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Structure and microwave-absorbing properties of Fe-particle containing alumina prepared by micro-arc discharge oxidation

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Abstract

Alumina films containing Fe particles were produced on aluminum alloy substrates by micro-arc oxidation with additive Fe particles in the electrolyte. The permittivity–frequency and permeability–frequency properties were determined in the microwave frequency regime of 6.5–18 GHz by vector network analysis. Scanning electron microscopy (SEM), X-ray diffraction (XRD), and X-ray fluorescence (XRF) were employed to investigate the microstructures and compositions. Our results show that about 16.16 wt.% Fe particles are embedded in the alumina films and the acquired complex permittivity and permeability values match the microwave frequency. When the matching thickness is 2 mm, the minimum RL values are −10.5 dB at 9.6 GHz and −9.1 dB at 16.3 GHz. The results suggest a new design of microwave absorbers based on electromagnetic wave-absorbing materials that eliminate the use of a mixture of agglutinants in the preparation.

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Keywords: Alumina films; Fe particles; Complex permittivity and permeability

1. Introduction

Eelectromagnetic wave-absorbing materials are being investigated as anti-electromagnetic interference coatings in self-concealing technology, and properties such as dielectric loss, conductive loss, and magnetic loss have been reported in the literature [1–8]. These materials include metal oxides, alloy–epoxy composites, and especially metallic magnetic materials which have generated particular interest. Because of their desirable complex permeability ($\mu' - j\mu''$) and permittivity ($\varepsilon' - j\varepsilon''$) which determine the reflection and attenuation characteristics, metallic magnetic materials such as Fe and Co are some of the suitable candidates for applications in GHz electromagnetic wave absorption because these materials possess high magnetization values at high frequencies [4,5]. Conventionally, these materials are fabricated using agglutinants. For example, a common method is to use small particles isolated from insulating materials such as epoxy resin.

In the paper, we report a novel fabrication technique utilizing micro-arc discharge oxidization (MDO) to fabricate

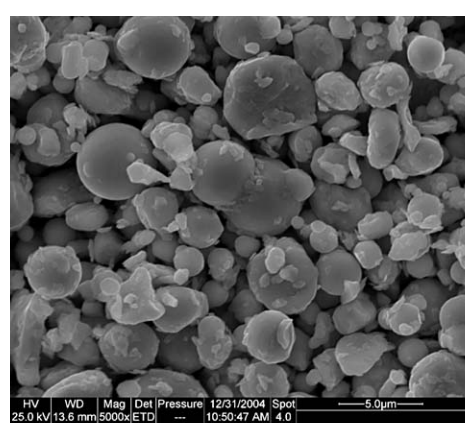

Fig. 1. SEM micrograph of the Fe particles.

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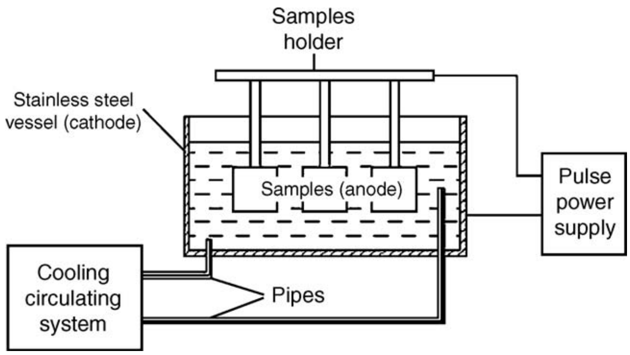

Fig. 2. Schematic of the MDO apparatus.

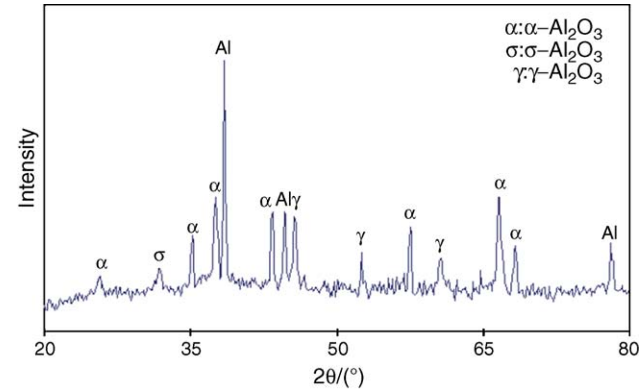

Fig. 4. XRD spectra of MDO coating.

electromagnetic wave-absorbing coatings without the use of agglutinants. Utilizing micro-arc discharge, diffusion, and electrophoresis, various kinds of conventional ceramic coatings containing adventitious elements have been synthesized on metal substrates such as Al, Ti, Ta, Mg and their alloys [9–13]. Electromagnetic wave-absorbing coatings can be formed by incorporating electromagnetic wave-absorbing materials in the electrolyte. In this work, Fe particle-containing alumina films were produced on aluminum alloys by micro-arc discharge oxidation using additive Fe particles in the electrolyte. The permittivity–frequency and permeability–frequency of the films were evaluated in the microwave frequency regime of 6.5–18 GHz employing vector network analysis. Scanning electron microscopy (SEM), X-ray diffraction (XRD), and X-ray fluorescence (XRF) were employed to investigate the microstructures and compositions. The electromagnetic properties and reflection loss in the GHz regime loss were studied.

2. Experimental details

Round disks (20 mm diameter, 4 mm thickness) and rectangular specimens (100 mm × 100 mm × 4 mm) made of LY2 aluminum alloys consisting of 2.6–3.2% Cu, 2.0–2.4% Mg, 0.45–0.7% Mn, and <0.8% impurities were mechanically polished prior to micro-arc oxidation. An aqueous solution of electrolytes was prepared using $NaWO_3$, $Na_3PO_4 \cdot 12H_2O$ and wave-absorbing Fe particles (Fig. 1), and the apparatus is schematically shown in Fig. 2. The output of the power supply was connected to the bath, and another one to the samples immersed in the electrolyte. The applied voltage was varied from 300 to 450 V DC. An average current density of 8 A/dm^2 was maintained on the surface by controlling the voltage and the electrolyte temperature was less than 50 °C. The coating thickness is about 50 μm.

The phase of the coating was characterized by glancing-angle (2°) X-ray diffraction using the CuK_α line at 30 kV. The surface morphologies were observed using scanning electron microscopy (SEM), and X-ray fluorescence spectrometry (XRF) was used to determine the elemental concentrations. The electromagnetic properties of the coatings were investigated based on the reflection coefficient (S_{11} parameter) and transmission coefficient (S_{21} parameter) measured by a network analyzer at a frequency range of 6.5–18 GHz. The tested coatings were peeled off from the large MDO specimens, pressed, and cut precisely to the size of 10.1 mm × 22.9 mm corresponding to the cross section of the waveguide. It was then inserted into the waveguide to measure the S-parameter using the HP-8520B network analyzer. The complex permittivity

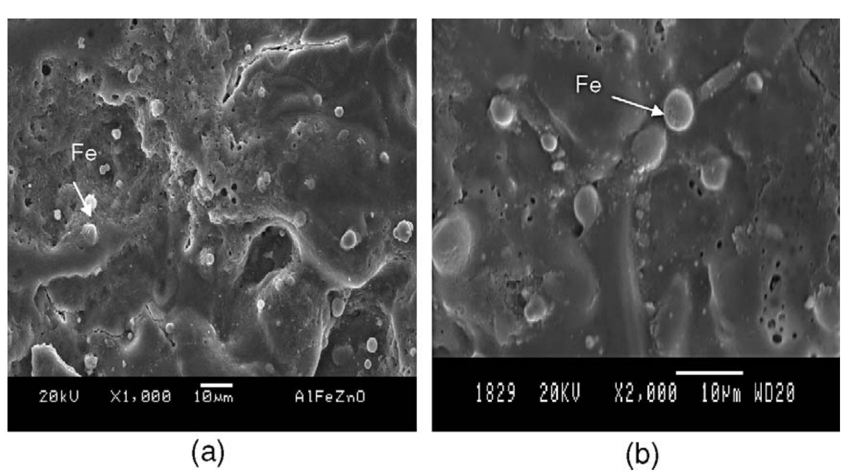

Fig. 3. SEM micrographs showing the surface morphologies of the Fe-containing MDO coatings: (a) low magnification, (b) high magnification.

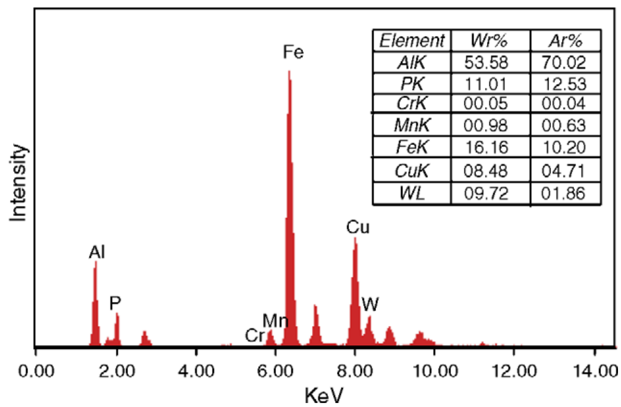

Fig. 5. Elemental concentrations of the Fe-containing MDO coatings determined by XRF.

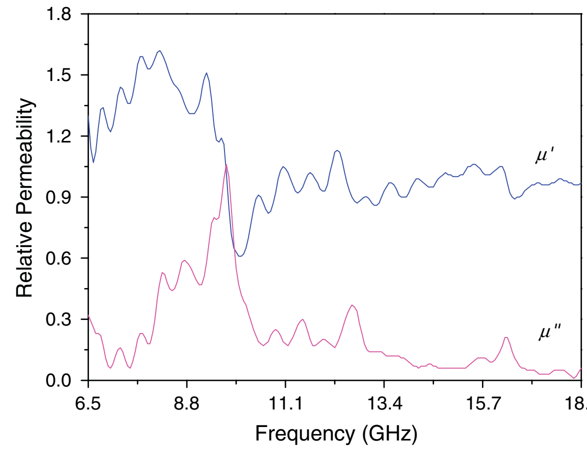

Fig. 7. Complex permeability of the coatings.

($\varepsilon = \varepsilon' - j\varepsilon''$) and permeability ($\mu = \mu' - j\mu''$) of the coatings were calculated from the S_{11} and S_{21} parameters.

3. Results and discussion

Because the electrolyte contains Fe particles, the Fe particles will participate in the reaction occurring in the micro-arc discharge channels and be incorporated into the coatings by diffusion and electrophoresis during MDO. The SEM photographs for the Fe-containing MDO coated samples are depicted in Fig. 3. It can be observed that many Fe particles several micrometers in size are randomly distributed on the surface. Some of them extend throughout the alumina films. Meanwhile, many micro-pores are formed by the micro-arc discharge process. Fig. 4 shows the X-ray diffraction patterns of the MDO-coated samples indicating the presence of α-Al_2O_3, γ-Al_2O_3. Since the Fe particles are in a Fe-based solid solution state, no Fe precipitation is detected by XRD. The elemental concentrations determined by XRF are displayed in Fig. 5 and the weight percent of Fe is approximately 16.16%.

The frequency dependence of the complex permittivity shown in Fig. 6 shows small values of dielectric constant ($\varepsilon' \cong 2.6$) and negligibly small dielectric loss ($\varepsilon'' \cong 0.2$). Since the Al_2O_3 and embedded Fe particles often provide high values of dielectric constant, the films exhibit low relative permittivity due to micro-pores formed in the film. Therefore, the conduction loss is observed to be small. Fig. 7 shows the frequency dispersion of the complex permeability. Compared to the non-magnetic properties of ceramic coatings ($\mu_r' = 1$ and $\mu_r'' = 0$), our Fe particle-containing materials exhibit obvious magnetic properties. In particular, the increase in the magnetic loss (μ_r'') is evident. The values of μ' increase with frequencies from 1.1 to 1.62 in the range of 6.5–8.2 GHz, decrease from 1.62 to 0.6 in the range of 8.2–10 GHz, and then become constant at about 1 between 10 and 18 GHz. At the same time, μ'' initially increases from 0.32 to 1.06 in the range of 6.5–9.72 GHz and then decreases to 0.02 at frequencies up to 18 GHz. This behavior stems from the magnetic Fe fillers embedded in the ceramic coatings. According to the tangent values of dielectric loss ($\tan \delta_\varepsilon = \varepsilon_r''/\varepsilon_r'$) and magnetic loss angle ($\tan \delta_\mu = \mu_r''/\mu_r'$) shown in Fig. 8, obvious magnetic loss is observed at about 9.6 GHz and 12.6 GHz.

According to the transmission line theory, the reflection loss (RL) of normal incident electromagnetic wave at the absorber surface can be calculated from the relative permeability and

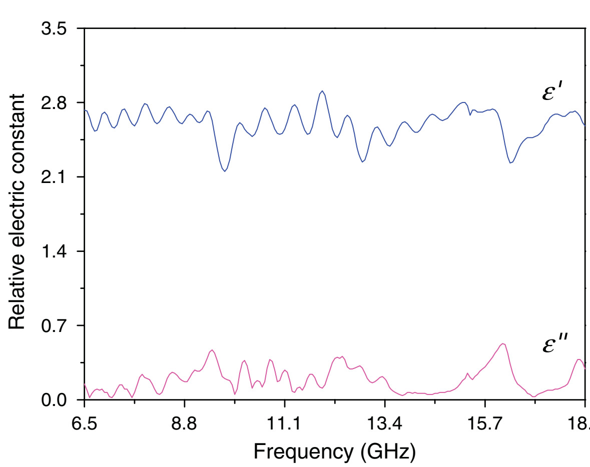

Fig. 6. Complex permittivity of the coatings.

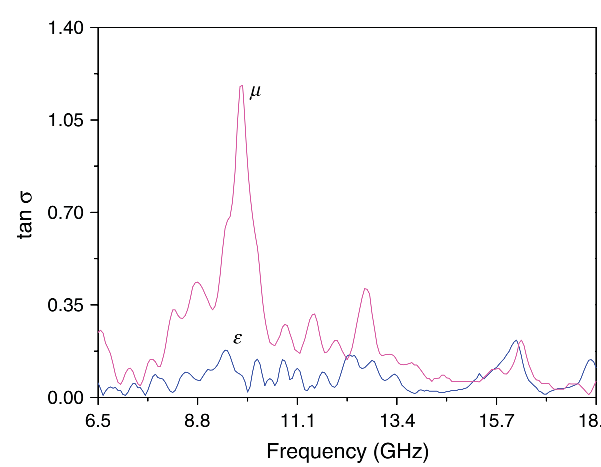

Fig. 8. Tangent values of dielectric loss and magnetic loss.

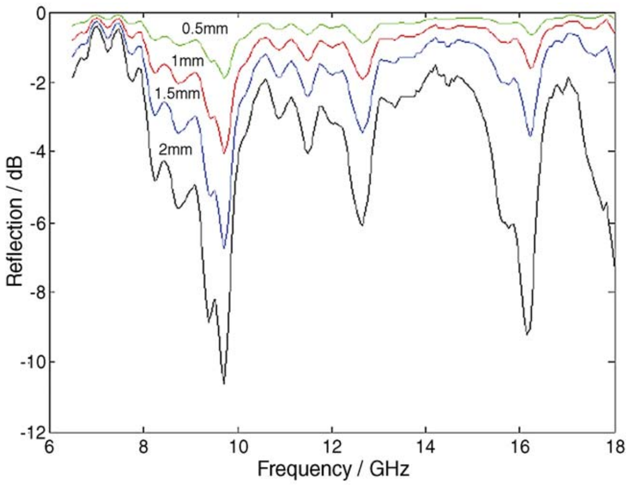

Fig. 9. Reflection loss (RL) of the Fe-doped films of various thicknesses.

permittivity at a given frequency and absorber thickness using the following equations [14]:

$$Z_0 = \sqrt{\frac{\mu_0}{\varepsilon_0}}\,, \tag{1}$$

$$Z_{in} = \sqrt{\frac{\mu}{\varepsilon}}\tanh\left(j\frac{2\pi fd}{c}\sqrt{\mu\cdot\varepsilon}\right)\,,\ \text{and} \tag{2}$$

$$RL = 20\lg\left|\frac{Z_{in}-Z_0}{Z_{in}+Z_0}\right|\,. \tag{3}$$

Here, Z_0 is the impedance of air, μ_0, ε_0 are the permeability and permittivity of air, respectively, f the frequency of the electromagnetic wave, d is the thickness of the absorber, c is the velocity of light, Z_{in} is the input impedance of the absorber, and μ and ε are the complex permeability and complex permittivity of absorber, respectively.

Fig. 9 shows the frequency dependence of the calculated reflection loss of the Fe-containing alumina films with thicknesses of 0.5, 1, 1.5, and 2 mm. It clearly shows that the reflection loss minimum depends on the thickness and the minimum reflection loss decreases with thickness. Three obvious absorption peaks located in 9.6, 12.6, and 16.3 GHz can be observed. When the thickness is 2 mm, the minimum RL value of −10.5 dB is observed at 9.6 GHz and −9.1 dB at 16.3 GHz.

In general, electromagnetic waves absorbers are required to have excellent reflection loss values and be thin to cater to military as well as civilian applications. Moreover, they must have good formability. The method described here satisfies these requirements and is projected to play an important role in the new design of microwave absorbers.

4. Conclusion

Alumina films containing approximately 16.16 wt.% Fe particles were formed on aluminum alloys by micro-arc oxidation by incorporating Fe particles in the electrolyte. The microwave frequency properties in the range of 6.5–18 GHz were determined by vector network analysis. The relative permittivity ($\varepsilon = \varepsilon' - j\varepsilon''$) exhibits small variations ($\varepsilon' \cong 2.6$, $\varepsilon'' \cong 0.2$), and the μ' and μ'' values of the relative permeability vary as a function of frequencies. When the thickness is 2 mm, the minimum RL value is −10.5 dB at 9.6 GHz and −9.1 dB at 16.3 GHz. This novel fabrication method opens the way for more effective and simpler design and synthesis of microwave absorbers in military and commercial applications.

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