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Fe_xCu_{1-x} ALLOYS FOR PERMANENT MAGNETS

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New magnetic composite materials, ribbon samples, with nominal compositions Fe_xCu_{1-x} (x = 0.2; 0.5; 0.8) were obtained by the melt spinning technique. Various information was provided via X-ray diffraction, Mössbauer and magnetic measurements. The structural aspects: phase composition, size and texture effects, have been corroborated with magnetic properties. Finally, the possibility to choose the adequate composition and the suitable processing in order to obtain desired anisotropic magnetic materials were presented.

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1. Introduction

The equilibrium diagram of the Fe-Cu binary system (Fig.1) points to a very low, practically insignificant miscibility of the two metals, each one crystallizing in very definite phase: b.c.c for iron and f.c.c for copper.

In 1962, Levi [1] reported that after inserting Fe-wires in a Cu-matrix and a subsequent drawing, Fe-wires with reduced diameters (down to 300 Å) in plastically deformed copper matrix can be obtained. In that case, the iron volume fraction was 45% and the system has shown an intrinsic coercivity of 320 A/cm. Latter in 1971, by lamination of a cast FeCu compound, Kawabuchi et al. [2] presented similar magnetic results. However, such magnets were not attractive mainly due to high costs compared with performances.

It is worth mentioning an industrially produced ternary system ($Fe_{20}Ni_{20}Cu_{60}$), known as "cunife". After a thermal homogenizing treatment at 1050 ºC, followed by quenching and subsequent annealing at 550 ºC, as a result of spinodal decomposition, inside of the matrix appeared a precipitated FeNi phase [3]. The performances of this isotropic magnetic system consist in a specific energy of 4 kJ/m^3 and a coercivity around 320 A/cm. If the system is strongly cold deformed, the specific energy can increase up to 12 kJ/m^3 and the coercivity up to 440 A/cm, as a result of shape induced anisotropy [4]. Because the Curie temperature is below the spinodal decomposition temperature [5] the application of an external magnetic field during the precipitation will have no effect on the grains orientation, due to the paramagnetic state of spins.

In Romania, the first attempt to Fe-Cu based permanent magnets dated back to 1976 when an $Fe_{70}Cu_{30}$ system has been melted in vacuum via induction furnace and then cast between two copper foils. The cast alloy was subsequently laminated in order to elongate the Fe domains. The maximum coercivity was about 100 A/cm [6,7].

The last two decades brought various alternative procedures to obtain new materials by promoting a larger miscibility of different metals. Among them, mechanical attrition (ball milling) or rapid quenching (melt-spinning) are suitable for producing amorphous or nano-cristallized systems including Fe-Cu ones.

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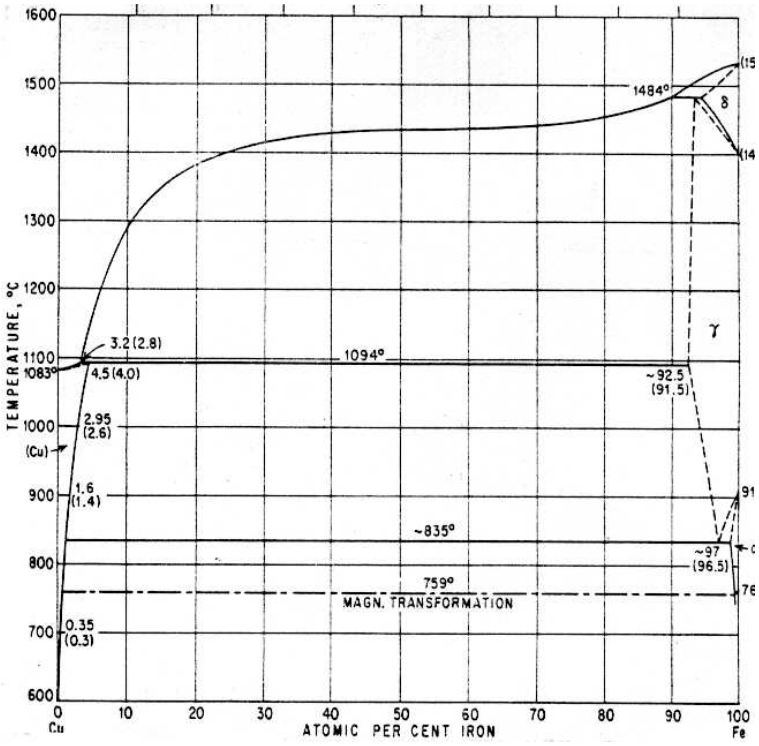

Fig. 1. Equilibrium diagram for Fe-Cu binary system.

The new processes are related to interface mechanisms induced by temperature or by external effects appearing at the large surface area offered by the nano-particles. There are reported magnetic properties of granular Fe-Cu systems obtained by melt spinning (see for example [8]). A precipitated α-Fe (bcc) phase, with different grain shape and sizes was obtained in a Cu (fcc) matrix for various Fe_xCu_{100-x} compositions (x=20, 50, 80). These heterogeneous nanosystems indicate magnetisations which are different from the values provided by the usual spherical α-Fe grains in Cu. The difference decreases with increasing annealing temperature. It is reported that the anisotropy field, H_K, is higher than that of Fe (H_K ~ 560 A/cm) calling attention that, beside the contribution of the magnetocrystalline anisotropy there is another one. A natural conclusion is that the shape anisotropy determines the positive difference. In this context, it has to be mentioned that the precipitates of $Fe_{70}Cu_{30}$ provided a length/diameter ratio of 4.4.

2. Experimental

A new set of the Fe_xCu_{1-x} (x = 20, 50, 80) alloys were prepared by melt spinning, starting from technical pure iron and electrolytical pure Cu. The chemical compositions and the processing conditions are presented in Table 1.

The samples were investigated using X-ray diffractometry, Mössbauer analysis and magnetometry. Structural analysis was performed by X-ray diffractometry via an AD 8 Advanced Bruker diffractometer. The Mössbauer data were obtained using a traditional set-up with symmetrical wave-form and the ^{57}Co source in a rhodium matrix. For the magnetic measurements the samples were inserted in a vibrating sample magnetometer (VSM).

Table 1. Chemical compositions and the processing conditions of the studied composites.

Sample code	Chemical composition (wt. %)	Melt spinning conditions		
		Nuzzle diameter (mm)	Overpressure Δp (atm)	Wheel velocity, v (m/s)
1	Fe20-Cu80	0.50	0.40	30
2	Fe20-Cu80	0.40	0.50	30
3	Fe20- Cu80	0.50	0.45	30
4	Fe50-Cu50	0.50	0.45	30
5	Fe80- Cu20	0.50	0.45	30

3. Results and discussion

The powder samples prepared from the melt spun alloys, measured by the VSM, provided the magnetic data presented in the Table 2. The main values describing the magnetic properties are derived from demagnetisation curves. An example of such curve is presented in Fig. 2, for the melt spun Fe50-Cu50 alloy.

Table 2.The magnetisations of the studied samples: experimental and theoretical estimations.

Sample	C_{mFe} (%)	C_{VFe} (%)	Sample volume (cm^3)	Sample mass (g)	Density ρ (g/cm^3)	Measured magnetic moment, μ (emu)	Measured magnetisation M_m (Gs) (Gs)	Theoretical magnetisation M_{th} (Gs) (Gs)
1/Fe20-Cu80	0.2	0.222	0.00205	0.0180	8.760	0.023	11.290	366.1
2/Fe20-Cu80	0.2	0.209	0.00134	0.0110	8.232	0.005	3.740	366.1
3/Fe20-Cu80	0.2	0.216	0.00199	0.0175	8.787	0.159	79.836	366.1
4/Fe50-Cu50	0.5	0.505	0.00223	0.0178	7.950	0.621	277.350	884.6
5/Fe80-Cu20	0.8	0.801	0.00240	0.0193	7.870	1.630	664.660	1403.0

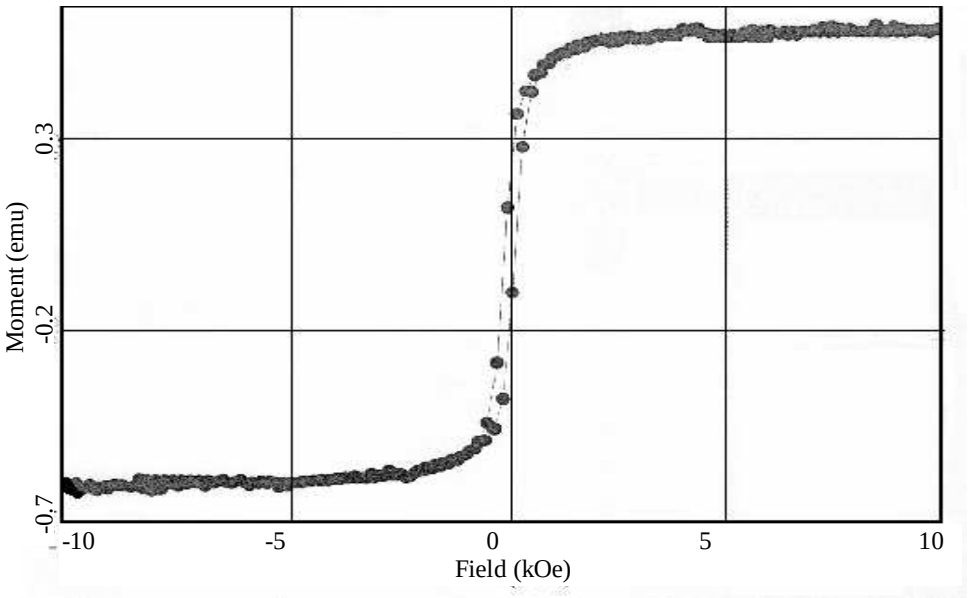

Fig. 2. The demagnetisation curve for the melt spun Fe50-Cu50 alloy.

The following relationship defines the magnetic moment μ:

$$\mu = M \cdot V = M \cdot \frac{m}{\rho} \tag{1}$$

where: μ is the magnetic moment; M is the magnetisation; V is the sample volume; m is the sample mass, in grams and ρ is the sample density.

$$M = \frac{\mu \cdot \rho}{m} \tag{2}$$

From (1) one can find the values of the magnetisation. In order to calculate the total magnetic moment of a sample, one has to consider it as resulted from: (i) the sum of the individual moments of each component, Fe and Cu, and (ii) the structure of the peculiar phases forming the composites. Then the relationship becomes:

$$M_{sat} = M_1 \cdot C_{vCu} + M_2 \cdot C_{vFe} \tag{3}$$

where: M_{sat} is the saturation magnetisation; C_{vCu} - Cu volumic concentration, C_{vFe} - Fe volumic concentration. $M_1 = 0$ (for Cu) and $\mu_o M_2 \cong 2.2$ T for Fe.

The relation can relate the volume concentrations function on the mass concentrations

$$C_{Fe} = \frac{V_{Fe}}{V} = \frac{V_{Fe}}{m/\rho} = \rho \cdot \frac{m_{Fe}}{\rho_{feu}} \cdot \frac{1}{m} = \frac{\rho}{\rho_{Feu}} \cdot \frac{m_{Fe}}{m} = \frac{\rho}{\rho_{Fe}} \cdot C_{mFe} \tag{4}$$

$$C_{mCu} = 1 - C_{mFe} \tag{5}$$

where: C_{mFe} – Fe mass concentration; ρ_{Fe} – Fe density, equal with 7.86 g/cm^3; ρ_{Cu} – Cu density, equal to 8.96 g/cm^3.

Using the volume concentrations for Fe and Cu in the relation (3) and having in mind that the saturation magnetisation of pure α–Fe, is $4\pi M_2 = 22000$ Gs and the Cu magnetisation $M_1 = 0$, the theoretical values of the alloy magnetisations will be obtained and compared with those deduced from the hysteresis loops. In this manner, magnetic information about the sample phase constituents can be obtained, and then corroborated with those provided by X-ray diffraction and the Mössbauer spectroscopy.

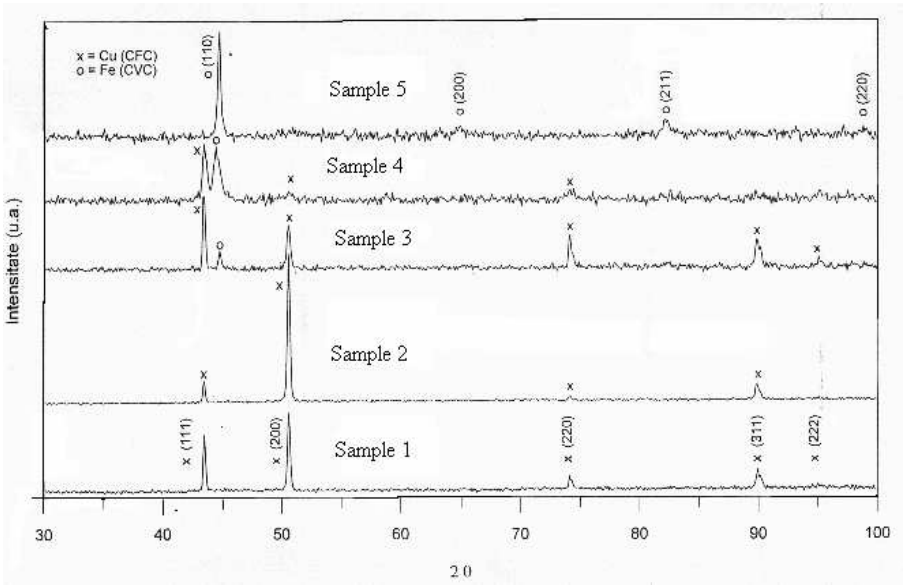

Fig. 3. The X-ray diffraction pattern for the melt spun Fe-Cu alloys (×: fcc-Cu, o: bcc-Fe).

The higher values of the magnetisations observed for the samples 4 and 5 derive from the presence, in a higher concentration, of α – Fe (evidenced also by the shape of the hysteresis loops). From the analysis of the hysteresis loops, it results that the coercivity is higher (H_c = 121 Oe) for sample 4, Fe50-Cu50 alloy, in comparison with the coercivity (H_c ~17 Oe) of the sample 5, Fe80-Cu20 alloy. It is worth to mention that the shape of the hysteresis loop of sample 5 is specific to soft iron. A structural texture was induced in the equi-atomic composition (sample 4), as resulted from the inversion of the line intensities in the X-ray pattern (Fig. 3).

As observed by the X-ray diffractometry (see Fig. 3) and Mössbauer analysis of samples 1 and 2 (Fe20-Cu80 alloys obtained in different conditions), fcc-Cu is the major phase and Fe mainly dissolved in the Cu matrix. That is also in concordance with the lower values of the magnetisation: 11.29 Gs for the sample 1 and 3.74 for the sample 2. It is obvious that these samples need a subsequent recrystallisation treatment. In the case of the sample 3 (also an Fe20–Cu80 alloy), with the same chemical composition as reference samples 1 and 2, the value of magnetisation is higher (79.836 Gs), due to the clearly evidenced α - Fe (b.c.c) phase. The main peak belonging to the bcc-Fe phase in the X-ray pattern of sample 3 is wide, supporting the suggestion that Fe is dissolved under the form of very fine precipitates. The coercivity of this sample is H_c = 169 Oe, also confirming the above description.

In sample 4 (Fe50 – Cu50 alloy), only the main peak of bcc-Fe is evidenced among the fcc-Cu peaks. All peaks (including fcc-Cu ones) are large, suggesting that the sample granulation is reduced: very fine crystallites of Cu (f.c.c) and Fe (b.c.c.) phases are characterized by intergrowth. The hysteresis loop is wider too, supporting the existence of anisotropic Fe precipitates. The coercivity of this sample is H_c = 121 Oe.

In sample 5 (Fe80 – Cu20 alloy), α- Fe phase is present as the main phase. The Cu lattice seems to present a short coherence length, related to the absence the fcc-Cu peculiar peaks. It is far evidently that the magnetisation of this sample is the highest: M_m = 664.66 Gs. The presence of Cu in Fe has distorted the crystalline configuration of Fe. Following a recrystallization process, the Cu could be separated from this surroundings and a structure characterised by even a higher magnetisation, near to the theoretical value M_{th} = 1403 Gs can be formed.

The Mössbauer measurements, providing the highest and more detailed information about Fe compounds, confirmed the clear existence of a f.c.c.- Fe phase, by a singlet near to zero velocity beside the α - Fe phase, that appears as a sextet with a field of 33 T.

From the corroborated data offered by the magnetic, Mössbauer and structural X-ray characterisation, the most suitable samples showing the desired characteristics (larger coercive fields and magnetisations) and related structure are the samples labelled as samples 3 and 4. Therefore, one can affirm that in the Fe-Cu system, processing via melt spinning, in well selected conditions, composites with grain size ranging from a few nm to hundreds of nm can be obtained.

4. Conclusions

Depending on the processing conditions, the melt-spinning method provided composites with various phase concentrations, different size and shape of nano-grains and with the related magnetic properties.

In some samples, γ-Fe phase was detected, its concentration increasing with Cu/Fe ratio in the composite. A definite soft- iron phase (magnetic measurements) and a texture of the b.c.c. α-Fe (X-ray diffraction) was observed in the Fe50-Cu50 sample.

The magnetic properties are in good agreement with data of the X-ray and Mössbauer investigations. The corroborated data has allowed the selection of the compositions and the adequate processing to obtain anisotropic permanent magnets. It involves the control of the phase composition and also of the grain size and shape, finally leading to the local and spin interactions, proper to magnetic anisotropic systems.

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