Current Applied Physics 18 (2018) 411–416

Contents lists available at ScienceDirect

Current Applied Physics

journal homepage: www.elsevier.com/locate/cap

Unusual flow behavior of Fe-based soft magnetic amorphous ribbons under high temperature tensile loading

Jiyun Oh [a], Jin-Yoo Suh [b], Woo-Jin Kim [c], Jae-il Jang [c], Ki Hoon Kang [c], Chong Seung Yoon [c], Haein Choi-Yim [a, *]

[a] Department of Physics, Sookmyung Women's University, Seoul 04310, South Korea
[b] High Temperature Energy Materials Research Center, Korea Institute of Science and Technology, Seoul 02792, South Korea
[c] Department of Materials Science and Engineering, Hanyang University, Seoul 04763, South Korea

ARTICLE INFO

Article history:
Received 24 October 2017
Received in revised form
3 January 2018
Accepted 12 February 2018
Available online 13 February 2018

Keywords:
Fe-based amorphous alloy
Viscous flow
Nanoindentation
Soft-magnetic property

ABSTRACT

The viscous thermal flow behavior and mechanical property of $[Fe_{0.6}Co_{0.15}B_{0.2}Si_{0.05}]_{(100-x)}Ta_x$ (x = 0, 1, 2, 3, 4, and 5) soft magnetic amorphous ribbons were studied. The characteristics of melt-spun amorphous ribbons were measured by using vibration sample magnetometer (VSM), nanoindentation, differential scanning calorimetry (DSC) and thermo-mechanical analysis (TMA) to study the effects of Ta content variation on the thermal stability, mechanical, and soft magnetic properties. We observed that the nanoindentation hardness, Young's modulus, and glass transition and crystallization temperatures were improved by the addition of Ta. Using dilatometry measurement, TMA, by heating at a constant rate under tension mode, we examined not only the glass transition and crystallization behaviors but also the possibility of coexistence of multiple amorphous phases.

1. Introduction

The synthesis of an Fe-based amorphous alloy was first conducted with the Fe-P-C system in 1967 by Paul Duwez's group [1], and Fe-based bulk metallic glasses (BMGs), of the Fe-Al-Ga-P-C-B alloy system, were first synthesized in 1995 by Inoue et al. [2]. Thereafter, research on Fe-based amorphous alloys has been consistently pursued [3–8]. Fe-based amorphous alloys are well known for their excellent soft magnetic properties, such as high saturation magnetization (M_s) and low coercivity (H_c), as well as other promising engineering properties including physical, mechanical, and corrosion-resistance properties [9–11].

Among the Fe-based amorphous alloys, the Fe-Co-B-Si alloy system was reported to demonstrate promising soft magnetic properties [12–15]. Additionally, the substitution of Fe with Co turned out to be an effective method to enhance the M_s value of the amorphous metallic structure [16]. In tailoring the properties of interest, the addition of minor alloying elements and optimization of their concentrations are key processes in alloy development. The effect of a refractory element, Ta, on the magnetic and other properties of Fe-based amorphous alloys has rarely been reported [3,17]; systematic study on the effect of Ta concentration was little carried out in Refs. [3,17]. Therefore, the major effect of Ta concentration variation on the physical properties of Fe-based amorphous alloys including magnetic, mechanical, and thermomechanical properties needs to be clarified. Ta has a high density, high elastic modulus, and high melting point, and thus, the Ta is mainly used as a minor alloying element for improving various properties of alloys [18]. Therefore, in this study, we added Ta to the Fe-Co-B-Si alloy system and reported the effect of Ta addition and its variation in concentration on the mechanical and viscous flow behaviors.

In the development and characterization of Fe-based amorphous alloys, defining the glass transition temperature (T_g) is one of the most tricky tasks, probably owing to the limited glass-forming ability of the alloys, which is generally measured using differential scanning calorimetry (DSC) at a constant heating rate. The glass transition taking place during the heating of an amorphous structure is a transition from a structurally frozen state to a super-cooled liquid state, which is accompanied by an increase in heat capacity resulting in a slight deflection of the DSC baseline toward the direction of heat absorption. However, for most Fe-based amorphous alloys investigated for their soft magnetic properties, the DSC baselines do not show a sufficiently clear deflection to define the T_g.

* Corresponding author.
E-mail address: haein@sm.ac.kr (H. Choi-Yim).

https://doi.org/10.1016/j.cap.2018.02.005
1567-1739/

To date, the unclear T_g has been tentatively attributed to the limited glass-forming ability of the alloys, and no further studies have been performed in this regard. Additionally, the thermal behaviors of Fe-based amorphous alloys have been less thoroughly investigated than those of other alloys such as Cu-, Mg-, and Zr-based alloys [19–23].

In our previous work, we focused on the magnetic properties of the Fe-Co-B-Si-Ta amorphous alloy system with focusing on the variation of Fe/Co concentration ratio utilizing the positive effect of Ta addition by fixing its concentration [24,25]. Here, we focused on the thermal analysis of the amorphous phases and mechanical properties of the Fe-Co-B-Si-Ta amorphous alloy system. Moreover, we performed a different thermal analysis, utilizing dilatometry measurements of thin ribbons under tension mode to define the glass transition behavior of Fe-based amorphous alloys with varying chemical compositions. It turned out that the dilatometry measurement of amorphous ribbons not only enabled the successful evaluation of the glass transition and crystallization behavior, but also suggested the possibility of coexistence of multiple amorphous phases. Therefore, in this study, we reported the unusual viscous flow behavior of Fe-based amorphous alloy ribbons with varying Ta content and discussed the measured thermal stability of the amorphous phases and other properties.

2. Experiments

Multi-component alloys with nominal atomic percentage compositions of $[Fe_{0.6}Co_{0.15}B_{0.2}Si_{0.05}]_{(100-x)}Ta_x$ (x = 0, 1, 2, 3, 4, and 5) were prepared by arc-melting under a Ti-gettered argon atmosphere. The alloys were prepared by using high-purity metals of Fe (99.95%), Co (99.95%), B (99.5%), Si (99.999%), and Ta (99.95%). To minimize the chemical inhomogeneity, we remelted the alloy ingots at least six times. Amorphous alloy ribbons were rapidly solidified by melt-spinning method under an argon atmosphere with a copper wheel speed of 45.81 m/s. The as-spun amorphous ribbons had a thickness and width of approximately 30 μm and 2–2.5 mm, respectively.

The amorphous structure of the as-spun ribbons was confirmed by X-ray diffraction (XRD) with Cu-Kα radiation. The saturation magnetization, M_s, was measured by vibration sample magnetometer (VSM) under in-plane applied magnetic fields in the range from −1.0 to 1.0 T. The nanoindentation hardness (H_N) and Young's modulus (E) were examined by a nanoindentation tester (MTS Nanoindenter XP) with a pyramidal Berkovich diamond tip using a continuous stiffness measurement (CSM) technique at a controlled frequency 45 Hz. In CSM mode, the contact stiffness is continuously measured while the nanoindenter is loading at a regular frequency [26]. The nanoindentation was performed with a maximum load of 50 mN at constant loading/unloading rates of 1.67 mN/s and the indenter was held at the maximum load for 1 s. We also assigned a Poisson's ratio of 0.3. Each indentation test was repeated 20 times under the same experimental conditions to increase accuracy of the data and the average values of 20 separate measurements were used. The ribbon samples were mechanically polished down to 0.05 μm colloidal silica suspension to obtain a mirror-like appearance. After the nanoindentation experiment, the surface profile was examined by atomic force microscopy (AFM) and we applied a Pt-coating to the surface with indented regions to prepare for scanning electron microscopy (SEM) observations. Then, the appearance of each indentation was examined by SEM.

The crystallization temperature ($T_{x,\ DSC}$) was measured by DSC curves under an argon atmosphere at a heating rate of 0.34 K/s. Amorphous samples with an approximate mass of 20–30 mg were used for the DSC measurements. The viscous flow behavior of the super-cooled liquid region, temperature dependence of the relative displacement, T_g, and crystallization temperature ($T_{x,\ TMA}$) were measured by thermomechanical analysis (TMA) using the thermomechanical analyzer device under a tensile loading mode. The as-spun ribbons were directly cut by using scissors to be installed in the sample holder of the TMA apparatus with a gauge length of 10 mm (shown in Fig. 8(a)). The applied load and heating rate were 700 mN and 0.17 K/s, respectively.

3. Results and discussion

The chemical composition with the nominal atomic percentage of the Fe-Co-B-Si-Ta alloy system is given in Table 1. Fig. 1 shows the XRD patterns of the $[Fe_{0.6}Co_{0.15}B_{0.2}Si_{0.05}]_{(100-x)}Ta_x$ (x = 0, 1, 2, 3, 4, and 5) melt-spun ribbons. The XRD patterns show only broad halo humps without any crystalline peaks, which indicates the non-periodic atomic arrangement of an amorphous phase.

The intrinsic magnetic property of the Fe-Co-B-Si-Ta melt-spun ribbon was studied by using VSM. Fig. 2(a) shows hysteresis M-H loops of the melt-spun ribbons; Fig. 2(b) shows the dependency of M_s on the Ta content and the obtained values of M_s are also listed in Table 2. It is obvious from the hysteresis M-H loops that all the amorphous ribbons have soft magnetic characteristics. The highest and lowest values of M_s were 149.6 and 101.2 emu/g for the $Fe_{60}Co_{15}B_{20}Si_5$ (x = 0) and $Fe_{57}Co_{14.25}B_{19}Si_{4.75}Ta_5$ (x = 5) ribbons, respectively. As the Ta content increased from 0 to 5 at.%, M_s decreased from 149.6 to 101.2 emu/g. This significant drop in M_s appeared to be more than that expected owing to the change in Fe content from 60 to 57 at.% (with Ta contents ranging from 0 to 5 at.%). Although this is difficult to explain, the unusual behavior might suggest the existence of multiple amorphous phases and the variation of the relative phase fractions as a function of Ta concentration, for example, a Fe-rich clustering contributing to high M_s value that could exist in the amorphous structure could have been suppressed by the addition of Ta. No direct proof has been provided yet, but indirect evidences are reported in the following sections. Nevertheless, further study seeking the direct evidence for the discussion given above should be conducted.

The mechanical properties such as hardness and modulus are shown in Fig. 3(a) and (b). Typical load–displacement (p–h) curves of the Fe-Co-B-Si-Ta melt-spun ribbons are shown in Fig. 3(a) except for the thermal drift section. With the increase in Ta content, the maximum indentation depth decreased from 1086.16 to 535.11 nm, which indicates that the hardness is significantly influenced by the Ta content. Additionally, it was confirmed that the addition of Ta induces mechanical hardening of the ribbons. H_N and E are presented in Fig. 3(b) with error bars, which indicate the scattering of the values acquired by 20 repeated indentations. As shown in Fig. 3(b), the H_N and E values consistently increase with increasing Ta content, and the average values of H_N and E are listed in Table 2. The maximum values of H_N and E are 11.90 and 112.31 GPa, respectively, for the $Fe_{57}Co_{14.25}B_{19}Si_{4.75}Ta_5$ (x = 5) alloy. The H_N values range from 5.44 to 11.90 GPa and the E values range from 21.66 to 112.31 GPa. It should be noted here that Ta addition to a level of 5 at.% induced a two-fold increase in H_N and a five-fold

Table 1
Chemical composition of Fe-Co-B-Si-Ta amorphous alloys (at.%).

$[Fe_{0.6}\ Co_{0.15}\ B_{0.2}\ Si_{0.05}]_{(100-x)}Ta_x$	Fe	Co	B	Si	Ta
x = 0	60	15	20	5	0
x = 1	59.4	14.85	19.8	4.95	1
x = 2	58.8	14.7	19.6	4.9	2
x = 3	58.2	14.55	19.4	4.85	3
x = 4	57.6	14.4	19.2	4.8	4
x = 5	57	14.25	19	4.75	5