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The effect of metalloid content on glass forming ability, thermal stability and magnetic properties of Fe-Ta-Si-C powders prepared by mechanical alloying

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ABSTRACT

In this study, the structural, thermal and magnetic characteristics of $Fe_{75-x}Ta_5Si_{10}C_{10+x}$ (x = 0, 5, 10) powders processed by mechanical alloying (MA) were investigated. The X-ray diffraction (XRD) indicated that by increasing the level of carbon, the structural refinement and lattice strain of the nanocrystalline Fe-based solid solution formed in the early stages of milling are enhanced. Furthermore, quantitative XRD analyses demonstrated that the glass forming ability (GFA) is remarkably improved by increasing the amount of carbon, where the percentage of amorphous phase increases from 56% for x = 0 to 98% for x = 10 after 120 h of milling. In addition, thermodynamic calculations, based on the extended Miedema's model, showed that the driving force for the glass formation increases with the concentration of carbon. Also, thermal analyses demonstrated that both the glass transition (T_g) and the onset of the crystallization (T_x) temperatures increase with the content of carbon, suggesting an enhanced thermal stability of the glassy phase. Additionally, the extension of the supercooled liquid region for the alloys with x = 5 and 10 reached a large value of 75 and 78 K, respectively, manifesting a high thermal stability of their supercooled liquid. Magnetic measurements showed that the soft magnetic behavior of the powders milled for 120 h is improved by increasing the level of carbon in terms of a decrease in coercivity (H_c) from 3.7 kA/m for x = 0 to 2.1 kA/m for x = 10. The evolution of the magnetic properties with annealing temperature reflected that the alloy with x = 10 displays a minimum H_c of 1.2 kA/m after annealing at 673 K.

1. Introduction

Recently, the invention of new materials with low loss has become essential to improve the energy efficiency in producing electricity. A low loss leads to saving energy and cost, as well as reducing CO_2 emissions [1–3]. In the field of energy saving, the amorphous alloys, especially the Fe-base ones received much attention due to their excellent soft magnetic properties, such as high saturation magnetization (M_s), very low coercivity (H_c), high magnetic permeability, and particularly very low core losses [4]. The magnetic amorphous alloys can be used in transformer cores, telecommunications and electronic devices working at high frequencies [5–7]. The two important factors that greatly affect the size and the noise of an amorphous core are low H_c and high M_s [8]. Accordingly, there is much demand for producing the new amorphous alloys with high M_s, low H_c and low costs. The choice of the constituents and composition of amorphous materials with high glass forming ability (GFA) and high Fe content is crucial in attaining high M_s. Moreover, the produced amorphous alloys should have sufficient GFA to achieve the excellent soft magnetic properties [9].

There are many different systems for the Fe-based amorphous alloys, such as Fe—B [10], Fe-B-Si [11], Fe-Si-C [12], Fe-P-C [13] and Fe-Si-B-P-C [14]. Among these systems, Fe-Si-C amorphous alloys are highly attractive, owning to the low cost of raw materials. Previously, it has been shown that the addition of P and C can improve GFA [15]. In addition, GFA can be enhanced further by the small addition of a transition element with a large negative heat of mixing (ΔH_{mix}) and a noticeable atomic size difference with the constituents, according to the Inoue's empirical rule [16]. Therefore, the simultaneous addition of metalloids and transition metals can be useful to promote the GFA, to improve the soft magnetic properties and to enhance the thermal stability of the amorphous phase [17–19].

The mechanical alloying (MA) is an effective process for producing the amorphous alloys due to its advantages over other methods, such as the low cost of equipment, easy control of process parameters, the

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overcoming of the restrictions of the phase diagrams and a greater flexibility in the choice of the constituent materials [20–22]. Furthermore, as compared to the casting process, the mechanically alloyed glassy powders can be converted into the bulk metallic glasses (BMGs)/composites with arbitrary dimensions or complicated shapes by a suitable powder metallurgy technique [23].

It has been shown that the formation of a single amorphous phase in the Fe-Si-C ternary system is possible by the MA process after 200 h milling, although no supercooled liquid region (SLR) was observed for the prepared alloy and although their magnetic behavior was not investigated [12]. In addition, the results indicated that glass formation in the Fe-Si-C system is provoked in the C-rich compositions, as compared to those rich in Si [12]. It has recently been showed that with the addition of Ta to the Fe-Si-C system, amorphization could take place faster and the obtained glassy alloy with a composition of $Fe_{70}Ta_5Si_{10}C_{15}$ exhibits a high thermal stability and tunable soft magnetic properties, depending on the milling time and annealing temperature [24]. To the best of our knowledge, the effects of the metalloids fraction, particularly C, on GFA, thermal stability and magnetic behavior of the mechanically alloyed Fe-Ta-Si-C powders were not investigated. In the present work, the influence of C content on the phase evolution, microstructure, powders morphology, thermal behavior and magnetic properties of the mechanically alloyed $Fe_{75-x}Ta_5Si_{10}C_{10+x}$ (x = 0, 5, 10) powders was studied.

2. Materials and methods

The starting materials for producing the $Fe_{75-x}Ta_5Si_{10}C_{10+x}$ (x = 0, 5, 10) powders consist of Fe (Merck, > 99.5%, $D_{avg} \sim 10\,\mu m$), Ta (Merck, > 99.5%, $D_{avg} \sim 40\,\mu m$), Si (Merck, > 99.5%, $D_{avg} \sim 44\,\mu m$) and C (Merck, > 99.5%, $D_{avg} \sim 150\,\mu m$). The prepared compositions were named Fe75, Fe70 and Fe65 for brevity. The MA was performed in a planetary ball mill (Sepahan 84D, Iran) at a rotation speed of 400 rpm for different periods. The vials and balls were made of hardened steel, and the total weight of the powders, the balls diameter and the ball-to-powder weight ratio (BPR) were 10 g, 10 mm and 20:1, respectively. Moreover, in order to avoid powders oxidation, the MA was carried out under a high-purity argon atmosphere (99.99%). The milling was conducted with a sequence of 30 min working, followed by 15 min of rest to prevent a large temperature rise. The characterization of the milled powders at selected times was carried out by X-ray diffraction (XRD, D8 advance Bruker, Cu $_{K\alpha}$ radiation). The measurements of the XRD patterns were performed at an angle range of 20–90° with the step size and time of 0.02° and 1 s, respectively. The Rietveld refinement was carried out using the MAUD software (version 2.33) [25]. The fitting process was performed by a degree 3 polynomial function for the background of each pattern, and Gaussian and loerntzian peaks were used to simulate the diffraction peaks of the amorphous and crystalline phases, respectively [13]. The fitting process was stopped when the difference between the experimental and calculated diffraction patterns reached the minimum and the ratio between residual factor (R_{wp}) and expected residual factor (R_{exp}) approached unity. The powders morphology and microstructure were studied by electron microscopy (SEM, Camscan mv2300) and transmission electron microscopy (TEM, Philips, Model CM120). For the TEM measurements, the powders were dispersed in ethanol using an ultrasonic probe for about 5 min and then a droplet of the ethanol-powder mixture was put on a copper grid. The thermal stability of the milled powders was evaluated by a differential scanning calorimeter (DSC, NETZSCH, 409 PC Luxx) at a constant heating rate of 20 K/min under a flow of highly pure argon. The magnetic hysteresis curves, saturation magnetization (M_s) and coercivity (H_c) at room temperature were measured by a vibrating sample magnetometer (VSM, Lake Shore, Model 7400). The values of the structural parameters, including the crystallite size and lattice strain, fraction of amorphous phase, average size of the particles and the magnetic parameters were reported as the average values of three measurements.

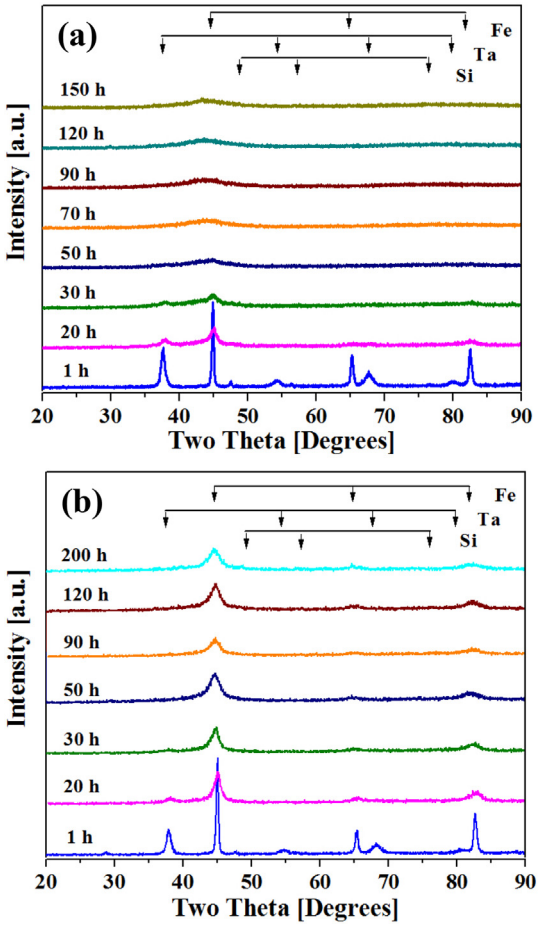

Fig. 1. The XRD patterns for different milling times: (a) Fe_{65} powders and (b) Fe_{75} powders.

In addition, the standard deviations from the corresponding mean values were reported as the random errors of the measured data.

3. Results and discussion

Fig. 1 shows the changes in XRD patterns with milling time for the Fe_{75} and Fe_{65} compositions. The corresponding XRD pattern for the Fe70 powder has been shown in Ref [24]. The figures show the diffraction peaks of all elements except C at the initial milling stage. The absence of C peaks is due to a low atomic scattering factor of this element. Furthermore, a solid solution of α-Fe is formed by milling up to 20 h. The patterns still show some undissolved Ta, even after milling up to 30 h. Generally, the diffusion rate of the interstitial elements, such as C is faster than substitutional ones like Si and Ta. This effect results from a smaller size of the interstitial atoms, as well as the existence of many empty interstitial sites in the lattice, which in turn decreases the corresponding activation energy for diffusion [26]. In addition, the diffusion rate of Si is higher than that of Ta due to its lower melting temperature (T_m). Hence, the diffraction peaks of Ta still exist even after 30 h of milling. Moreover, all patterns show a decreasing trend for intensity, followed by a notable increase in the peaks broadening with the extension of the milling time and the rate of these evolutions is faster by increasing the C fraction. The observed changes in the patterns can be attributed to the crystallite size refinement, as well as the introduction of the lattice strain and amorphization upon milling. According to Fig.1, a broad diffuse peak, characteristic of the amorphous phase, is obviously detected after 50 h milling for the Fe65 powders, while the other compositions, particularly Fe75 show sharper diffraction peaks at this milling stage, indicating a smaller fraction of the