Formation of ferromagnetic bulk amorphous $Fe_{70}Cr_6Ga_4P_{12}Si_5C_3$ alloys

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The necessity of obtaining amorphous alloys with higher thickness which correspond to different industrial applications requirements, determined the intensification of researchers regarding the processing possibility of some amorphous alloys characterized by critical cooling rate as small as possible. Consequently, the focus of this paper was to process ferromagnetic bulk amorphous alloy rods, with 1 and 2 mm diameter, using copper mould casting method. The samples obtained were structural investigated by X-Ray diffraction (XRD), differential thermal analysis (DTA), differential scanning calorimetry (DSC) and magnetic characterized by conventional low frequency induction method, in order to observe the influence of the elaboration technology on glass forming ability and magnetic properties of the elaborated alloy.

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1. Introduction

In the last decades a large number of amorphous alloys has been produced with special soft magnetic properties, most notably Fe and Co based alloys. These alloys are characterized by a high amorphization critical cooling rate ($10^5 \ldots 10^6$ K/s). Consequently, these alloys can be produced as ribbons, wires or foils with thickness up to 60 μm [1].

The necessity of products with higher thickness, for industrial applications, lead to wide researches on the ferromagnetic bulk amorphous alloys.

Recently, the researches [2, 3] led to the discovery of some new multicomponent amorphous Zr, Pd, Pt, Mg, Ti based alloys characterized by critical amorphization speed lower as 10^3 K/s, from which one can obtain products of tens millimeters thickness. It should be noted that these alloys are characterized by:

- large ratio of T_g / T_m (T_g is the glass transition temperature and Tm is melting temperature);
- the temperature range between the crystallization temperature T_x and the glass transition temperature T_g high, it means $\Delta T_x = T_x - T_g$ has high values.

Consequently, these alloys are characterized by high glass forming ability (GFA), its value depending especially on the chemical composition.

Japanese researchers [3,4,5] have established 3 empirical rules which should be fulfilled, from chemical point of view, by GFA alloys:

- multicomponent system consisting of more than three elements;
- significant difference (beyond 12 %) in atomic size ratio among the three main constituent elements;
- negative heat of mixing among the three main constituent elements.

Taking into account these rules, in order to obtain ferromagnetic bulk amorphous alloys, other elements like P, Si, C, Ga must be introduced in the chemical composition beside the base element (iron) [6,7]. But these elements must be in a definite ratio in order to ensure the one hand a high GFA and on the other hand no effect on the magnetic properties of the alloy. In addition to these elements, sometimes is required the presence of other alloying elements as Cr, Ni, which can contribute to the improvement of some mechanical properties or corrosion resistance, required in different industrial applications.

2. Experimental procedures

Experiments were aimed to obtain amorphous $Fe_{70}Cr_6Ga_4P_{12}Si_5C_3$ alloys as rod shapes having diameters about 1 or 2 mm. For this purpose, metallic Fe, Cr, Ga pure powders and ferroalloys: FeP_{28}, $Fesi_{75}$, $FeC_{0.17}$ were used as raw materials.

In a first stage the alloy was produced in the bars, by inductive heating and melting of the raw materials in argon atmosphere. The microscopic structure of the alloy shows the presence of a fine polinary eutectic and of some dendritic crystals of an iron based solid solution (Fig. 1). For chemical composition certification, the obtained master alloy was investigated by EDX, using a Scanning Electron Microscope XL 30 ESEM. The EDX spectrum and the percentage contents of the elements are shown in Fig. 2.

As it results from EDX analysis, the chemical composition of the obtained alloy, is very close to the theoretical one. It is noted, however, a slight increase of Ga and C content, and in compensation, a reduction Cr, Fe and P content.

In order to obtain the amorphous alloy as rods, there was used the casting mould copper method. Two different technologies were applied.

The first one presumes the following steps:

- Cutting the master alloy in order to obtain pieces of about 5 grams;

- Inserting the master alloy in the quartz crucible, endowed at the bottom with a 1mm diameter hole for melt ejection. In this crucible, the master alloy is remelted by inductive heating using an argon atmosphere;
- Relative positioning of the crucible against the copper mould and melt ejectation in mould cavity.

The second technology presumes the performing of the same steps, except that, in the second step the master alloy was 4 times melted and remelted under B_2O_3 flux.

The main process parameters are:

- preparation temperature, having a value higher than fusion temperature of the alloy by 100 ^oC;
- overpressure applied to the melt: 0.35 atm;

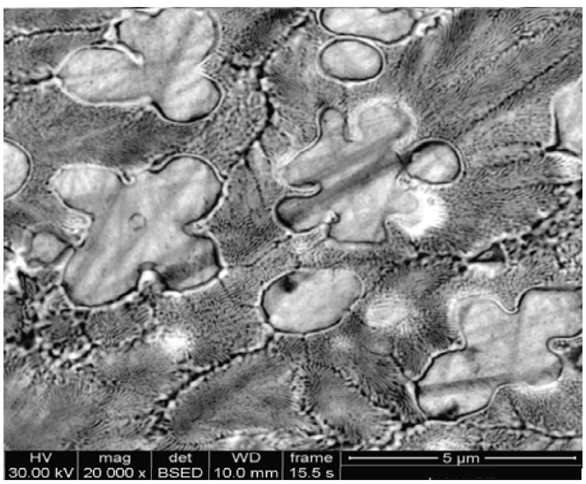


Fig. 1. SEM micrograph of the master alloy.

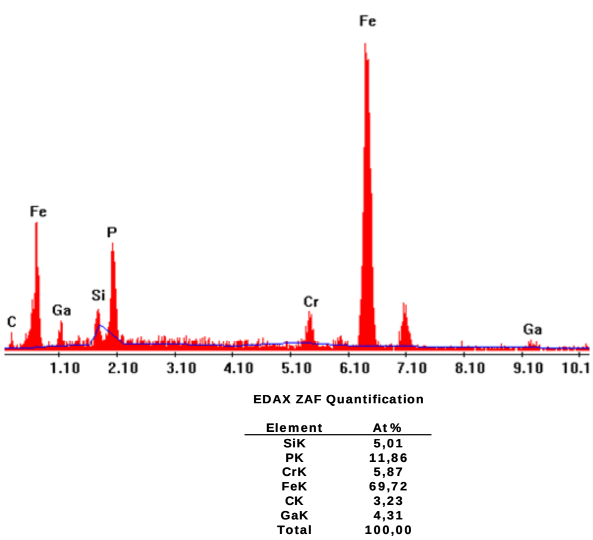


Fig. 2. EDX analysis of the obtained master alloys.

As a result of the mould casting, rods having diameters of 1 or 2 mm and lengths of 20-25 mm were obtained (Fig. 3).

Fig. 3. The rods prepared by mould casting.

The structural characterization of the alloy obtained as rods using both technologies was carried out by X-ray diffraction (XRD) and differential thermal analysis (DTA).

The structural analysis by X-ray diffraction was performed in a DRON 3 diffractometer, using the Mo radiation with wavelength λ = 0.71 Å. The obtained patterns (Fig. 4) show an amorphous structure in case of 1 mm rods, obtained by both technologies. In case of 2 mm rods the amorphous structure appears in case of the homogenization by multiples remeltings of the master alloy under the shielding of a bore oxide (B_2O_3) flux.

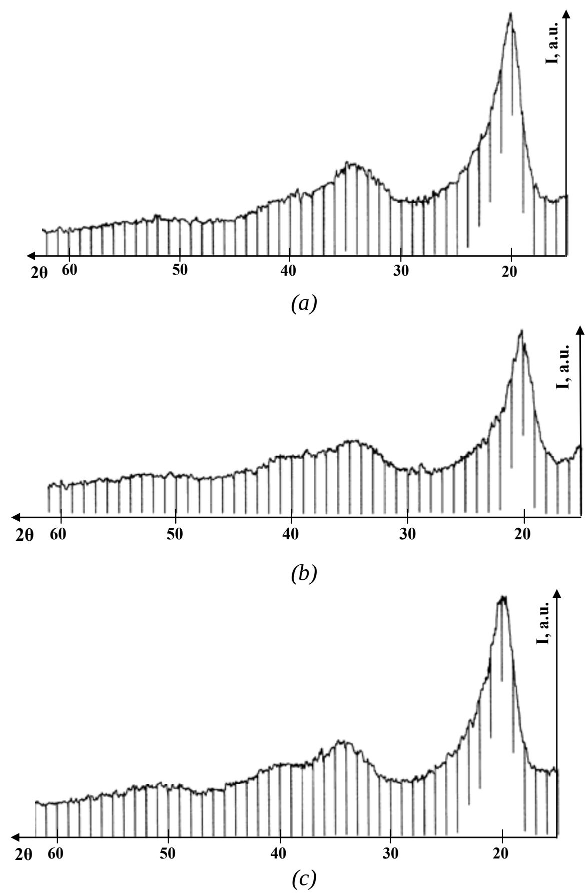


Fig. 4. X-ray patterns of the obtained alloys: (a) 1mm diameter rod; (b) 1mm diameter rod with multiples remeltings under bore oxide flux; (c) 2 mm diameter rod with multiples remeltings under bore oxide flux.

The differential thermal analysis was realized using the apparatus Baehr 1600 °C type, which has a data acquisition and processing system.

DTA curves obtained (Fig. 5) show an exoothermic effect which marks the crystallization of amorphous phase and an endothermic effect on the melting temperature.

For getting the glass transition temperature and crystallization temperature, the samples were investigated by DSC analysis.

It can be noticed a good stability of the amorphous phase, especially in case of 1 mm diameter rods, obtained by remelting master alloy technology under bore oxide flux. The beginning temperature of the crystallization process was at 468.6 °C (curve b). In case of the alloy obtained in the shape of 1 mm diameter rod, without the remelting of the master alloy under bore oxide flux (curve c) and in case of 2 mm diameter amorphous rods (curve a), the crystallization temperatures are situated at 455.6 °C, and 456.1°C respectively.

The a and c curves show a peak splitting which certifies the presence of two processes: a primary crystallization of the amorphous phase followed by a transformation of the crystalline phase. The largest ΔT_x (T_x-T_g) is 38 °C for the 1 mm diameter rods obtained by remelting under bore oxide (curve b) resulting a better glass forming ability. The other values for ΔT_x were 27 °C for 1 mm diameter rods without remelting and 25 °C for the 2 mm diameter rods.

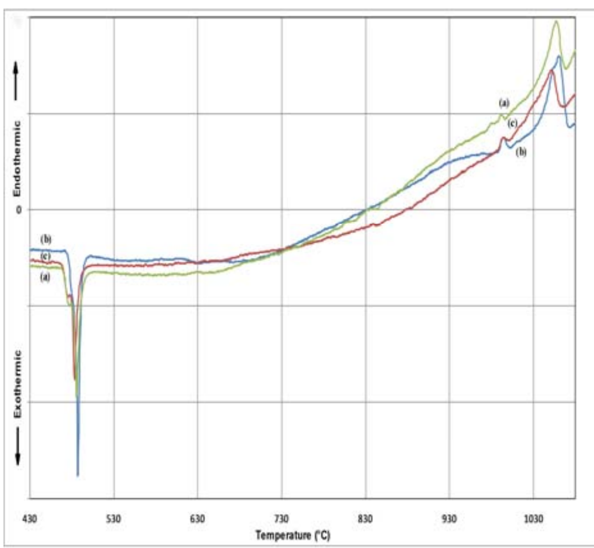


Fig. 5. DTA curves of the amorphous alloys obtained in the shape of rods.

The magnetic measurements on the obtained alloys were performed using a fluxmeter type installation, by conventional low frequency induction method [8, 9]. The hysteresis cycles of amorphous rods (Fig. 7) have been recorded. They allowed the estimation of the saturation magnetic induction and of the coercive field.

The results of measurements are summarised in Table 1. The results obtained are comparable with the ones obtained previously by other research groups.[10]

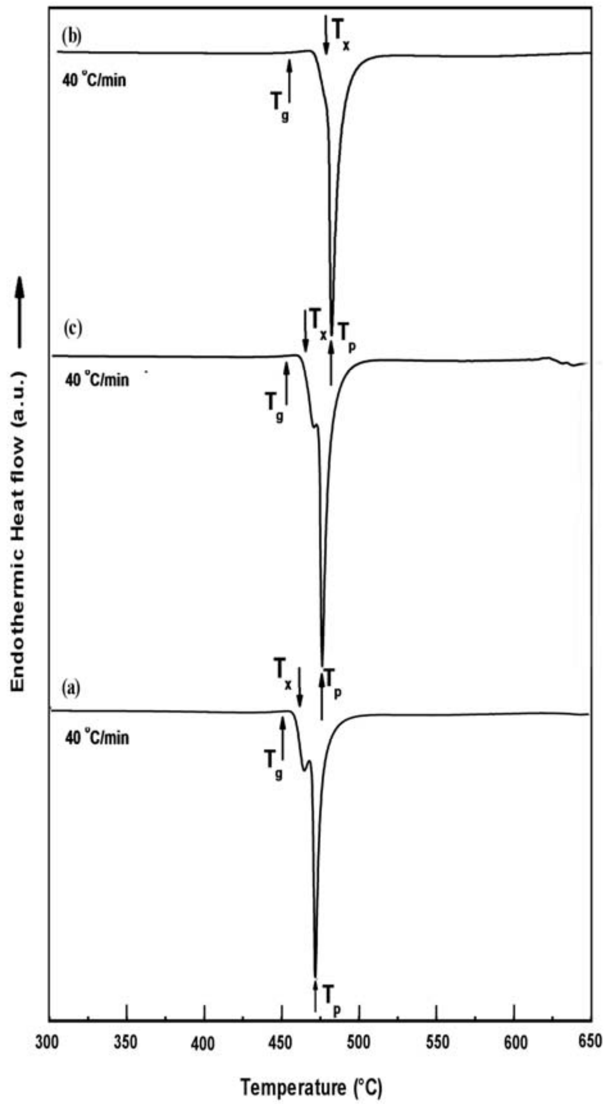
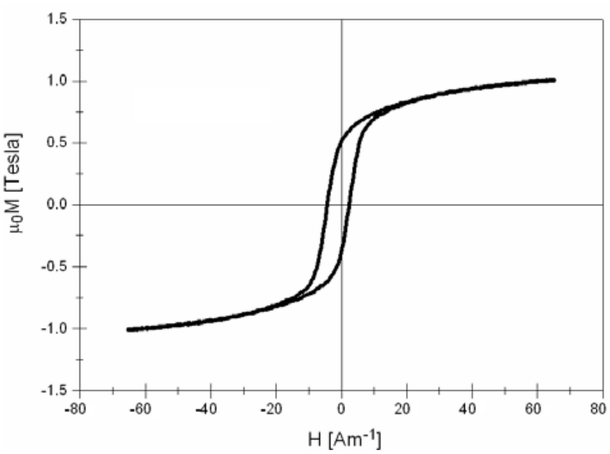


Fig. 6. DSC curves of the amorphous alloys obtained in the shape of rods: (a) – 1 mm diamater rod; (b) - 1mm diameter rod with multiples remeltings under bore oxide flux; (c) - 2 mm diameter rod with multiples remeltings under bore oxide flux.

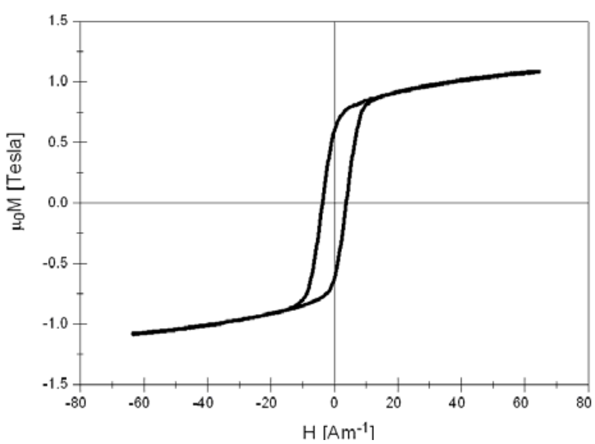
It is noticeably that in case of the alloy with 1 mm diameter rod, by multiple remeltings technology under flux layer of the master alloy (Fig. 7b) the saturation magnetization is higher, and the coercitive field is lower than in case of the alloy obtained without master alloys remelting.

Table 1. Values of the magnetic characteristics.

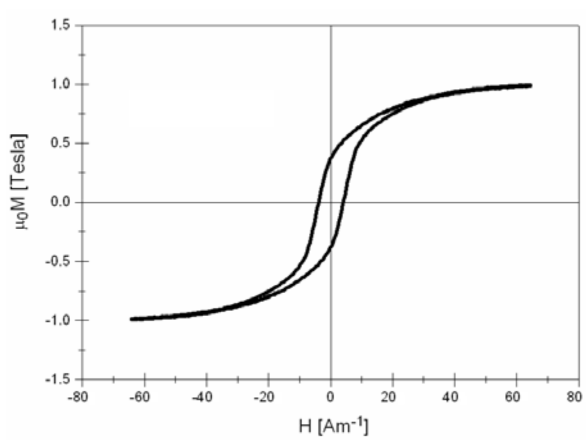
Magnetic property	1mm diameter rod	1mm diameter rod with multiples remeltings	2 mm diameter rod with multiples remeltings
Saturation magnetic induction, T	1	1.2	0.95
Coercive field, Am^{-1}	9.5	8	10



(a)



(b)



(c)

Fig. 7. Hysteresis cycles of the obtained bulk alloys: (a) 1mm diameter rod; b) 1mm diameter rod with multiples remeltings under bore oxide flux; (c) 2 mm diameter rod with multiples remeltings under bore oxide flux.

4. Conclusions

The investigations carried out in this paper have pointed out the possibility of getting bulk amorphous alloys in the shape of rods from Fe-Cr-Ga-P-Si-C family by casting under pressure using a copper mould.

The structural analysis indicated that the usage of a technology which involves multiples remelting of the master alloys under bore oxide flux, before mould casting, assures the obtaining of a stable amorphous structure and permits to get amorphous structures with larger thicknesses of the bulk products.

The magnetic characterization of the elaborated rods revealed that at the product with the same thickness the soft magnetic characteristics are superior in case of using multiples remeltings technology of the master alloys under bore oxide flux.

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