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ScienceDirect

Acta Materialia 61 (2013) 6609–6621

Acta MATERIALIA

www.elsevier.com/locate/actamat

Fabrication and characterization of bulk glassy $Co_{40}Fe_{22}Ta_8B_{30}$ alloy with high thermal stability and excellent soft magnetic properties

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Received 18 April 2013; received in revised form 19 July 2013; accepted 20 July 2013
Available online 16 August 2013

Abstract

In this work, $Co_{40}Fe_{22}Ta_8B_{30}$ alloy as a new bulk metallic glass with a wide supercooled liquid region of 74 K and excellent soft magnetic properties was prepared by the powder metallurgy method. Glassy $Co_{40}Fe_{22}Ta_8B_{30}$ powders were obtained by ball milling of melt-spun glassy ribbons at a cryogenic temperature and subsequently consolidated by hot pressing into disk-shaped specimens 10 mm in diameter and 2 mm thick. It was found that the new glassy alloy exhibited the largest diameter compared with the other Co-based bulk metallic glasses produced in the well-known Co–Fe–Ta–B alloying system up to now. The influence of the consolidation time on the microstructure and magnetic properties of the bulk samples was investigated by X-ray diffraction, differential scanning calorimetry, vibrating sample magnetometry and Faraday magnetometry. The results indicate that the new alloy exhibits a long incubation time before crystallization upon annealing above the glass transition temperature: noticeably longer than for other known (Co,Fe)-based amorphous alloys. The glassy bulk sample consolidated for 600 s at 923 K had a relative density of 99.2%, a saturation magnetization of 46.6 A m^2 kg^{-1}, a Curie temperature of 425 K and a low coercivity of 6 A m^{-1}. In addition, the new bulk glassy alloy exhibited ultra-high hardness of 13.48 GPa. The mechanisms by which the thermal stability and incubation time prior to crystallization increase are explained in accordance with pair correlation function analysis.

Keywords: Bulk metallic glass; Magnetic properties; Pair correlation function

1. Introduction

Bulk metallic glasses (BMG) have attracted increasing interest owing to their excellent properties, such as ultra-high mechanical strength, high corrosion resistance, high electrochemical discharge capacity and soft magnetic properties [1–4]. Multicomponent BMG several millimeters thick have been produced by rapid solidification techniques such as copper mold casting of different alloy systems, e.g., Pd-based, Zr-based, Cu-based, Ni-based, Fe and Co-based alloys [3]. BMG with shapes and dimensions that cannot be produced by direct casting may be accessible by powder processing routes [5]. For this purpose, glassy bulk specimens could be produced by the consolidation of glassy powders above the glass transition T_g in the supercooled liquid region (SLR), where the viscosity of the powders is significantly suppressed [5]. In order to achieve a high density product upon compaction, it is essential to allow enough consolidation time in the temperature range of the SLR, without crystallizing the sample. Therefore, a large width of SLR and long incubation time before crystallization are essential requirements for sintering.

Co-based BMG are well known for their excellent soft magnetic properties, such as very low coercivity and high magnetic permeability [1,6]. Almost 10 years ago, glassy $Co_{43}Fe_{20}Ta_{5.5}B_{31.5}$ rods 2 mm in diameter, with an ultra-high fracture strength of 5 GPa, low coercivity of 0.25 A m^{-1} and maximum magnetic permeability of 550,000, were produced by copper mold casting [1,7].

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1359-6454/$36.00
http://dx.doi.org/10.1016/j.actamat.2013.07.045

Glassy $Co_{43}Fe_{20}Ta_{5.5}B_{31.5}$ alloy showed the maximum width of SLR (72 K) in the composition range 5–30 at.% Fe, 4–6 at.% Ta and 20–33 at.% B [1]. In contrast to its high glass-forming ability compared with other Co-based BMG, annealing of $Co_{43}Fe_{20}Ta_{5.5}B_{31.5}$ alloy in the SLR results in crystallization with very short incubation times, even when the annealing temperature is slightly above T_g. As an example, the $(Co,Fe)_{21}Ta_2B_6$ phase precipitates upon annealing at 928 K for 60 s, i.e., just 18 K above the T_g [1].

In this work, $Co_{40}Fe_{22}Ta_8B_{30}$ BMG (i.e., with higher Ta content compared with $Co_{43}Fe_{20}Ta_{5.5}B_{31.5}$ alloy) was produced by the consolidation of glassy powders above the corresponding T_g. Previous results indicated that the mechanical alloying process was unsuccessful at producing fully glassy $Co_{40}Fe_{22}Ta_8B_{30}$ powders [8,9]. The unreacted boron inclusions, besides the small fraction of nanocrystalline α-(Fe, Co) phase, remained in the glassy matrix even after milling for 200 h [8,9]. In the current research, the glassy powders were obtained by controlled ball milling of melt-spun glassy ribbons with a wide SLR of 74 K and a significantly longer incubation time prior to crystallization compared with the $Co_{43}Fe_{20}Ta_{5.5}B_{31.5}$ alloy. Subsequent consolidation of the milled powders allowed bulk glassy samples 10 mm in diameter and 2 mm thick to be produced. Besides a very good thermal stability, the resulting samples possess a high relative density of 99.2%, low coercivity of 6 A m^{-1} and ultrahigh hardness of 13.48 GPa. To the present authors' knowledge, there are no reports on the fabrication of Co-based BMG more than 3 mm in diameter in the Co–Fe–Ta–B system with such excellent thermal stability and good soft magnetic properties.

2. Experimental procedure

A master alloy with nominal composition of $Co_{40}Fe_{22}Ta_8B_{30}$ (at.%) was prepared by arc-melting of pure Co (99.9%), Fe (99.95%), Ta (99.96%) and crystalline B (99.5%) under a Ti-gettered Ar atmosphere. Amorphous ribbons (width 3.5 mm, thickness 29 μm) were prepared under Ar flow by a single-roller Bühler melt spinner on a copper wheel rotating at 41 m s^{-1} tangential velocity. The ball milling of the ribbons was performed on a planetary ball mill (Retsch PM4000), using hardened steel balls 10 mm in diameter and hardened steel vials with a capacity of 250 ml. Before the milling, the as-cast ribbons were cut into small pieces (3.5×10 mm^2). A typical charge of 4 g of ribbons was milled for 3 h at a ball-to-powder weight ratio of 15:1 and a rotation speed of 200 rpm. To avoid a strong temperature rise, the milling was interrupted at each 15 min of working, and the vials were cooled down for 20 min in a bath of liquid nitrogen. To minimize oxidation, the sample handling was carried out in a glove box under purified argon atmosphere (<1 ppm O_2 and H_2O). The as-milled powders were sieved to <100 μm and used for the subsequent consolidation. Disk-shaped samples 10 mm in diameter and 2 mm thick were prepared under an argon atmosphere by consolidation of the powders, using a uniaxial hot pressing machine. The powders were placed under a pressure of 800 MPa, heated at a rate of 20 K min^{-1} to 923 K and kept at this temperature for different pressing times, then cooled down to ambient temperature using a forced flow of Ar gas.

The phase analysis of the samples was performed using X-ray diffraction (XRD) in reflection configuration, using Co K_α ($\lambda = 1.7889$ Å) radiation, as well as in transmission configuration, using a high-intensity, high-energy monochromatic synchrotron beam with an incident beam energy of 100 keV at the BW5 beamline at Hamburger Synchrotronstrahlungslabor, HASYLAB (Deutsches Elektronen-Synchrotron, DESY, Hamburg, Germany). The two-dimensional XRD patterns were integrated to the Q-space, where Q is the wave vector, using the FIT2D software package [10]. The integrated data were corrected for polarization, sample absorption, fluorescence contribution and inelastic scattering using the pair correlation function PDFgetX2 software [11]. The total structure factor $S(Q)$ was determined from the normalized elastically scattered intensity according to Faber–Ziman equation [12]. The pair distribution function $PDF(r)$ was calculated as

$$PDF = 1 + \frac{1}{2\pi^2\rho_0 r}\int_0^{Q_{max}} Q(S(Q)-1)\sin(Qr)dQ \qquad (1)$$

where ρ_0 is the average atomic number density.

The microstructure was characterized by high-resolution transmission electron microscopy (HRTEM; Tecnai F30 operating at 300 kV) and scanning electron microscopy (SEM; Hitachi TM 1000). The thermal stability was investigated with a differential scanning calorimeter (NETZSCH DSC 404) at a 20 K min^{-1} heating rate under a flow of high-purity argon. The viscosity of the samples was measured by parallel plate rheometry using a Perkin-Elmer thermomechanical analyzer (TMA7) device operated at a heating rate of 20 K min^{-1} and under a static load of 2.6 N. The density of ribbons and bulk samples was measured by the Archimedes method, using a computer-controlled microbalance and glycol as the working liquid. The final values were obtained by averaging over 50 experimental values. The Vickers hardness was determined for the bulk samples using a computer-controlled Struers Duramin 5 Vickers hardness tester. The tests were performed using a typical diamond indenter in the form of a pyramid with a square base and an angle of 136° between opposite faces, applying a load of 1.96 N for 10 s.

For magnetic investigations, the M–H hysteresis loops were recorded by vibrating sample magnetometry (VSM) at ambient temperature. For the bulk samples, VSM measurements were carried out on rectangular specimens (8×4 mm^2) cut from the consolidated disks. The coercivity was measured using a Foerster Coercimat under an applied field high enough to saturate the samples. The variation in magnetization with temperature and the Curie temperature T_c of the samples were determined using a