SOFT MAGNETIC PROPERTIES OF BULK AMORPHOUS Co-BASED SAMPLES[1]

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Ball milling of melt-spun ribbons and subsequent compaction of the resulting powders in the supercooled liquid region were used to prepare disc shaped bulk amorphous Co-based samples. The several bulk samples have been prepared by hot compaction with subsequent heat treatment (500°C – 575°C). The influence of the consolidation temperature and follow-up heat treatment on the magnetic properties of bulk samples was investigated. The final heat treatment leads to decrease of the coercivity to the value between the 7.5 to 9 A/m.

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1 Introduction

Co-based alloys exhibit excellent soft magnetic properties in the amorphous state [1, 2]. The ribbon, the shape in which amorphous or nanocrystalline material is usually prepared, is in many cases not suitable shape for applications, therefore it is logical to attempt to prepare such material in a more bulk form, for example in the form of a cylinder or a ring, that would be more convenient for industrial applications. Recently, Inoue et al. found some bulk amorphous and nanocrystalline alloys in Fe-(Al, Ga)-(P, C, B, Si) [3] and (Fe, Co, Ni)-(Zr, Hf)-B [4, 5] systems, which combine a large glass-forming ability with good soft magnetic properties of the amorphous phase. An alternative way of producing bulk material makes use of powder metallurgy methods [6, 7]. Ball milling technique has been successfully used to prepare many alloys in powder form which are therefore suitable for compaction into a variety of shapes [8]. For our investigations we have chosen $Co_{70.3}Fe_{4.7}Si_{10}B_{15}$ alloy with close to zero magnetostriction [9] and $Co_{56}Fe_{16}Zr_8B_{20}$ alloy with good stability of permeability up to high frequencies with a supercooled liquid region of 39 K before crystallization [4].

2 Experimental

Amorphous $Co_{70.3}Fe_{4.7}Si_{10}B_{15}$ and $Co_{56}Fe_{16}Zr_8B_{20}$ alloys in the form of thin ribbons were prepared by melt spinning. The as-quenched ribbons were subsequently short time milled using a RETSCH PM4000 planetary ball mill. The milling was performed under argon atmosphere

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with speed of 200 rpm at a ball-to-powder mass ratio of 6:1. The milling times were chosen so that to avoid structural changes during the milling and prepare fine-grained powders. Amorphous $Co_{70.3}Fe_{4.7}Si_{10}B_{15}$ and $Co_{56}Fe_{16}Zr_8B_{20}$ ribbons were mechanicaly milled for 4h and 1h, respectively. All powder handling was carried out in glove box under argon atmosphere (less than 1 ppm O_2 and H_2O). The powder consolidation into bulk samples (cylindrical discs: 10 mm in diameter and 3 mm in thickness) was done in a uniaxial hot press under argon atmosphere. During the consolidation experiments the powders were constant-rate heated at 60 K/min to the consolidation temperature, held isothermally at this temperature and pressed for 2 min using a pressure of about 900 MPa. The amorphous nature of the samples was proved by X-ray diffraction (XRD) experiments with Co Kα radiation ($\lambda = 0.17889$ nm) (Philips PW 3020) . The thermal stability of the samples was analyzed by a differential scanning calorimetry (DSC) (NETZSCH DSC 404) at 20 K/min heating rate under flowing argon. The glass transition temperature (T_g), the crystallization onset temperature (T_x), the extension of the supercooled liquid region ($\Delta T_x = T_x - T_g$) and the crystallization enthalpy (ΔH) were determined from the DSC scans. The temperature dependence of magnetization (from 300 up to 1073 K) was measured using a vibrating sample magnetometer (VSM) at 10 K/min heating rate, applying constant magnetic field with the strength of $\mu_0 H = 0.3$ T. The coercivity (H_c) of the compacted bulk samples was measured by a coercimeter Förster Koerzimat. In order to prepare ring-shaped samples more suitable for magnetic measurements, selected as-compacted samples were drilled using spark plasma erosion. We have annealed the samples at 500°C, 525°C, 550°C, 575°C a 600°C for 30 minutes at 5 K/min heating rate under flowing argon. The saturation polarization (J_s) and H_c of ring-shaped samples were determined from hysteresis loops traced in a maximum applied field of 5 kA/m using DC-hysteresigraph. All magnetic measurements were performed at room temperature. The density (ρ) of the ribbon and the consolidated bulk specimens was measured using the Archimedes principle with bromoform as the immersion fluid. The losses were measured for magnetic cores, wound into a toroid at sinusoidal induction in the cores with feedback and computer supporting the measuring unit.

3 Results and discussion

The XRD measurements indicated that the mechanical milling of amorphous as-spun ribbons of $Co_{70.3}Fe_{4.7}Si_{10}B_{15}$ and $Co_{56}Fe_{16}Zr_8B_{20}$ has no substantial influence on their structure and both samples remain amorphous [10]. The DSC scans for $Co_{70.3}Fe_{4.7}Si_{10}B_{15}$ and $Co_{56}Fe_{16}Zr_8B_{20}$ in the as-quenched state reveal the presence of a supercooled liquid region before single-stage crystallization. Ball milling of both mentioned alloys causes the change from a single-stage crystallization mode to a two-stage crystallization mode, while maintaining the supercooled liquid region before crystallization [10]. Furthemore, our recent studies showed that the bulk samples obtained by consolidation in the supercooled liquid state at temperatures slightly lower then the onset of crystallization exhibit magnetic properties comparable to those of corresponding as-spun ribbons. It is well known that the supercooled liquid state provides easy flow and relatively low viscosity, which can be utilized for the fabrication of high density and full strength bulk metallic glasses by powder consolidation. Therefore we decided to consolidate $Co_{70.3}Fe_{4.7}Si_{10}B_{15}$ powder at 540°C and $Co_{56}Fe_{16}Zr_8B_{20}$ powder at 600°C, respectively.

Structural relaxation of amorphous alloys can be also reached by annealing the amorphous

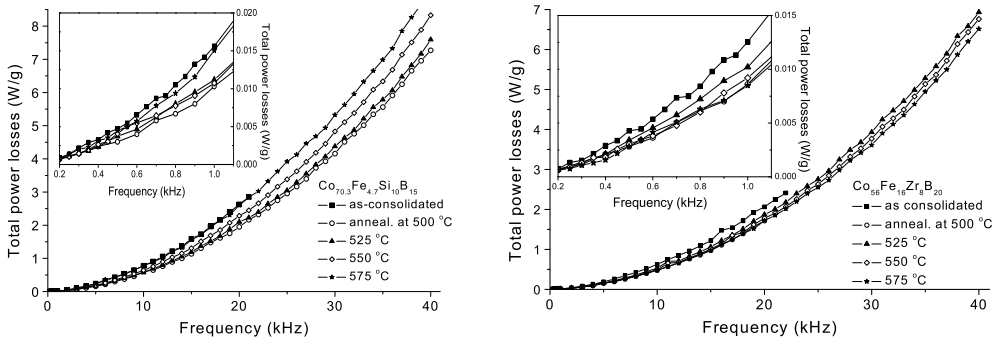

Fig. 1. Dependencies of total power losses on frequency for $Co_{70.3}Fe_{4.7}Si_{10}B_{15}$ and $Co_{56}Fe_{16}Zr_8B_{20}$ bulk samples.

samples. Accordingly, a series of bulk samples was prepared with annealing temperature T_{an} ranging from 500 up to 600 °C. In order to measure the soft magnetic properties of the consolidated bulk samples by the fluxmetric method, ring-shaped samples were made from the consolidated discs by spark erosion. Fig.1 presents the evolution of the total power losses as a function of the annealing temperature in the frequency range 200 Hz-50 kHz at B_{max}=0.2T (as-consolidated samples up to 22 kHz). Total power losses of samples after annealing at 600°C were too high for our measurement. These experiments indicated that the annealing temperatures 500 °C and 575 °C seem to be optimal for thermal treatment of amorphous bulk samples $Co_{70.3}Fe_{4.7}Si_{10}B_{15}$(1.72 W/g) and $Co_{56}Fe_{16}Zr_8B_{20}$ (1.95 W/g), respectively (at f=20 kHz, B_{max}=0.2 T). The subsequent increase of the annealing temperature results in the onset of the nucleation leading to the increase of the coercivity and total losses. It seems that crystalline phase pins domain wall displacement. Soft magnetic behaviour of our samples is strongly related to the existence of the amorphous phase. The coercivity of the compacted bulk samples was determined from demagnetization curves. Table 1. shows a detailed comparison of the magnetic properties of the samples in the form of ribbons and rings.

Tab. 1. The density and DC magnetic properties of $Co_{70.3}Fe_{4.7}Si_{10}B_{15}$ and $Co_{56}Fe_{16}Zr_8B_{20}$ bulk samples and as-quenched ribbons.

Sample		ρ [kg/m^3]	J_s [T]	H_c[A/m]
$Co_{70.3}Fe_{4.7}Si_{10}B_{15}$	ribbon	7510	0.83	3
	ring	6890	0.77	23
	anneal. ring at 500°C	6890	0.77	9
$Co_{56}Fe_{16}Zr_8B_{20}$	ribbon	7570	0.88	7
	ring	7490	0.87	13
	anneal. ring at 575°C	7490	0.87	7.5

The saturation polarization and the coercivity of ring samples were determined from magnetic hysteresis loops traced in a maximum applied field of 3 kA/m using DC-hysteresigraph. Consequently, the bulk $Co_{70.3}Fe_{4.7}Si_{10}B_{15}$ and $Co_{56}Fe_{16}Zr_8B_{20}$ glassy alloys should be useful as an engineering material because of the bulk shape as well as due to its good soft magnetic properties.

4 Conclusion

$Co_{70.3}Fe_{4.7}Si_{10}B_{15}$ and $Co_{56}Fe_{16}Zr_8B_{20}$ bulk amorphous samples were prepared by hot pressing of powders obtained by ball milling of pieces of melt-spun ribbons. Hot pressing followed by heat treatment allows to reduce the stress introduced into the material during ball milling and consolidation. Thermal treatments at temperatures below glass transition temperature lead to the improvement of the magnetic properties. This method enables the preparation of bulk samples with larger dimensions than specimens prepared by copper mold casting and with very good soft magnetic properties comparable to those of the corresponding as-quenched ribbons.

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