Materials and Design 86 (2015) 703–708

Contents lists available at ScienceDirect

Materials and Design

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

Fabrication of Fe-based bulk metallic glass by selective laser melting: A parameter study

Hyo Yun Jung [a,*], Su Ji Choi [b], Konda G. Prashanth [a], Mihai Stoica [a,c], Sergio Scudino [a], Seonghoon Yi [b], Uta Kühn [a], Do Hyang Kim [d], Ki Buem Kim [e], Jürgen Eckert [a,f]

[a] IFW Dresden, Institute for Complex Materials, Helmholtzstr. 20, D-01069 Dresden, Germany
[b] Department of Materials Science and Metallurgical Engineering, Kyungpook National University, 702-701 Daegu, South Korea
[c] Politehnica University of Timisoara, P-ta Victoriei 2, 300006 Timisoara, Romania
[d] Center for Non-crystalline Materials, Department of Metallurgical Engineering, Yonsei University, 120-749 Seoul, South Korea
[e] Department of Advanced Materials Engineering, Sejong University, 143-747 Seoul, South Korea
[f] University of Technology Dresden, Institute of Materials Science, D-01062 Dresden, Germany

ARTICLE INFO

Article history:
Received 10 March 2015
Received in revised form 10 July 2015
Accepted 27 July 2015
Available online 30 July 2015

Keywords:
Bulk metallic glasses
Selective laser melting (SLM)
Soft magnetic properties
Microstructure

ABSTRACT

Soft magnetic Fe-based bulk metallic glass cylindrical specimens with a diameter of 2 mm and height of 6 mm have been successfully fabricated by selective laser melting (SLM) and the effect of scan speed *v* and laser power *P* on the microstructure, thermal stability and soft magnetic properties has been investigated. The results indicate that low *v* and high *P* lead to the formation of SLM samples with high relative densities, which can reach values of about 99.7%. This can be ascribed to the optimal energy transfer during processing at low *v* and high *P*. Structural and calorimetric studies prove that the SLM samples are fully amorphous. In addition, magnetic measurements reveal that the amorphous structure of the SLM material is identical to the parent atomized powders. Although additional work is required to remove the residual porosity and to avoid the formation of cracks during processing, the present results confirm that additive manufacturing by SLM represents an alternative processing route for the preparation of bulk metallic glass components with designed geometry having excellent magnetic softness.

1. Introduction

Fe- and Co-based metallic glasses have attracted significant attention as promising soft magnetic materials having high-energy efficiency and performance [1–3]. With regard to this, the fabrication of 3-dimensional magnetic components has been considered as one of the important issues for extending the industrial applications of these materials.

During the last two decades, there has been increasing understanding of alloying design with the aim of improving the glass forming ability (GFA) of soft magnetic amorphous alloys [4,5]. As a result, several new bulk metallic glass (BMG) systems with large GFA have been reported [6–11]. Because of their enhanced GFA, casting techniques such as water-cooled suction casting and injection casting can be used for the fabrication of soft magnetic bulk amorphous components. For example, Wu et al. [12] and Zhang et al. [13] reported on the direct fabrication of toroidal cores with good magnetic properties. The bulk glassy core of $Fe_{66}Co_{10}Mo_{3.5}P_{10}C_4B_4Si_{2.5}$ BMG having an inner diameter of 6 mm and an outer diameter of 10 mm exhibits a low coercive force of 1.0 A/m, a high maximum permeability of 450,000 as well as a low core loss of 0.4 W/kg at 50 Hz and a maximum magnetic flux density of 1 T [13]. However, owing to dimensional limitations and brittleness, there have been a few reports on the direct fabrication of bulk magnetic cores using casting process.

As an alternative, powder metallurgy (P/M) has been widely used to synthesize bulk amorphous components. Because of the high mechanical strength and lack of plasticity, the compaction of amorphous powders at low temperatures requires the use of binders [14], which are not necessary for compaction at elevated temperature within the supercooled liquid region, where metallic glasses display a significant decrease of viscosity and a viscous flow behavior [15–17]. Major processes for the P/M fabrication of 3-dimensional amorphous components are hot pressing and spark plasma sintering. To minimize the possible deterioration of the magnetic softness resulting from partial crystallization of the glassy precursor, the consolidation behavior as well as the thermal stability of the amorphous structure at elevated temperatures should be carefully controlled. This drawback can be overcome through the production of BMGs by additive manufacturing, as demonstrated by Pauly et al., who reported on the fabrication of Fe-based BMG by selective laser melting (SLM) [18]. SLM is a layer-based additive manufacturing technology highly advantageous for the fabrication of bulk metallic

* Corresponding author.
E-mail address: hyoyun.jung@gmail.com (H.Y. Jung).

http://dx.doi.org/10.1016/j.matdes.2015.07.145

components with refined microstructures and remarkable mechanical properties [19–22]. By using an optimum laser energy density, metal powders are melted, rapidly solidified and built in a 3-dimensional structure. Although the SLM specimens prepared in the previous study [18] were partially crystalline and displayed a high density of process-induced cracks, this study nevertheless provided a valuable insight for the fabrication of bulk amorphous/nanocrystallized components by SLM.

In contrast to the compaction of amorphous powders, SLM involves melting and rapid solidification of thin layers of metallic powders. Although the imposed cooling rate during SLM depends on the processing parameters used [23], results suggest that solidification during SLM occurs relatively rapidly with typical cooling rates in the range of 10^3–10^4 K/s [18,24]. Thus, the SLM process is a suitable alternative to fabricate bulk amorphous components. Recently, Li et al. reported on the fabrication of an Al-based metallic glass by SLM [25]. Although the processing window for the fabrication of a fully amorphous phase was relatively narrow, they successfully fabricated single line amorphous tracks avoiding crystallization. However, up to now, there has been no report of the synthesis of 3-dimensional fully glassy components using SLM.

In the present work, Fe-based BMG was fabricated by SLM. Because of its large GFA and good magnetic properties, an Fe–C–Si–B–P–Cr–Mo–Al BMG developed from industrial raw materials was selected for the present study. According to the previous report by Li et al. [26], the Fe-based BMG with composition $Fe_{68.3}C_{6.9}Si_{2.5}B_{6.7}P_{8.7}Cr_{2.3}Mo_{2.5}Al_{2.1}$ (at.%) can be prepared as amorphous rods with a maximum diameter of 4 mm by water-cooled suction casting. Gas atomization was used to prepare glass-formable powders for the present study. Along with the parameter studies, the effect of the SLM process on the microstructure, thermal properties and magnetic properties of the Fe-based BMG was investigated. In order to evaluate the properties of the materials, the BMG samples produced by SLM were compared with standard samples and in this case the samples produced by casting process.

2. Experimental procedures

A master alloy with nominal composition $Fe_{68.3}C_{6.9}Si_{2.5}B_{6.7}P_{8.7}Cr_{2.3}Mo_{2.5}Al_{2.1}$ (at.%) was prepared under N_2 atmosphere by induction melting of hot metal, industrial ferroalloys (Fe–75Si, Fe–25P, Fe–60Cr, Fe–60Mo, Fe–15B) and pure elements (Al, Fe) having commercial grade purity (>99%). Cylindrical specimens with a length of 55 mm and a diameter of 3 mm and 4 mm were fabricated by suction casting. Glass forming powders were prepared by gas atomization. Under vacuum condition (3×10^{-2} Torr), the master alloy was heated up to the operating temperature ($T_m + 200$ K), and the molten alloy was ejected through a nozzle with an Ar pressure of 1.2 MPa. The gas-atomized powders with particle size between 75 μm and 150 μm were selected for the present study because of their good flowability, which is a necessary prerequisite for optimal layer deposition during SLM. Cylindrical specimens with a diameter of 2 mm and height of 6 mm were fabricated on an Al substrate plate by using a SLM 250 HL device (SLM Solutions) equipped with a Yb-YAG laser at room temperature. Several parameter combinations with the laser power P ranging between 280 and 340 W and the scan speed v between 1500 and 4500 mm/s were employed with a layer thickness of 75 μm, a hatch spacing of 110 μm and a hatch style rotation of 90°. High purity argon gas was used throughout the SLM process to avoid oxygen contamination. The microstructure of the samples was analyzed by optical microscopy (OM) using an Olympus optical microscope. Phase analysis of the specimens was performed by X-ray diffraction (XRD) in reflection geometry using a laboratory D3290 PANalytical X'pert PRO diffractometer with Co radiation ($\lambda = 0.179$ nm) and in transmission at the ID11 beamline of the European Synchrotron Radiation Facilities (ESRF) using a high-intensity high-energy monochromatic synchrotron beam ($\lambda = 0.0206642$ nm). Two-dimensional powder XRD pattern was integrated to Q-space using the FIT2D [27], where Q is the wave vector. The thermal stability of the amorphous specimens was investigated by differential scanning calorimetry in constant-rate heating mode (40 K/min) with a Perkin-Elmer DSC7 calorimeter under a continuous flow of purified argon. Differential thermal analysis (DTA, Sinco, S-1600) was performed to investigate melting behavior of the specimens, revealing same melting temperature (T_m) of the alloy after SLM process. The T_m of the gas-atomized powders and SLM sample prepared with $v = 2500$ mm/s and $P = 340$ W was 1216 K and 1217 K, respectively. Hysteresis M–H loops of the specimens were measured using the VSM option of a Quantum Design physical property measurement system. The coercivity was measured with a Foerster Coercimat under an applied field high enough to maintain magnetic saturation. The density of the specimens was measured by the Archimedes method using distilled water as working liquid. All specimens for density measurement were tested ten times, and average density of each specimen was calculated. Relative density of the SLM samples was obtained by comparing the average density of the SLM specimens and suction cast rod with a diameter of 3 mm. Table 1 lists details of average density of the specimens.

3. Results

Fig. 1 compares the XRD patterns ($\lambda = 0.179$ nm) of the gas-atomized $Fe_{68.3}C_{6.9}Si_{2.5}B_{6.7}P_{8.7}Cr_{2.3}Mo_{2.5}Al_{2.1}$ powders with different sizes below 355 μm, and the corresponding suction cast BMG rods with a diameter of 3 and 4 mm. Except for the rod with a diameter of 4 mm, all specimens show only broad and diffuse halo patterns without any sharp diffraction peaks, indicating the amorphous nature of the specimens. On the other hand, the sample with a diameter of 4 mm shows sharp diffraction peaks of metastable Fe_3C, FeB, Fe_2P and α-Fe phases, suggesting that the maximum diameter for glass formation (d_{max}) of the present alloy is 3 mm.

Fig. 2 shows the relative density map of the SLM samples as a function of laser power (P) and scan speed (v). Relative density of the SLM samples was obtained by comparing the average density of the SLM specimens and suction cast rod with a diameter of 3 mm. In the case of the examined processing parameters with $v > 2500$ mm/s, the formation of bulk specimens was not possible. At such high scan speeds, the powder bed was not able to receive enough energy input during SLM processing, leading to incomplete melting and poor inter-particle bonding [28–30]. On the other hand, processing at lower speeds ($v \le 2500$ mm/s) leads to improved melting and consolidation of the powders, allowing the formation of rod-shaped specimens with various densification levels: lower v and higher P lead to the formation of SLM samples with high relative densities. When v decreases to 1500 mm/s and P increases above 300 W, the SLM parts show relative densities higher than 99%.

Fig. 3 presents the cross-sectional OM images of the parent gas-atomized powders and SLM samples prepared using different combinations of processing parameters. The OM image of the atomized powders (Fig. 3(a)) reveals that most particles are spherical, but some have an elongated shape and spherical micro-pores formed by trapped gases [31].

Table 1
Density of suction rod and SLM samples as a function of laser power (P) and scan speed (v).

		Scan speed, v [mm/s]	
		2500 mm/s	1500 mm/s
Laser power, P [W]	280 W	5.03 ± 0.26 g/cm^3	5.86 ± 0.04 g/cm^3
	300 W	5.18 ± 0.19 g/cm^3	7.12 ± 0.01 g/cm^3
	320 W	5.53 ± 0.15 g/cm^3	7.13 ± 0.01 g/cm^3
	340 W	6.01 ± 0.03 g/cm^3	7.16 ± 0.01 g/cm^3
Suction rod (ϕ = 3 mm)		7.18 ± 0.01 g/cm^3	