$Al$_{0.3}$ and it was sieved below 45 $\mu$m. The two constituent fine powders were mixed at different weight ratios (F/A: 100/0, 90/10, 80/20) to form the respective composite samples denoted for brevity as “A0”, “A10” and “A20”. For the mechanical reinforcement of the composites, the powder mixtures were doped with 2wt% of a glass compound and 5wt% of polyvinyl alcohol solution. The prepared mixtures were compacted under 200 MPa in disc shape and finally annealed at temperatures varying from 700°C to 900°C for 30 minutes in air. The upper temperature limit serves to avoid the extended oxidation of the alloy particles.

Among the main objectives is to identify the optimal production process of the specific ceramic-alloy composites with regard to their electromagnetic performance, thus the materials were characterized as to their density and their microstructure was recorded by scanning electron microscopy (JEOL JSM6300). For the electromagnetic characterization of the specimens, ring-shaped samples were machined to fit in a coaxial sample holder and their complex permeability $\mu^*$ and permittivity $\varepsilon^*$ were measured in the frequency range 1 MHz-10 GHz, by means of impedance and network analysis (Agilent E4991A RF Impedance-Material Analyzer, Agilent 8720C Network Analyzer). The knowledge of these properties has allowed the calculation of the return losses ($RL$ in dB) for a single metal-backed layer as a function of thickness in the 1-10 GHz region.

3 Results

The post-annealing densities of the prepared ferrite/alloy composites, demonstrated in Figure 1, indicate an increasing trend with annealing temperature ($T_a$), especially for $T_a=900°C$. This is basically ascribed to the offset of densification of the Cu-doped ferrite particles, which is further supported by the glass compound acting as a sintering aid. The observation of composites’ microstructure by SEM revealed that the recorded densification is not related to the occurrence of the mechanism of ferrite grain coarsening (Figure 2). Besides, at this early stage we rather expect the initial interparticle neck formation than grain growth to take place. Still, the attained values remain well below the equivalent theoretical densities (relative densities: 72-77%). These low post-annealing densities are attributed to the relatively low press density and the absence of a strong densifying mechanism in this temperature range, such as the ferrite diffusion towards the pores.
3.1 Complex permeability and permittivity

The complex permeability spectra ($\mu^* = \mu' - j\mu''$) of the produced composites, as shown in Figures 3-5, exhibit a single and wide dispersion-type peak, which probably encompasses the contribution of different dynamic magnetization mechanisms (domain wall bulging and magnetization rotation). Thus, we observe that the addition of Fe-Si-Al alloy generally lowers the permeability level and shifts the loss peak towards higher frequencies. This is additionally evidenced by the variation of the characteristic frequency at maximum $\mu''$ ($f_{\text{MAX}}$), which is displayed in Figure 6. The experimental data indicate that the effect of permeability degradation is intensified for $T_a = 900^\circ$C, whereas the maximum frequency shift occurs in the composites annealed at 700°C.

The graphs in Figure 7 depict the complex permittivity spectra ($\varepsilon^* = \varepsilon' - j\varepsilon''$) of the composites annealed at 700°C in the 1-10 GHz region, where for both parts $\varepsilon'$ and $\varepsilon''$ no dispersion with frequency is observed. Additionally, the real parts of permittivity of the composites annealed at 800°C and 900°C are displayed in Figure 8. In all cases, the inclusion of alloy particles in the ferrite matrix yields the enhancement of dielectric polarization, which is mainly demonstrated by the real part spectra as the recorded differences in $\varepsilon''$ are on the level of the measurement accuracy. It is also important to notice that, with the exception of A10, permittivity of A0 and A20 is raised by annealing at higher $T_a$. 

Figure 1: Post-annealing density ($d$) of ferrite/alloy composite materials.

Figure 2: SEM images of the composite A0 annealed at (a) 700°C, (b) 800°C and (c) 900°C (scale length ~10 μm).
Figure 3: Real and imaginary part of permeability of A0, A10 and A20 composites annealed at 700°C.

Figure 4: Real and imaginary part of permeability of A0, A10 and A20 composites annealed at 800°C.

Figure 5: Real and imaginary part of permeability of A0, A10 and A20 composites annealed at 900°C.
Figure 6: Variation of $f_{\text{MAX}}$ (frequency of max $\mu''$) with composition and annealing temperature.

Figure 7: Complex permittivity of A0, A10 and A20 composites annealed at 700°C.

Figure 8: Real parts of permittivity of A0, A10 and A20 composites annealed at 800°C and 900°C.
3.2 Return losses

For the evaluation of the new composites in microwave attenuation, we have calculated the return losses ($RL$) in the 1-10 GHz range, by applying the transmission-line theory for the case of a single metal-backed layer. Actually, the contours of $RL$ are plotted as a function of frequency and material thickness and depicted in Figures 9-11 for the different applied annealing temperatures. From these figures, we derive that the presence of sendust alloy in the test composites A10 and A20 results in the extended bandwidth of the recorded absorption peaks. Particularly, the narrowband peaks of A0 at 6 GHz shift to higher frequencies and wide peaks with $RL$ higher than 20dB (99% power decay) now cover the band from 6.5 GHz to 10 GHz. Moreover, we notice that the alloy-doped composites require a decreased layer thickness in order to achieve minimum reflections. Thus, we deduce that the underlying impedance matching, which occurs for thicknesses close to a quarter-wavelength, is favoured by the low but non negligible magnetic losses $\mu''$ in combination with the high dielectric constant as it is also analyzed in [8].

**Figure 9:** Return Losses of A0, A10 and A20 composites annealed at 700°C.

**Figure 10:** Return Losses of A0, A10 and A20 composites annealed at 800°C.

**Figure 11:** Return Losses of A0, A10 and A20 composites annealed at 900°C.
4 Conclusion

New ferrite/alloy composites were prepared in order to investigate the synergy between the two magnetic components, with a view to their high frequency electromagnetic properties. Specifically, the addition of alloy particles has caused the suppression of permeability levels and the displacement of the respective spectra to higher frequencies. Additionally, the ferrite/alloy interaction has generally enhanced the dielectric properties of the composites. The observed non-linearities in the described effects should be further investigated in terms of the morphological and structural characteristics of the composites. Yet, the induced variations of the constitutive parameters ($\mu^*$ and $\varepsilon^*$) were found to promote the plane wave attenuation at microwave frequencies above 6 GHz.

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