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Effect of partial substitution of Cr with Co on glass forming ability, mechanical and magnetic properties in $Fe_{48}Mo_{14}Cr_{15-x}Co_xB_6P_8Si_7Y_2$ bulk metallic glasses

Efecto de la sustitución parcial de Cr con Co sobre la capacidad de formación de vidrio, propiedades mecánicas y magnéticas en vidrios metálicos a granel $Fe_{48}Mo_{14}Cr_{15-x}Co_xB_6P_8Si_7Y_2$

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Abstract

Fe-based BMGs are well known for its attractive combination of structural, magnetic and corrosion properties. These alloys are widely used in industry because of low price and good glass forming ability (GFA). In this paper, the effect of partial substitution of Cr with Co on GFA, mechanical and magnetic properties of Fe48Mo14Cr15-xCoxB6P8Si7Y2 bulk metallic glasses was studied. The samples were prepared by copper mold casting technique in rod form. The elaborated alloys were structurally investigated by X-Ray diffraction (XRD) and differential scanning calorimetry (DSC), while the mechanical properties were investigated by compressive and nanoindentation tests. Also, the magnetic properties were investigated. The GFA was estimated by several criterion or parameters (reduced transition temperature Trg, ΔTx, α, β, γ and δ parameters). It was found that the addition of Co instead of Cr leads to a slight increase of the glass forming ability of Fe-Mo-Cr-B-P-Si-Y family. Also, an increase in magnetic properties, hardness, elastic modulus and compressive strength has been noted.

Keywords: Bulk metallic glasses; GFA; DSC; Nanoindentation; compressive strength; soft magnetic properties.

Resumen

Los BMG a base de Fe son bien conocidos por su atractiva combinación de propiedades estructurales, magnéticas y de corrosión. Estas aleaciones se utilizan ampliamente en la industria debido a su bajo precio y buena capacidad de formación de vidrio (GFA). En este trabajo se estudió el efecto de la sustitución parcial de Cr con Co sobre GFA, propiedades mecánicas y magnéticas de vidrios metálicos a granel Fe48Mo14Cr15-xCoxB6P8Si7Y2. Las muestras fueron preparadas por la técnica de fundición en molde de cobre en forma de varilla. Las aleaciones elaboradas fueron

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investigadas estructuralmente por difracción de rayos X (XRD) y calorimetría diferencial de barrido (DSC), mientras que las propiedades mecánicas fueron investigadas por pruebas de compresión y nanoindentación. Además, se investigaron las propiedades magnéticas. La GFA se estimó por varios criterios o parámetros (temperatura de transición reducida Trg, ΔTx, parámetros α, β, γ y δ). Se encontró que la adición de Co en lugar de Cr conduce a un ligero aumento de la capacidad de formación de vidrio de la familia Fe-Mo-Cr-B-P-Si-Y. Además, se ha observado un aumento en las propiedades magnéticas, dureza, módulo elástico y resistencia a la compresión.

Palabras clave: vidrios metálicos a granel; GFA; DSC; nanoindentación; resistencia a la compresión; propiedades magnéticas blandas.

1. Introduction

Fe-based bulk metallic glasses (BMGs) are representative alloys of a new class of engineering materials - Amorphous alloys – having an attractive combination of structural, magnetic, hardness and properties with thicknesses of few millimeters. Due to their amorphous structure, it may achieve interesting properties, like high strength, high hardness, increased wear and corrosion resistance, and good soft magnetic properties. These entire properties make the Fe based BMGs very attractive for industrial application [1], [2], [3], [4], [5], [6], [7], [8], [9], [10], [11], [12], [13], [14].

The ability of a metallic alloy to transform into a glassy state is defined as the glass-forming ability (GFA). The GFA evaluation was estimated by calculating the popular parameters summarized in Table 1 [1], [2].

Table 1. GFA parameters

Parameter	Equation	Ideal Value
T_{rg}	$T_{rg}=T_g/T_l$	1.0
ΔT_x	$\Delta T_x=T_x-T_g$	High
α Parameter	$\alpha=T_x/T_l$	1.0
β Parameter	$\beta=1+T_x/T_l$	2.0
γ Parameter	$\gamma=T_x/(T_g+T_l)$	0.5
δ Parameter	$\delta=T_x/(T_l-T_g)$	High

Source: [2].

The reduced glass transition temperature T_{rg}, defined as the ratio between glass transition temperature T_g and the liquidus temperature T_l ($T_{rg}=T_g/T_l$) [15].

The supercooled liquid region ΔT_x, defined as the difference between the crystallization temperature T_x and glass transition temperature T_g. For a good GFA, it should be as large as possible [1]. Also, the GFA may be estimated by the enthalpy of crystallization ΔH_x, which should be high [1].

Another important parameter γ ($\gamma=T_x/(T_g+T_l)$) was developed by Lu et al. [16]. The stability of the liquid phase is related in particular to the reduced distance between the constituent elements, the structural arrangement of atoms in the liquid state and the thermodynamic stability of the liquid, expressed by the minimum free energy for certain chemical compositions. A high T_x means a higher stability of the glass and a low T_l means a higher stability of the supercooled liquid.

Combining these aspects, Mondal and Murty proposed a new parameter α ($\alpha=T_x/T_l$) [17]. They also proposed parameter β, by taking into account the ability to form glass during cooling from the liquid state and the stability of the glass $\beta=1+T_x/T_l$ [17].

Considering that glass formation requires avoiding the formation of a crystalline phase, Chen et al. [18] suggested that the GFA of alloys should be inversely proportional to the rates of nucleation and growth, proposing a new parameter δ defined as $T_x/(T_l-T_g)$ [18].

Obtaining the glassy state is conditioned by the fulfillment of mainly two conditions which refers to the chemical composition of the alloy and the cooling rate. According to Inoue [2], a chemical composition favorable to glassy state in Fe-based BMGs, should have transition metals, metalloids or other metals in proportion of 20-25 at.%. The presence of metalloids like B, P, Si and Y increase the glass forming ability [1], [2], [3], [4]. The atomic size difference between metals and metalloids leads to favor an increasing of the atomic packing density of the liquid structure. Also, the presence of Y in the chemical composition in Fe-based alloys contributes to a deoxidizing effect of the melt which can lead to suppression of heterogeneous nucleation, thus improving the glass forming ability [14].

Another chemical element that can affect GFA while having a favorable influence on thermal stability, hardness, anti-corrosion performance and magnetic property is Cobalt (Co). It is well known that Co, along with Mo and Cr contributes greatly on increasing the mechanical strength, corrosion and wear resistance. In a lot of papers [19], [20], [21]. is concluded that by substituting Fe with a proper amount of Co in Fe-based BMG alloy, or addition of a small amount of Co causes an increase in the glass-forming ability through the decrease in melting and liquidus temperatures, leading to the formation of bulk glassy alloys with diameters up to at least 5 mm.

Chromium (Cr) in Fe-based BMGs leads to increase the mechanical strength and hardness and in particular, provides excellent wear and corrosion performance even under relatively low percentage compared with stainless steels but affects negatively the GFA and soft magnetic properties [22], [23].

Co was chosen to partially replace Cr in Fe-based BMGs. Co has almost the same atomic radius as Cr (Co: 125 pm and Cr: 128 pm) and Cobalt is a ferromagnetic element meanwhile Cr is an anti-ferromagnetic element. Co has higher values of the atomic magnetic moment and saturation magnetization than Cr and also a stronger corrosion resistance than Fe element [22]. Therefore, it can be assumed that partial substitution of Cr with Co will increase the ferromagnetic element concentration and increase the total atomic magnetic moment.

This research study was aimed to establish the role of Co on GFA, magnetic and mechanical properties by partially replacing the chromium not the iron in the alloy composition, as reported in other studies [23], [24], [25], [26]. It was found that proportions higher than 7% at. have a negative influence on corrosion resistance and coercivity [20], [21], [22], [23], [24], [25], [26], [27]. The present paper reports the effects of Co-Cr substitution up to 7% at. in $Fe_{48}Mo_{14}Cr_{15-x}Co_xB_6P_8Si_7Y_2$ BMGs on GFA, magnetic and mechanical properties, considering new possible applications of these alloys.

2. Experimental procedure

Ingots of $Fe_{48}Mo_{14}Cr_{15-x}Co_xB_6P_8Si_7Y_2$ (x = 0, 4, 7) were prepared by induction melting the mixture of pure elements Fe, Cr, Co, Y metals and Fe–Mo, Fe–B, Fe–P and Fe–Si ferroalloys in an argon atmosphere. The alloy compositions represent the nominal atomic percent. The master alloys were remelted four times to obtain a better homogeneity. Bulk amorphous alloys in rods form with the diameter of 2 mm and length of 20 - 25 mm were prepared by the copper mold casting method.

The amorphous structure of the elaborated samples was examined by X-ray diffraction XRD using an X'Pert³ Powder diffraction system, with the radiation of a Cu anode with a wavelength λ = 1.54 Å. The thermal stability was investigated by differential scanning calorimetry (DSC) using a Netzsch STA 441 Jupiter under a flow of purified nitrogen. The glass transition temperature T_g, the crystallization temperature T_x and the melting temperature T_m were determined as the onset temperatures of the glass transition the crystallization and melting peak, respectively, and the liquidus temperature T_l as the offset of the melting peak during heating with a constant rate of 0.33 K/s.

The mechanical properties were determined by nanoindentation and compressive tests. The compressive tests were done at room temperature, at a loading speed of 1mm/min on a Zwick/Roell- machine. The specimens used in compressive tests were 2mm in diameter and 3mm height. The nanoindentation tests were conducted with a Berkovich diamond tip using an Anton Paar Nanoindentation Tester. The indentations were performed in the load-control mode with a maximum load of 20 mN at a constant loading/unloading rate of 40mN/min. At least 15 indents were performed to verify the accuracy and scatter of the indentation data. The results were obtained using the Oliver & Pharr method. The magnetic properties were examined in open magnetic circuit using a conventional AC measuring stand at low frequency of 9 Hz.

3. Results and discusions

The rods obtained by copper mold casting (Figure 1) were structurally analyzed by X-ray diffraction. In the diffraction pattern (Figure 2) can be observed only broad peaks characteristic of an amorphous structure.

The DSC curves of the elaborated alloys of $Fe_{48}Mo_{14}Cr_{15-x}Co_xB_6P_8Si_7Y_2$ (x = 0, 4, 7) are showed in Figure 3. The alloys exhibit an exothermic peak which marks the crystallization event and an endothermic peak for the melting. The glass transition temperature T_g, the crystallization temperature T_x, the melting temperature T_m and the liquidus temperature T_l for each alloy were determined and listed in Table 2. With increasing the Co content, the glass transition temperature T_g and the crystallization temperature T_x move to higher values, meanwhile the melting temperature and liquidus temperature T_l slightly moves to lower values.

Figure 1. BMG rod obtained by copper mold casting.

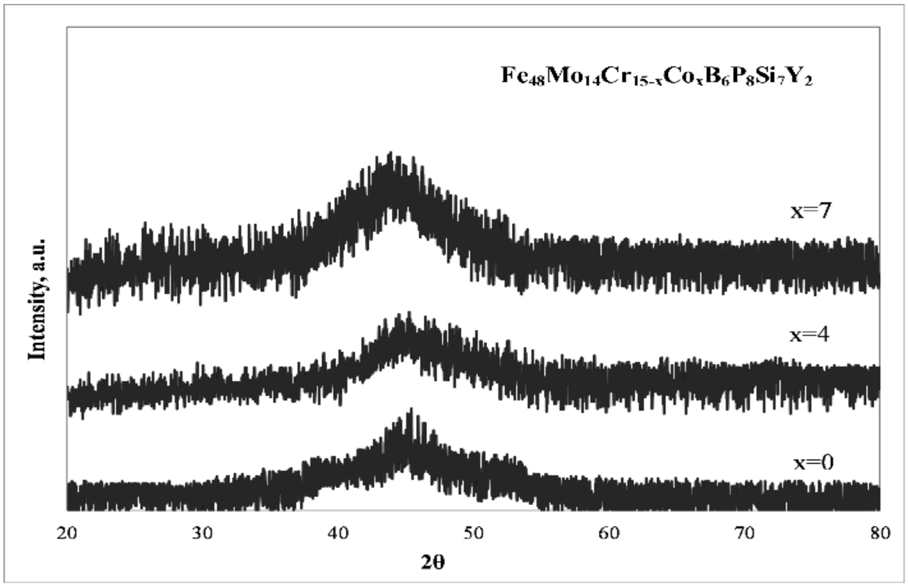


Fig 2. XRD patterns of BMG rods.

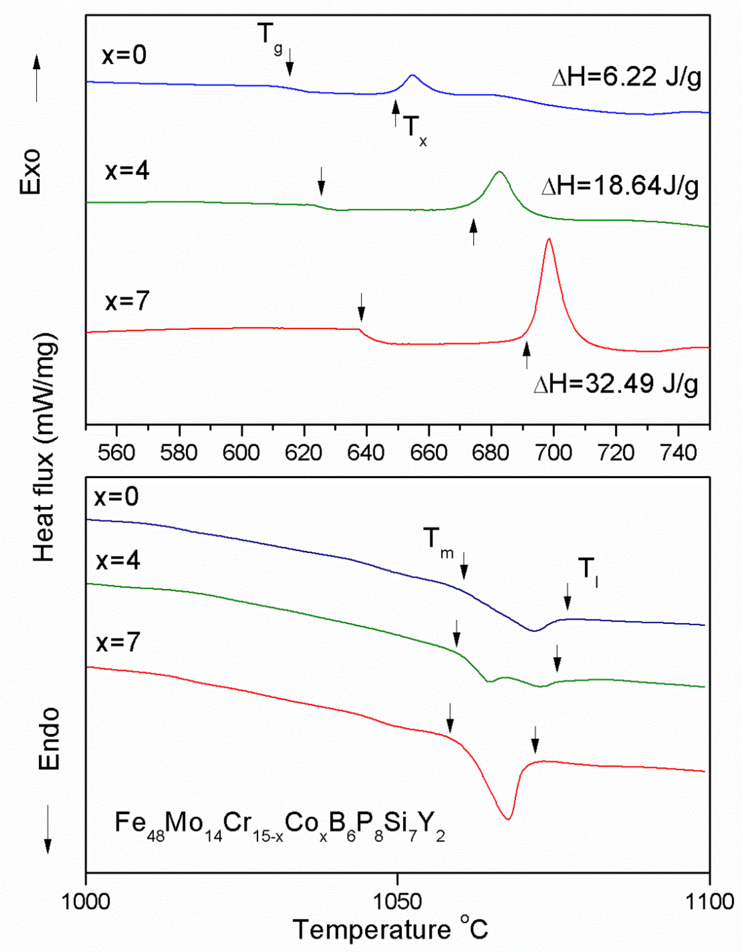


Fig 3. DSC curves of the $Fe_{48}Mo_{14}Cr_{15-x}Co_xB_6P_8Si_7Y_2$ BMGs.

Table 2. Transformation temperatures for $Fe_{48}Mo_{14}Cr_{15-x}Co_xB_6P_8Si_7Y_2$ BMG

Alloy **Temperatures**	**x=0**	**x=4**	**x=7**
T_g[°C]	614	628	633
T_x[°C]	649	675	692
T_m [°C]	1061	1059	1058
T_l [°C]	1077	1075	1072

The GFA evaluation of $Fe_{48}Mo_{14}Cr_{15-x}Co_xB_6P_8Si_7Y_2$ (x = 0, 4, 7) was done by calculating the proposed parameters listed in Table 3. Analyzing the results obtained by comparing them with the ideal ones, it was found that the partial addition of Co instead of Cr has led to a slight increase in the glass forming ability of Fe-Mo-Cr-Co-B-P-Si-Y family alloys. By adding Co, the T_l drops few degrees and the T_g and T_x increase to higher values. The most common parameter used for GFA estimation ΔT_x moves from 35° to 59°.

Table 3. GFA Parameters for $Fe_{48}Mo_{14}Cr_{15-x}Co_xB_6P_8Si_7Y_2$ BMG

Alloy **GFA Criterion**	**x=0**	**x=4**	**x=7**
$T_{rg}=T_g/T_l$	0.57	0.58	0.59
$\Delta T_x=T_x-T_g$	35	47	59
$\alpha=T_x/T_l$	0.60	0.63	0.65
$\beta=1+T_x/T_l$	1.60	1.63	1.65
$\gamma=T_x/(T_g+T_l)$	0.38	0.40	0.41
$\delta=T_x/(T_l-T_g)$	1.40	1.51	1.58

Since Co has a similar electronegativity with Fe [28], it presents a higher tendency to form solid solution as it indicates the Fe-Co binary phase diagram. Therefore, according to Jiao [29] the effect of Co on GFA is limited in a small proportion. Also, by increasing the number of components and the differences in atomic size and valence electron concentration between the alloy components according to Wang [28], the GFA can be improved. There is a large atomic size difference between Co and B, P and Si which leads to an increase of the atomic packing density, therefore enhancing the supercooled liquid region [2].

The thermal stability of the amorphous phase increases by the addition of Co, which is indicated by the growth of the enthalpy crystallization as resulted from DSC analysis.

For the same alloy family, the crystallization enthalpy may be used as a measure of the amorphous degree, the higher is the enthalpy, the more stable is the amorphous phase [3]. The amount of the crystallization enthalpy grows from 6.22 J/g to 32.49 J/g with Co addition which indicates a better glass forming ability.

In Figure 4 is represented the typical load–displacement (P–h) curves of as-cast rods (x=0, 4, 7) obtained from the constant loading rate of a 40mN/min. With the increase of Co content, the maximum indentation depth is decreased from 325 to 281 and to 248 nm, respectively.

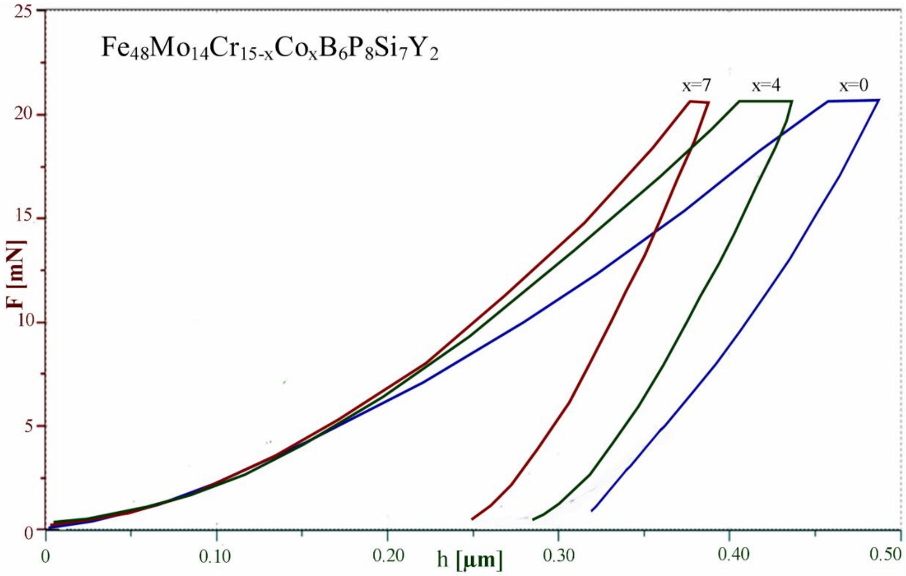


Fig 4. Typical Load–penetration depth (F–h) curves measured during nanoindentation tests.

This indicates that the hardness depends on Co content. The hardness, elastic modulus obtained by Oliver–Pharr method, are summarized in Table 4. The addition of Co induces mechanical hardening, manifested in an increase of indentation testing hardness (H_{IT}) and indentation elastic modulus (E_{IT}).

Table 4. Mechanical properties for $Fe_{48}Mo_{14}Cr_{15-x}Co_xB_6P_8Si_7Y_2$ by nanoindentation

Mechanical properties	**x=0**	**x=4**	**x=7**
H_{IT}, GPa	12.8 ± 0.5	13.9 ± 0.2	15.2 ± 0.4
E_{IT}, GPa	190.6 ± 3	206.1 ± 1	225.2 ± 3

Nanoindentation tests carried out showed that Co addition over Cr leads to a higher hardness (from 12.8 to 15.29 GPa) and elastic modulus (from 190.6 to 225.2 GPa). A higher content of Co, (over 4% at.) has a positive influence on hardness and elastic modulus, due to a more densely packed structure which was also noticed in other Fe-based BMGs [19], [22], [23].

Figure 5 presents the compressive stress-strain curves for the as-cast rods (x = 0, 4, 7) obtained by compression tests. It can be observed that the stress–strain curves present a purely elastic deformation without plastic deformation showing that it is a high resistance material.

The results derived from compression test are presented in Table 5. It can be noted that the Co addition increases the fracture strength. The values obtained are comparable with the data reported in literature for the Fe-based bulk amorphous alloys by Stoica et al.[30].

Table 5. Mechanical properties for $Fe_{48}Mo_{14}Cr_{15-x}Co_xB_6P_8Si_7Y_2$ by compression tests

Compressive strength, σ_f [MPa]	**x=0**	**x=4**	**x=7**
	2823 ±25	2885 ±20	2958 ±18

The compression tests showed also an increase of compressive strength from 2823 MPa to 2958 MPa with increasing the Co content. The compressive strength depends on the atomic size distribution and the chemical bonding between the atoms of the alloy elements. Increasing the atomic size distribution and bonding strength leads to a higher fracture strength [29]. These alloys containing metals and metalloids, form strong covalent bonds. Increasing the number of metals in the alloy can generate new covalent bonds [8]. Therefore the addition of Co, which forms chemical compounds with B, P, and Si increases the number of covalent bonds and thus increase the strength of the material.

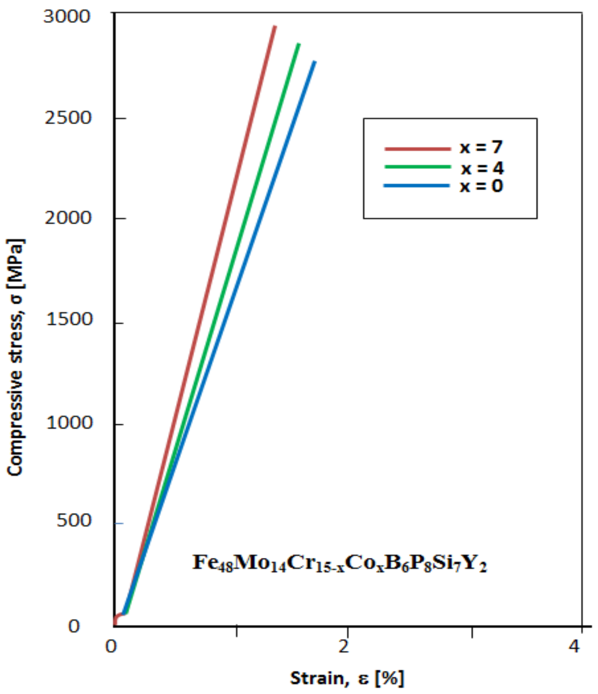
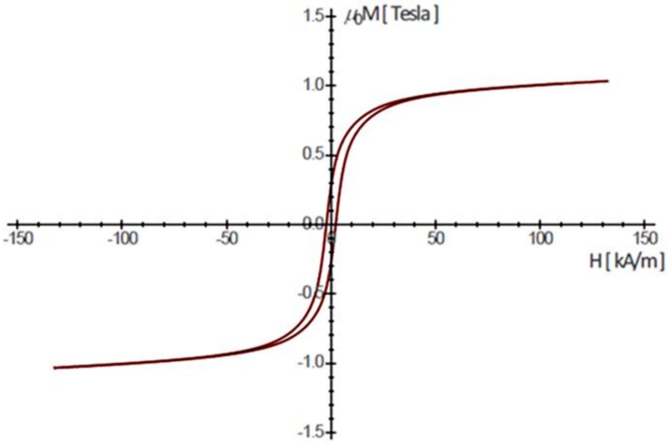


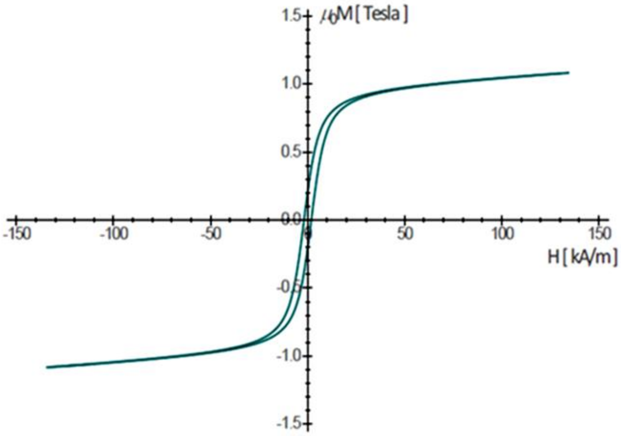
Fig 5. The stress-strain curves for $Fe_{48}Mo_{14}Cr_{15-x}Co_xB_6P_8Si_7Y_2$.

On the other hand, the presence of Co leads to a wider atomic size distribution that favors a short-distance arrangement of atoms, leading to a higher dense packing structure [8], [20]. The denser is the packing, the more covalent bonds are shorter and stronger and therefore the fracture strength is higher [8].

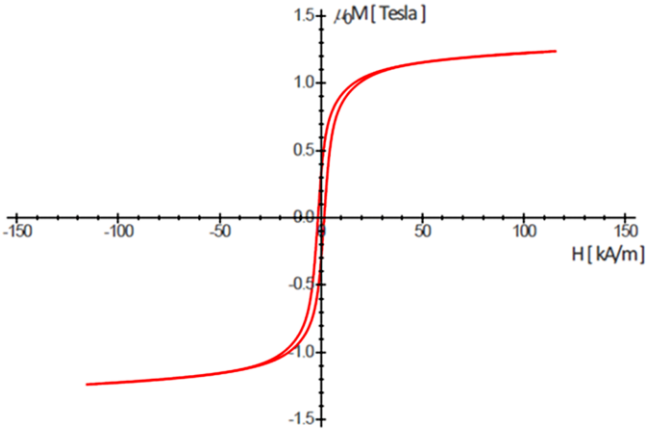
The magnetic hysteresis loops of the as-cast rods at room temperature are presented in Figure 6, and the values of saturation magnetization and coercivity are listed in Table 6.



a) x=0



b) x= 4



c) x=7

Fig 6. Magnetic hysteresis loops.

Table 6. Magnetic properties for $Fe_{48}Mo_{14}Cr_{15-x}Co_xB_6P_8Si_7Y_2$

Magnetic properties	x=0	x=4	x=7
Coercivity (H_C), A/m	8.76	6.18	5.24
Saturation magnetization (μ0Msat), T	0.95	1.16	1.21

The as-cast rods display good soft magnetic properties, the saturation magnetization increases and the coercivity decreases with Co. The values obtained are comparable with the data reported in literature for Fe-based BMGs [20], [27].

4. Conclusions

Bulk $Fe_{48}Mo_{14}Cr_{15-x}Co_xB_6P_8Si_7Y_2$ (x=0, 4, 7) metallic glasses in form of rods of 2 mm in diameter have been successfully obtained by copper mold casting method.

The GFA of the elaborated alloys was estimated using the following criterion T_{rg}, ΔT_x, α, β, γ, and δ. It was found that the partial addition of Co instead of Cr leads to an increase of the glass forming ability of Fe-Mo-Cr-Co-B-P-Si-Y family alloys which was confirmed also by the crystallization enthalpy values.

Nanoindentation and compression tests carried out showed that Co addition over Cr leads to a higher hardness, elastic modulus and compressive strength. Also, it was noted a slightly improvement in soft magnetic properties, higher saturation magnetization and lower coercivity.

Considering that the partial substitution of Cr with Co improves GFA, mechanical and magnetic properties it can be appreciated that Fe-Mo-Cr-Co-B-P-Si-Y family alloys represent an opportunity to develop the use of BMG in new engineering applications, like components of micro-electro-mechanical systems (MEMS) for example microgears, microsprings, sensors and actuators.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reports in this manuscript.

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