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Magnetostriction and stress measurements in plasma-sprayed MnZn ferrite films

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The coercivity of the plasma-sprayed MnZn ferrite films decreases significantly after annealing in air at the temperature range of 500–800 °C. Magnetostriction and surface stress changes incurred in these ferrite films after the annealing process has been measured. It is found that the effects of the external stress on the magnetic properties of the plasma-sprayed MnZn ferrite films are minor. The coercivity mechanism arising from the magnetoelastic effects due to the nonzero magnetostriction has been analyzed to be small and is not dominant in controlling the magnetization process in these ferrite films. © 2005 American Institute of Physics. [DOI: 10.1063/1.1847431]

INTRODUCTION

With the advantages of bulk productivity, simple procedures, and high cost efficiency, plasma spraying of MnZn ferrites (MZF) offers a promising route for industrial applications in sensors and transformers. However, previous work has demonstrated¹ that the magnetic properties ($H_c = 120$ – 130 Oe) of the as-sprayed MnZn ferrite films are inferior compared to those of sintered MnZn ferrites ($H_c < 10$ Oe).² Although plasma-sprayed planar ferrite inductors may be employed in industrial applications,³ the high coercivity-induced hysteresis loss is considerable and may counteract the processing advantages.

It is found that the coercivity of plasma-sprayed ferrite films decreases to 80 Oe after heat treatment at 500 °C for 120 min in air and can even decrease to 10 Oe with an increased annealing temperature of 800 °C. As plasma-sprayed deposits contain relatively high residual stress,⁴ it is important to examine the effect of magnetoelastic factors on the coercivity mechanisms, which may evolve during the heat treatments. Magnetoelastic effects can be separated into two categories: (1) for small intrinsic stress created during the magnetization process due to nonzero magnetostriction, may be written as⁵ $h_c(3\lambda_s\sigma/4\pi M_s)$. Here λ_s and σ are saturation magnetostriction and stress, respectively, h_c is the reduced coercive force that has been numerically calculated.⁶ (2) For very large external stress, magnetoelastic Villari effect¹² due to the deformation of crystalline lattice may take place. The data obtained in the course of this study are used to estimate the magnetoelastic effects on the coercivity of these plasma-sprayed MnZn ferrite films.

EXPERIMENTS

Samples of MnZn ferrite films were plasma sprayed onto titanium substrates, with the conditions described in Ref. 1. The thickness of the ferrite films were in the range of 120–150 μm . The as-sprayed films were annealed in air at temperatures of 500 and 800 °C for up to 180 min. Phases present in the starting powder and in the ferrite films before

and after annealing were identified with x-ray diffraction (XRD) patterns obtained from a SCINTAG/PAD-V diffractometer using Cu $K\alpha$ radiation. The density of the free standing ferrite films was measured by with a helium pycnometer. The magnetic properties of the ferrite films before and after annealing were assessed with a vibrating-sample magnetometer at ambient temperature in a field range from -5000 to 5000 Oe. The magnetic field in the magnetic measurement is applied parallel to the film surface.

The field-induced strain (magnetostriction) of the MnZn ferrite films was measured at ambient temperature by using the laser deflection method. In this technique, bimorph cantilever samples in rectangular shape (3×2 cm²) were subjected to a uniform magnetic-field area produced by a pair of Helmholtz coils. The cantilever sample was clamped on one end and a He–Ne laser beam was reflected off the free end. A UDT SLC-5D linear position sensor⁷ was placed at 1.5 m away from the cantilever samples, which monitored the reflected beam position changes and are capable of detecting a beam displacement of 2 μm . The sensor output current was sent to a position-to-voltage converter, producing an output voltage linearly proportional to the beam position. Thus the smallest detectable sample deflection was approximately 40 nm. The cantilever samples were fixed with their surfaces perpendicular to the applied dc magnetic field, with a magnitude determined by a Gaussmeter. The deflection of the sample, Y , was converted to the in-plane strain by using the Stoney equation⁵ $e = Yt_s^2 E_s(1 - \nu_f)/2t_f L^2 E_f(1 - \nu_s)$, where t_s and t_f are the thickness of the substrate and film, respectively, E_s and E_f are the moduli of the substrate and film, ν_s and ν_f are the Poisson ratio of the substrate and film, respectively, and Y is the deflection of the sample at distance L from the clamped end. The values of E_s , E_f , ν_s , and ν_f appropriate to bulk titanium and MnZn ferrites were taken from the literature.^{8,9}

Free-standing films were made by peeling off the films from the substrates. The stress level in both freestanding and substrated ferrite films was determined by measuring the changes in crystalline lattice spacing⁴ obtained by x-ray diffraction. Due to the limited penetration depth of the x rays, only the surface stress in the ferrite films has been examined.

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FIG. 1. XRD patterns of (1) initial powder, (2) as-sprayed MnZn ferrite films, (3) ferrite films annealed at 500 °C for 120 min, and (4) ferrite films annealed at 800 °C for 120 min.

An elastic strain tensor was constructed from a set of lattice spacing obtained from different film orientations, which were converted to a stress tensor using Hooke's law. The in-plane stress was measured at points away from free edges.⁴ The XRD measurements were performed on a Bruker GADDS microdiffractometer with Cu K α radiation. The in-plane stress was determined by using the "sin² Ψ " method¹⁰ with a fixed 2θ at 89° that corresponds to MnZn ferrite (731) Bragg-peak location and seven Ψ orientations ranging from -45° to 45°.

RESULTS AND DISCUSSION

The XRD patterns of the initial powder and ferrite films (as-sprayed and annealed) are presented in Fig. 1. It is noted that the initial powder is a pure spinel phase while wustite FeO forms in the as-sprayed films [see pattern (2)] under the conditions of oxygen deficiency. After annealing the MZF films at 500 °C in air for 120 min, the wustite Bragg peaks disappear and hematite Fe₂O₃ peaks are observed in pattern (3), indicating oxidation concurrent with the decomposition of the metastable wustite phase. It can be seen that the XRD pattern (4) shows no significant difference from pattern (3) aside from a higher hematite Bragg-peak intensity, suggesting either more Fe²⁺ is oxidized to Fe³⁺ or a better crystallization in ferrite films after annealing at higher temperature.

The magnetic properties of ferrite films before and after annealing are summarized in Table I. The increase of the magnetization after annealing is a result of the cation ordering within the spinel lattice,¹ which is confirmed by the

TABLE I. Magnetic Properties of MnZn ferrite films after annealing in air.

T_{annl} (°C)	t_{annl} (min)	M_s (emu/cc)	H_c (Oe)
As-sprayed	0	205 ^{(1)c}	130 ⁽¹⁾
		210 ^{(2)d}	128 ⁽²⁾
500	120	358 ⁽¹⁾	80 ⁽¹⁾
		362 ⁽²⁾	80 ⁽²⁾
500	180	364 ⁽¹⁾	78 ⁽¹⁾
800	120	377 ⁽¹⁾	10 ⁽¹⁾
800	180	375 ⁽¹⁾	8 ⁽¹⁾

^a T_{annl} is the annealing temperature.

^b t_{annl} the annealing time.

^c(1) free standing films.

^d(2) ferrite films on titanium substrates.

FIG. 2. Field-induced strain (magnetostriction) in the plasma-sprayed MnZn ferrite films before and after annealing at 500 °C in air for 120 min. The ferrite films are on titanium substrates. The magnetic field is applied in the direction perpendicular to the film surface while the strain is measured in the direction parallel to the film plane.

neutron-diffraction study. It is also seen that the coercivity decreases from 128 to 80 Oe after annealing the MZF films at 500 °C in air, decreasing further to 10 Oe after annealing at 800 °C. The quantification of magnetoelastic effects in these films was carried out to examine the origins of these coercivity changes.

The field-induced strain (magnetostriction) measured in the direction perpendicular to the applied magnetic field is presented in Fig. 2. According to $\epsilon = \frac{3}{2}\lambda_s(\cos^2\theta - \frac{1}{3})$ with $\theta = 90^\circ$, the saturation magnetostriction is calculated to be 5.25×10^{-6} and 4.57×10^{-6} for MnZn ferrite films before and after annealing at 500 °C in air for 120 min, respectively. These values are comparable to that of bulk ferrites with similar composition.¹¹

The equation⁵ $h_c(3\lambda_s\sigma/4\pi M_s)$ can be rewritten as $h_c(3E\lambda_s^2/4\pi M_s)$, where E is the literature value of the modulus⁸ (160 GPa) of MnZn ferrites. Based on the magnetostriction data measured on the ferrite films on titanium substrates before and after annealing at 500 °C in air for 120 min, the coercivity difference, ΔH_c , due to the measured changes of λ_s and M_s is calculated to be 0.002 h_c . The upper limit value⁵ of h_c is 2, which gives a coercivity difference of $\Delta H_c < 0.004$ Oe. This value is much smaller than the measured coercivity change of ~ 50 Oe for ferrite films after annealing at 500 °C for 120 min, which suggests that the magnetoelastic effects due to nonzero magnetostriction are minor in these ferrite films.

The surface stress in the plasma-sprayed MnZn ferrite films determined by XRD is presented in Table II. A compressive stress exists in the surface of ferrite films on titanium substrates and tensile stress in both the top and bottom surfaces of freestanding ferrite films. The compressive stress in the ferrite films on the titanium substrates may result from the difference in thermal-expansion coefficients between the ferrite films and titanium substrates. When the film is released from the substrate, the net force imposed on it by the

TABLE II. Surface stress in plasma-sprayed MnZn ferrite films.

Samples	As-sprayed (on Ti sub)	Annealed (on Ti sub) ^a	As-sprayed (freestanding)	Annealed (freestanding) ^a
Stress (MPa)	-220 $\pm$ 30	-280 $\pm$ 30	80 $\pm$ 30 ^{(1)b} 95 $\pm$ 30 ^{(2)c}	40 $\pm$ 20 ⁽¹⁾ 45 $\pm$ 20 ⁽²⁾

^aAnnealed at 500 °C in air for 120 min.

^b(1) Top surface.

^c(2) Bottom surface.

substrate is removed. In this case, the tensile stress in both surfaces of the freestanding films may be balanced by the compression in the central part. This type of stress profile may originate from the nonlinearity of the stress distribution before substrate detachment.⁴ It is seen in Table II that the compressive stress in the ferrite films on titanium substrates increases after annealing the samples at 500 °C in air for 120 min while the tensile stress in the freestanding ferrite films decreases after the same annealing process.

The effects of the external stress on the magnetic properties of MnZn ferrites due to Villari effects have been reported.¹² The stress measurements indicate the existence of surface stress changes after the 500 °C annealing process for both freestanding ferrite films and ferrite films on titanium substrates, which may contribute to the observed coercivity changes. However, the differences between the magnetic properties of the two types of films are not large while the differences in the stress profiles are very different. This observation indicates that effects of external stress on the magnetic properties of the plasma-sprayed MnZn ferrites are likely small.

The coercivity changes in the ferrite films may be a product of changes in the compositional profiles and microstructures induced by annealing. The plasma-sprayed MnZn ferrite films are found to have an inhomogeneous composition¹ due to the preferential Zn evaporation during the spray process. The composition of the ferrite films homogenizes¹ after the annealing process due to cation diffusion, which in turn affects the magnetocrystalline anisotropy.¹³ Inhomogeneous composition of the as-sprayed ferrite films likely creates a position-dependent anisotropy variation, which sets up energy barriers that pin domain-wall movements.

CONCLUSION

The magnetoelastic effects on the magnetic properties of the ferrite films have been investigated. It is found that the magnetoelastic effects on the coercivity due to the nonzero magnetostriction are small in these ferrite films. The measurements of the surface stress on these ferrite films show very different stress profiles for freestanding and substrated ferrite films. However, the difference between the magnetic properties of these two types of samples is little, suggesting that the effects of large external stress on the magnetic properties of the plasma-sprayed MnZn ferrites are minor. The decrease of the coercivity during annealing may result from anisotropy changes arising from other origins, e.g., crystalline anisotropy.

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