FERROMAGNETIC MICROWIRES WITH LOW CURIE TEMPERATURE FOR SENSOR APPLICATIONS

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Glass-coated ferromagnetic microwires, with low Curie temperature, used for temperature sensor were designed and complex characterized. Two alloys with low Curie temperature consisting of Fe-Ni-Cr-B-Si and Fe-Ni-Cr-Co-B-Si were developed. The microwires are covered by a thin layer of glass, thus increasing their corrosive protection, making the temperature sensors suited to work in corrosive environments. The active elements have a temperature range between 20°C and 200°C, with a sensitivity that depend on the number of microwires used in the design of the temperature sensor.

Key words: microwires, low Curie temperature, sensor

1. Introduction

The paper describes multidisciplinary research involving materials processing and development, structural characterization, microstructure, and magnetic measurements. The main task that we focused on was to synthesize and characterize low Curie temperature (Tc) ferromagnetic materials as microwires.

The development of metallic microwires technologies (2000) became a revolutionary high-tech theme for a variety of consecrated applications, opening new ways for technological advances, and allowing the development of new

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applications. The technology strong points are: precision, reliability, high productivity, process automation and multiple functional possibilities. In sensor applications considerable attention is given to the temperature behaviour of magnetic glass-coated microwires [1 - 6].

In the presented work, we focused on the manufacturing process of the alloys, and on the production of low Curie temperature microwires with predefined properties. Emphasis is placed on the reproducibility of the core dimensions, microstructure, and on the properties of the microwires, so that they exhibit their optimized structural and their magnetic properties for this type of sensors application. With regard to low Curie temperature (Tc) alloys, we developed several alloys from the Fe-Ni-Cr and Fe-Co systems.

One of the possible applications that we expect for these types of microwires is a temperature sensor. This application is based on the ferromagnetic-paramagnetic transition at the Curie temperature. Positive results were obtained. The Curie temperature of Fe and Co-rich amorphous microwires is about 300 - 400^{o}C. Addition of Cr and Ni, results in a decrease of Tc to room temperature. In this way, a wide variety of Tc microwires can be obtained that varies between room temperature and 400°C [3, 4, 7, 10].

2. Experimental

An induction furnace -Leybold – Heraeus- was used for the melting of the alloys. The pre-alloys were placed in the crucible in descending order of melting temperature. Temperature was increased to 1500^{o}C. At this temperature Si was introduced as a deoxidizer. After the melting of all elements, the temperature was increased to 1600^{o}C, in order to homogenize the alloy, which it is drawn into ø5 mm quartz tubes by a suction system.

For the microwire fabrication, the alloy rod is placed in a sealed glass tube. Reaching the exact temperature, at which the alloy is melting and the glass softens, allows a very thin glass capillary (ranging between 20 mm and 50 mm).

The molten metal, streams into the capillary and is quickly solidified by water cooling jet. Then the metal core is in an amorphous state, and the drawing process introduces frozen-in tensile stress, which is a source of the magnetic anisotropy in microwires [8, 9].

Ferromagnetic glass-coated microwires, based on the Fe-Ni-Cr and Fe-Co system alloys, were prepared by the Taylor-Ulitovsky method. Two microwires with different Curie temperature were prepared (see details in Table 1).

The obtained microwires were structurally characterized by X-ray diffraction using a BRUKER AXS D8 DISCOVER diffractometer, configured in θ-2θ geometry, in parallel beam radiation provided by the X-ray Copper anode tube, followed by a 0.6 mm Göebel mirror.

Table 1

Composition of studied microwires

Sample code	Composition %at.
IA 4	$Fe_{5.71}Cr_{3.4}Ni_{0.03}Co_{64.04}B_{15.88}Si_{10.94}$
IA 5	$Fe_{30}Cr_{12}Ni_{35}B_{13}Si_{10}$

The obtained microwires (without removing the glass cover) were placed as a fascicle on a quartz zero background sample holder and the diffractogram was recorded using the LynxEye 1D multichannel detector, with an angular increment of 0.040, increment of 5s/step, in the angular range 10-1000. The experimental determinations were made by positioning the microwires both along the primary beam and perpendicular to it. For both types of microwires the diffractograms along and in the perpendicular direction did not show any differences, suggesting the absence of crystallographic texture along the microwires. The experimental diffraction peaks were identified using the ICDD PDF2 Release 2014 database.

The microwires were complex characterized by VSM Lake Shore 7300 for magnetic proprieties and determination of Curie temperature by MPMS (Magnetically Properties Measurement System) plotting the M = f (T) curves. The field in which determinations were made was 100 Oe.

Chemical composition, quality and quantity of microwires obtained and determination of microwire's thickness were performed by scanning electron microscopy (SEM) with high resolution Carl Zeiss Auriga microscope, using EDS analysis. In the case of the two microwires samples from IA4 and IA5 alloys, the chemical analysis was performed on the surface of the metal core. As a global remark, in both cases, a uniform distribution of the chemical composition was noted.

3. Results and Discussion

Curves for Curie temperature are shown in Figs. 5 and 8. They indicated a Curie temperature of 84°C for the $Fe_{5.71}Cr_{3.4}Ni_{0.03}Co_{64.04}B_{15.88}Si_{10.94}$ (IA4) alloy and 100^{o}C for the $Fe_{30}Cr_{12}Ni_{35}B_{13}Si_{10}$ (IA5) alloy. Thickness determination of the microwires is presented in Figs. 1 and 2 and are explained in table 2. From the measurements done, it was observed that the microwires kept their diameter constant, including their metal core.

Table 2

Thickness of studied microwires

Sample code	D (μm)	d (μm)
IA 4	23.5	15.8
IA 5	23.1	17.9

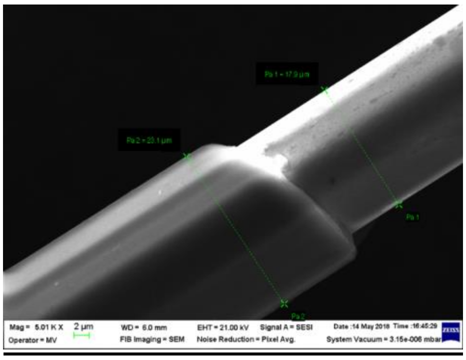

Fig.1. SEM Image of IA4 microwires

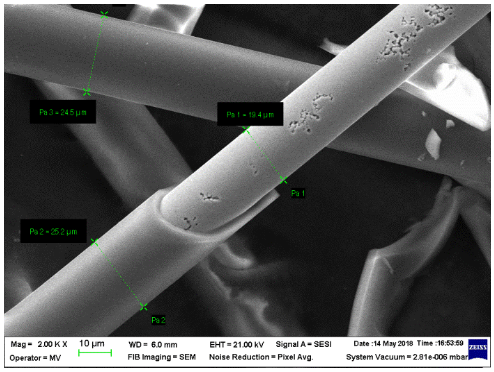

Fig.2. SEM Image of IA5 microwires

For the IA 4 type microwires the main diffraction peak (Fig 3) at the angular position 43.99°, respectively at the interplanar distance of 2,058 Å is associated with the crystallographic plane (111) specific to a face centred cubic phase of either Co (PDF 01-071-4651) or Cr-Fe-Ni (PDF 00-033-0945). Both theoretical patterns have the same crystallographic structure, belonging to the same spatial group Fm-3m (225), having the same lattice parameter of 3.554 Å. Experimentally, it was calculated the lattice parameter, a_{exp} = 3.564 Å. The other crystalline phases are evidenced by the presence of several diffraction peaks under the amorphous contribution of the solid solution formed due the rapid crystallization in the microwires manufacturing process, in the angular range 2θ: 40°-50°, a face centred cubic Fe3Co3Si2 (PDF 03-065-8930) and a hexagonal Co (PDF 01-089-4308) phase.

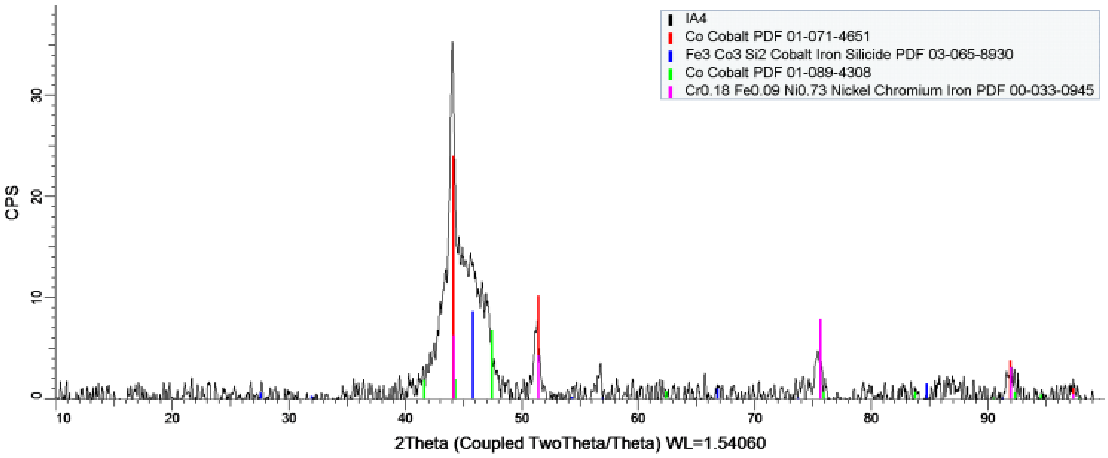

Fig.3. XRD patterns of IA 4 alloy

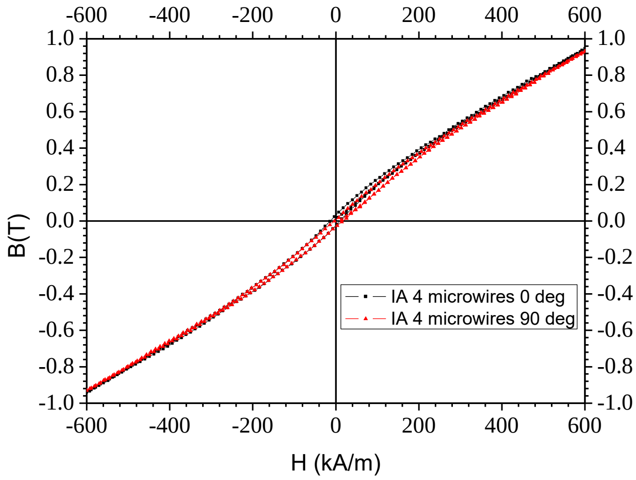

Fig.4. B (H) room temperature measurements of the IA4 microwires measured along the magnetic field and perpendicular to the magnetic field

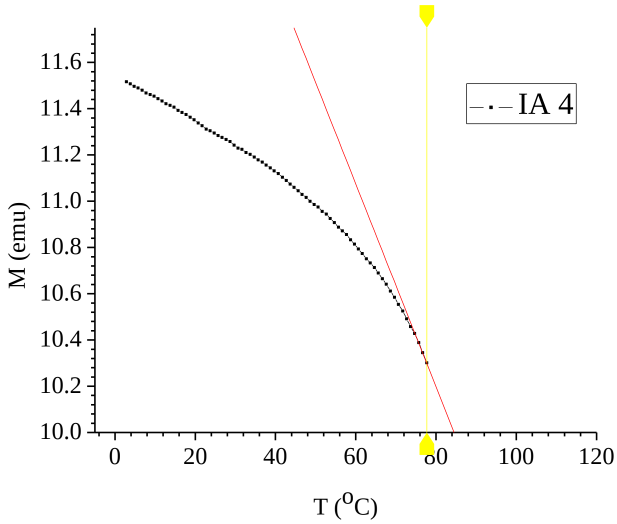

Fig. 5. M (T) measurement and the Tc determination point for the IA 4 alloy

For the IA 5 type microwires the main diffraction peak (Fig 6) at the angular position 43.94°, respectively at the interplanar distance of 2,061 Å is associated with the crystallographic plane (111) of a simple cubic phase of FeNi3, Pm-3m (221) space group (PDF 03-065-3244). We determined a lattice parameter of a_{exp} = 3.569 Å. It was noticed also the presence of two additional tetragonal crystalline phases of Cr7Ni3 (PDF 00-051-0637) and Fe4.9Si2.0 B1.0 (PDF 00-019-0626) under the amorphous contribution of the solid solution formed due the rapid crystallization in the microwires manufacturing process, in the angular range 2θ: 40°-50°, in a similar way as for IA4 type microwires.

From the magnetic point of view, the contribution of the crystalline cubic phases to the microwire anisotropy is minimal i.e. the microwires does not show magneto-crystalline anisotropy at room temperature.

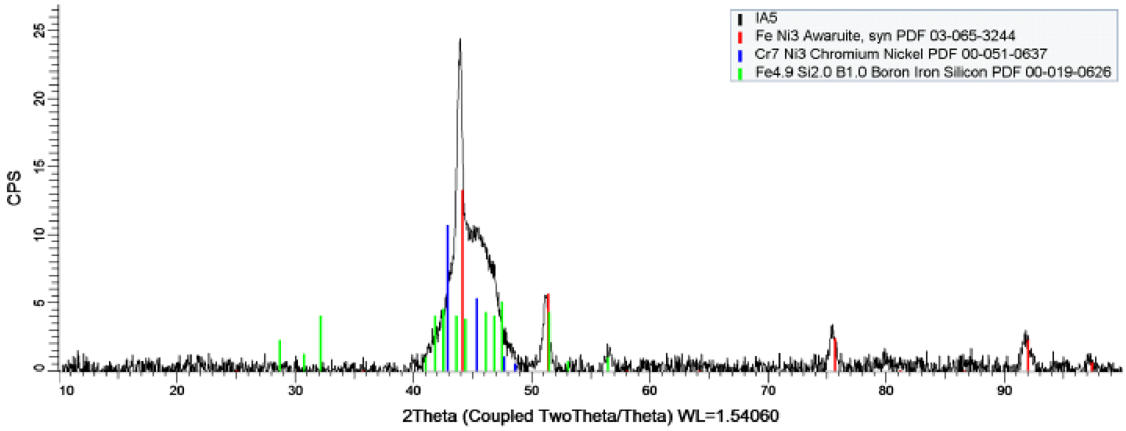

Fig. 6. XRD patterns of IA 5 microwire

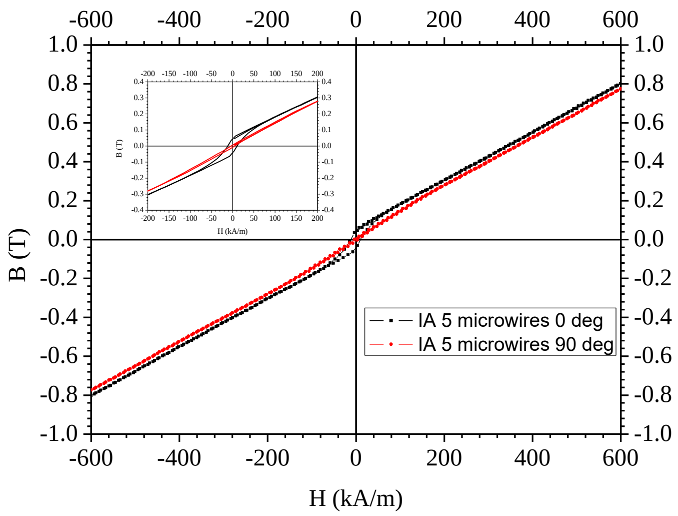

Fig.7. B (H) room temperature measurements of the IA5 microwires measured along the magnetic field and perpendicular to the magnetic field

In Figs. 4 and 7, we assume that the magnetic anisotropy is given by the gap structure from the interface between the glass and the metallic core.

Table 3

Magnetic characteristics of the microwires

Sample / measuring angle	M_r (emu/g)	M_S (emu/g)	H_c (Oe)
IA 4 / 0dgr	3.13	21.33	302.5
IA 4 / 90dgr	2.06	20.17	355.6
IA 5 / 0dgr	4.63	6.23	201.8
IA 5 / 90dgr	0.76	3.25	217.5

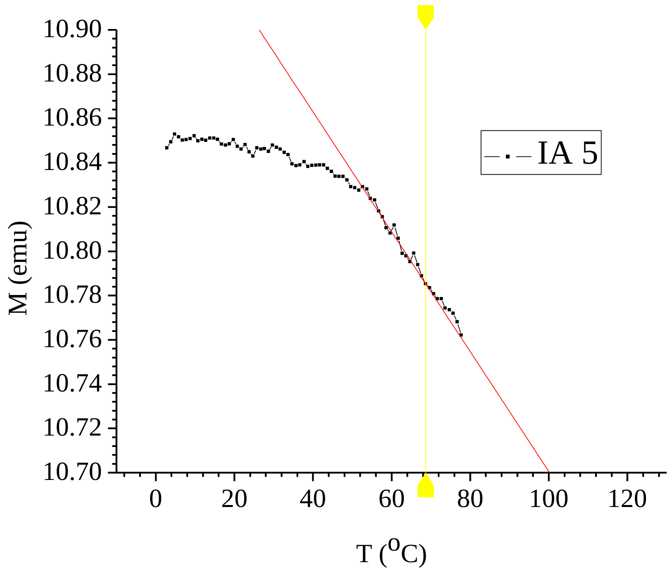

Fig. 8. Curve for Tc determination, IA 5

As can be observed, the chemical composition drastically affects their magnetization curves. The magnetic proprieties are presented in table 3.

Curie temperature Tc value depends essentially on the chemical composition and practically does not depend on the material structure. Also, the correlation between composition and Tc, has no linear trend, even within a single phase material. The predictability of Tc value is complicated even more because the material has 3 active magnetic components. The Curie temperature increases with the increase of Co content [6].

4. Conclusions

Microwires from $Fe_{5.71}Cr_{3.4}Ni_{0.03}Co_{64.04}B_{15.88}Si_{10.94}$ and $Fe_{30}Cr_{12}Ni_{35}B_{13}Si_{10}$ alloys with low Curie temperature were obtained and complex characterized. Based on the drastic change of magnetic properties near Curie temperature, one of the most common applications for these microwires, can be temperature sensors. We obtained microwires that are suited to be integrated into a temperature sensor working in temperature range of 20°C – 200°C. Also, these type of temperature sensors are ideal for the usage in corrosive environments, due to their special design of the active detectors, in our case the microwires.

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