

## Preparation and Characterization of FeAg Granular Alloys

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Fe<sub>10</sub>Ag<sub>90</sub> granular alloys have been fabricated, using a sol-gel method, for a range of nitric acid concentrations in the start solution. The samples have been characterized by X-ray diffraction and magnetization measurements. The average Fe particle sizes derived from X-ray diffraction are in the range 24-29 nm, indicating a large variation. The coercivity obtained from the hysteresis curve is two orders of magnitude larger than that of pure iron in all the samples. Moreover, the hysteresis curves do not saturate, even in fields of up to 1 T. These observations indicate that the samples contain both superparamagnetic and blocked particles. A comparison between the coercive field and the average particle diameter, determined by the Sherrer's formula, is displayed in all acid concentrations.

**Keywords:** *sol-gel, granular alloy, ferromagnetism*

### 1. Introduction

Granular magnetic materials composed of magnetic nanoparticles embedded in a nonmagnetic matrix show interesting physical properties, such as superparamagnetism (SPM), giant magnetoresistance and giant magneto-impedance<sup>1-3</sup>. Moreover, they have important technological applications in magnetic recording, in optical devices and in sensors<sup>4-7</sup>. Such materials have been produced as films by sputtering, as ribbons by melt-spinning, and in powder form by sol-gel methods<sup>8,9</sup> and mechanical alloying<sup>10</sup>.

The magnetic grain size of these materials varies from a few Å to hundreds of Å, with the grains randomly distributed in a nonmagnetic metallic matrix. When these alloys are prepared by melt-spinning or by sputtering methods, the mean grain size is very small (a few Å) displaying a predominantly SPM behavior. However, granular alloys produced by other techniques such as sol-gel and mechanical alloying contain both SPM and blocked (BL) particles<sup>10,11,14,15</sup>, displaying a more complex magnetic behavior.

Magnetization and Mössbauer Spectroscopy measurements have been used to study magnetic properties of the granular alloys FeCu and FeAg in powder form<sup>8,10</sup>. The average sizes of magnetic particles were estimated by the coercive field.

In this work a series of Fe<sub>10</sub>Ag<sub>90</sub> granular alloys has been produced by a sol-gel method and characterized by X-ray diffraction, Mössbauer spectroscopy and magnetization. We have observed that in all samples there is a size distribution of both SPM and BL particles with a strong influence of changes in acid concentration.

### 2. Experimental

Fe<sub>10</sub>Ag<sub>90</sub> granular alloys were produced by a sol-gel method<sup>8,9</sup>. The start solution was prepared from an aqueous solution of the nitrates with five different nitric acid concentrations: 0.25, 0.50, 0.75, 1.00 and 1.25 ml, respectively, in 300 ml of mixture. The powder obtained was reduced in a hydrogen atmosphere for 45 min at a temperature of 400 °C.

The crystalline structure of the sample was investigated by conventional X-ray diffraction using a Rigaku diffractometer with Mo-K<sub>α</sub> radiation. The magnetization and hysteresis curves were measured in a vibrating sample magnetometer with a maximum magnetic field of 1 T. The Mössbauer spectrum was obtained in a conventional constant acceleration spectrometer with a <sup>57</sup>Co source in a rhodium matrix at room temperature.

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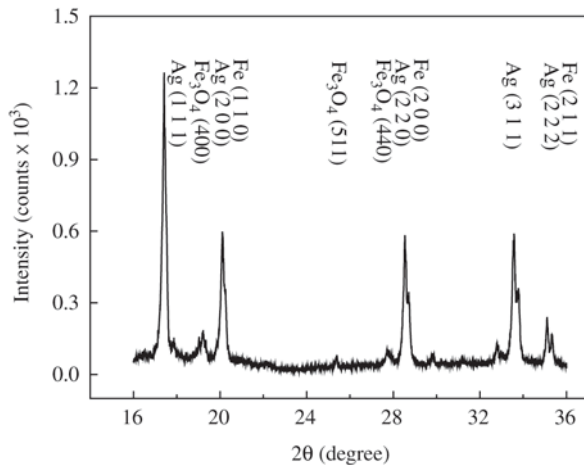
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### 3. Results and Discussion

Figure 1 shows the X-ray diffractometry patterns of  $\text{Fe}_{10}\text{Ag}_{90}$  granular alloy reduced at temperature of 400 °C. It is shown that the iron in the  $\text{Fe}_{10}\text{Ag}_{90}$  is b.c.c. structure and the silver is f.c.c. structure; there are also some low intensity reflections from  $\text{Fe}_3\text{O}_4$ . Assuming that the Fe in the sample is indeed distributed as small particles, the average particle diameter should be related to the Fe Bragg peak widths by Scherrer's formula,  $D_m = (0.9\lambda)/[\delta(2\theta)\cos\theta]$ , where  $\lambda$  is the Mo wavelength,  $\delta(2\theta)$  is the Fe(211) peak width, and  $\theta$  the scattering angle<sup>16</sup>. Figure 2 shows details of the spectral regions around the Ag (222) and Fe (211) Bragg peaks. Applying Scherrer's formula, we obtain the average Fe particle sizes  $D_m$  from the (211) line width for different nitric acid concentrations  $V_{ac}$ , as shown in Table 1. We observe that  $D_m$  increases with  $V_{ac}$  up to a concentration of 0.75 ml followed by a decrease.

Figure 3 shows the hysteresis curves at room temperature obtained for  $\text{Fe}_{10}\text{Ag}_{90}$  granular alloy samples with different nitric acid concentrations. None of the samples reach saturation in fields up to 1 T, showing the presence of SPM particles. The coercive field, determined from the hysteresis curves, ranges from 354 to 430 Oe, which is much higher than that of pure iron, indicating fine single-domain particles.

Figure 4 shows the Mössbauer spectrum of the  $\text{Fe}_{10}\text{Ag}_{90}$  (0.5 ml) sample. We can observe that the spectrum consists



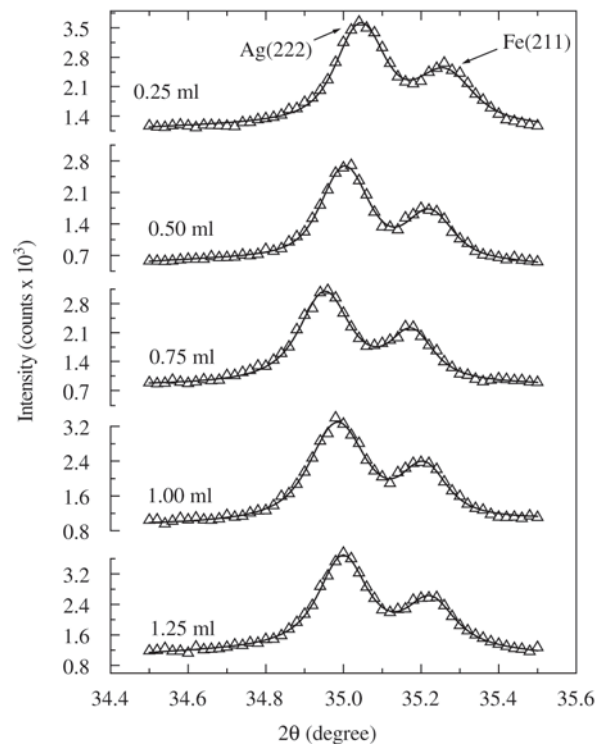
**Figure 1.** X-ray diffractogram of  $\text{Fe}_{10}\text{Ag}_{90}$  granular alloy (0.75 ml).

**Table 1.** Average Fe particles sizes  $D_m$  in the  $\text{Fe}_{10}\text{Ag}_{90}$  granular alloy in different nitric acid concentrations  $V_{ac}$ .

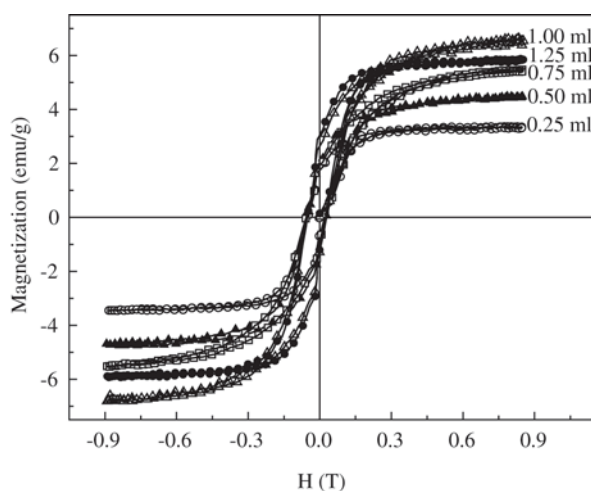
$V_{ac}$ (ml)	0.25	0.50	0.75	1.00	1.25
$D_m$ (nm)	24.5	28.0	28.6	26.2	25.3

of two components, a singlet, characteristic of an SPM phase, and a sextet corresponding to the blocked particles in the ferromagnetic  $\alpha$ -Fe. Observe that we were able to see the presence of SPM particles, even though the measurement time window of the Mössbauer effect is  $10^{-8}$  s.

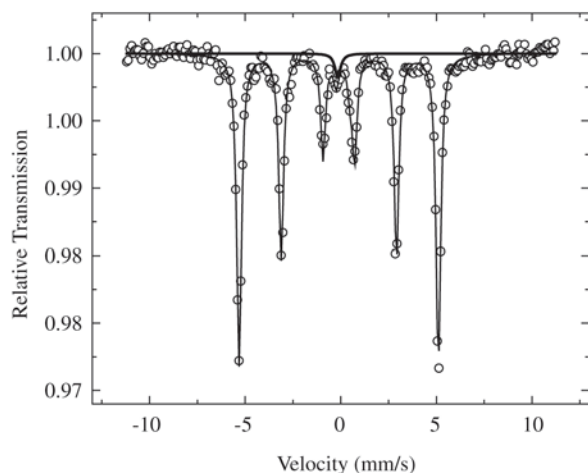
Figures 5a, 5b, 5c, and 5d show the change of the maximum field magnetization  $M_{H_{max}}$ , the remanent magnetization  $M_r$ , the coercive field  $H_c$ , and  $D_m$  with acid concentration.  $M_{H_{max}}$  increases linearly with acid concentration up to  $V_{ac} = 1.00$  ml, then decreases. This shows that the acid concentration in the start solution significantly affects the maximum field magnetization and consequently the atomic dispersion in the sample. In Fig. 5b, we can see that the remanent magnetization increases linearly with  $V_{ac}$ , despite a small dip at  $V_{ac} = 0.75$  ml. Figure 5c shows that  $H_c$  increases linearly with acid concentration, up to a maximum value ( $H_c = 434$  Oe) at  $V_{ac} = 0.75$  ml, then decreases linearly. In Fig. 5d, we can observe that  $D_m$  increases non-linearly with  $V_{ac}$  up to  $V_{ac} = 0.75$  ml, followed by a non-linear decrease. There is thus not a perfect correlation between  $H_c$  and  $D_m$ , as there is in a uniform particle system. Therefore, there appears to be a distribution of Fe particles



**Figure 2.** X-ray diffractogram of the Ag (222) and Fe (211) peaks of  $\text{Fe}_{10}\text{Ag}_{90}$  granular alloy, in different nitric acid concentrations. The solid line was the fitting obtained using two Pseudo-Voigt functions.



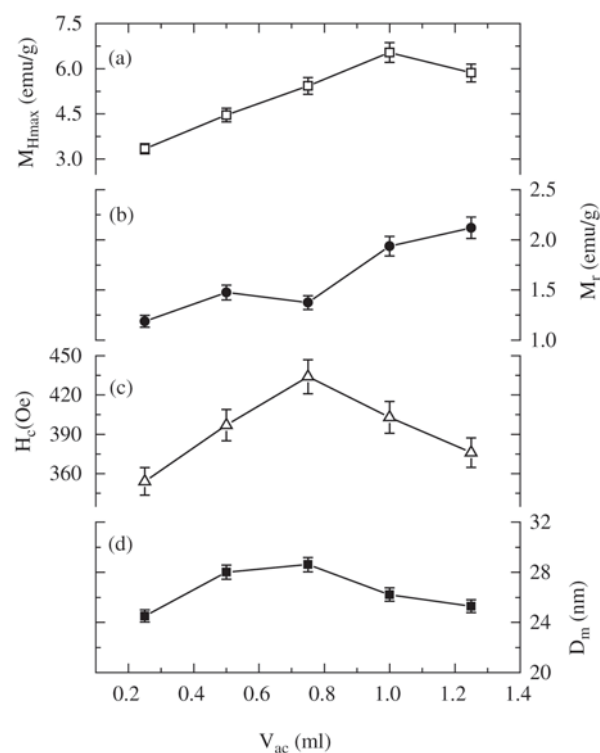
**Figure 3.** Room temperature hysteresis curves of the  $\text{Fe}_{10}\text{Ag}_{90}$  granular alloy samples in different nitric acid concentrations.



**Figure 4.** Mössbauer spectrum of the  $\text{Fe}_{10}\text{Ag}_{90}$  granular alloy at room temperature, showing singlet and sextet subspectra derived from an overall fit.

sizes whose width depends on the acid concentration. Since the  $H_c$  value depends directly on the number of blocked particles in the distribution, the coercive field ought to have a strong dependence on the distribution width, not just on the average particle sizes. However to obtain a correlation between the particles size distribution width and the coercive field more experimental and computational work is necessary. The distribution width varies between 1.14 and 1.67<sup>17</sup>.

The experimental results obtained from the hysteresis curve and Mössbauer spectroscopy measurements both indicate that the magnetization of the  $\text{Fe}_{10}\text{Ag}_{90}$  samples is



**Figure 5.** Magnetic properties of  $\text{Fe}_{10}\text{Ag}_{90}$  prepared in different nitric acid concentrations: a) Maximum field magnetization; b) Remanent magnetization; c) Coercive field; d) Fe means particle diameter obtained of X-ray diffraction.

composed of two contributions: one from the SPM particles and other from the BL particles, as observed in other systems reported in the literature<sup>11,14,15</sup>.

#### 4. Conclusion

Samples of  $\text{Fe}_{10}\text{Ag}_{90}$  granular alloys were produced in various concentrations of nitric acid. Magnetic measurements show that in all samples there was a size distribution of both SPM and BL particles. The magnetic properties of the samples are strongly influenced by changes in acid concentration.

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