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Preparation of Bulk Glassy $Fe_{76}Si_9B_{10}P_5$ as a Soft Magnetic Material by Spark Plasma Sintering

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Preparation of a soft magnetic $Fe_{76}Si_9B_{10}P_5$ glassy bulk material has been carried out by the spark plasma sintering (SPS) technique below the glass transition temperature. The glassy powders were consolidated into bulk forms with relative densities above 98.7% through sintering them at 740 K under a pressure of 600 MPa while the samples still keep a glassy state. These as-sintered samples with a diameter of 15 mm exhibited excellent soft magnetic characteristics, which is as good as that of the cast samples with a size of 2.5 mm.
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1. Introduction

Bulk glassy alloys (BGAs) are promising materials for structural application as they exhibit high mechanical strength, high hardness, good fracture toughness, superior corrosion resistance, and so on.[1,2] BGAs with excellent glasses forming ability were developed in the 1990s by conventional casting methods[2] where Fe- and Co-based glassy alloys were found to have good soft magnetism at room temperature.[3,4] Recent work reports that the $Fe_{76}Si_9B_{10}P_5$ bulk metallic glass has an unusual combination of high B_s of 1.51 T due to high Fe content and high glass forming ability forming a glassy rod with a diameter of 2.5 mm without any glass-forming metal elements[5] where B_s denotes the full magnetic induction.

The BGAs are usually low-dimensional, such as thin films, sheets, powders, wires and rods due to the necessity of high cooling rates, which limit the possible applications. Thus, the powders metallurgy technique is utilized to form bulk samples,[6] which however need extensive equipments. An alternative is SPS (spark plasma sintering) technique[7] where lower temperature and shorter time can be realized for the sintering. The technique thus can avoid any crystallization of BGAs and realized sintering for larger size samples.[8,9] Therefore, this technique has be applied for sintering of several Fe-base bulk glassy magnetic materials, such as Fe-Al-Ga-P-C-B-Si,[10] Fe-Co-Ga-P-C-B[11] and Fe–Co–Nd–Dy–B.[12]

In this contribution, $Fe_{76}Si_9B_{10}P_5$ glassy bulk samples are sintered using SPS technique. It is found the sample can be as large as 15 mm while the corresponding magnetic properties remain. Thus, the technique is beneficial for possible applications.

2. Experimental Procedure

Master ingots of the $Fe_{76}Si_9B_{10}P_5$ were prepared by induction melting the mixture of pure metals of Fe (99.98 mass%), pre-melted Fe-P (99.9 mass%), and pure metalloid of crystal B (99.5 mass%) and Si (99.999 mass%) in an argon atmosphere. The $Fe_{76}Si_9B_{10}P_5$ glassy powders were produced by a high pressure argon gas atomization method where the atomic compositions denote that of adding percentages. The ingots were remelt at 1473 K under atmosphere condition in a quartz tube using an induction heating coil, followed by injection through a nozzle with a diameter of 0.8 mm, and then atomized by high pressure argon gas with a dynamic pressure of about 9.3 MPa. The powders were classified and characterized by X-ray diffractometry (XRD), scanning electron microscopy (SEM), and differential scanning calorimetry (DSC). The size of the used powders for the sintering experiment is smaller than 63 µm. The glassy powders were pre-compacted, and then sintered in a vacuum using a SPS-1050 system. The sintering temperatures (T_s) selected below the glass transition temperature (T_g) are 680, 720, 740, 760 K. The heating rate is 100 K/min from room temperature to T_s with the holding time of 6 min. A uniaxial pressuring method was conducted using top and bottom WC hard metal punches. The loading pressure used was 600 MPa. The sintered samples obtained have a cylindrical shape with a diameter of 15 mm and a height of about 3 mm. The density of the sintered samples was determined by the Archimedean method using tetrabromoethane. The structures were examined by XRD. The thermal stability was determined by DSC at a heating rate of 0.67 K/s. Microstructures of the sintered samples were observed with a SEM. The B_s was measured under an applied field of 800 kA/m with a vibrating sample magnetometer. The coactivity (H_c) of the magnetic core, which was abraded after cut from the sintered disc by an electrical discharge machine, was measured with a DC B-H loop tracer in the applied field of 1.6 kA/m.

3. Results and Discussions

Figure 1 shows the surface morphology of the powders gas-atomized with a smaller size than 63 µm. No appreciable contrast revealing the formation of a crystalline phase is observed on the surface of any particles.

The DSC curve of the compact sintered at $T_s = 720$ K is shown in Fig. 2, together with the result of the original $Fe_{76}Si_9B_{10}P_5$ glassy powders. Typical endothermic reaction due to glass transition and two exothermic peaks due to crystallization are observed in the DSC curve of the glassy powders, which denote a Curie transition peak, a glass

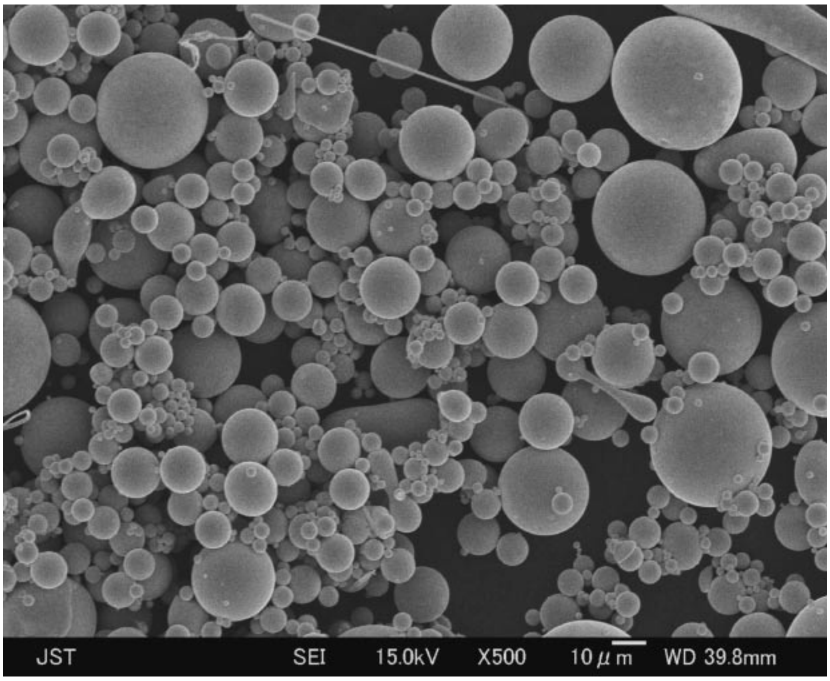

Fig. 1 Scanning electron micrograph of gas-atomized $Fe_{76}Si_9B_{10}P_5$ powders with particle size below 63 μm.

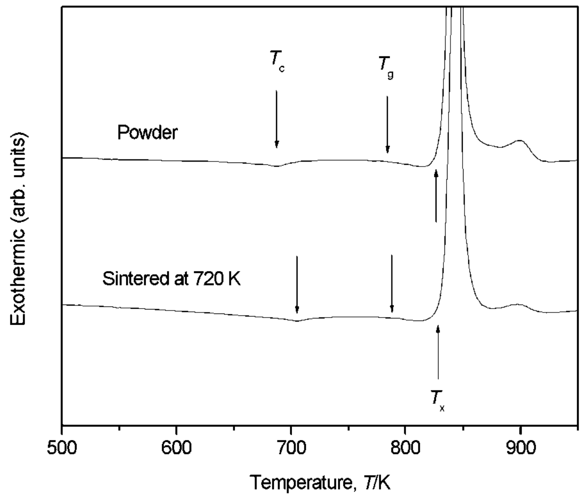

Fig. 2 DSC curves of the bulk $Fe_{76}Si_9B_{10}P_5$ alloy sintered at 720 K. The DSC curve of the powders is also shown for comparison.

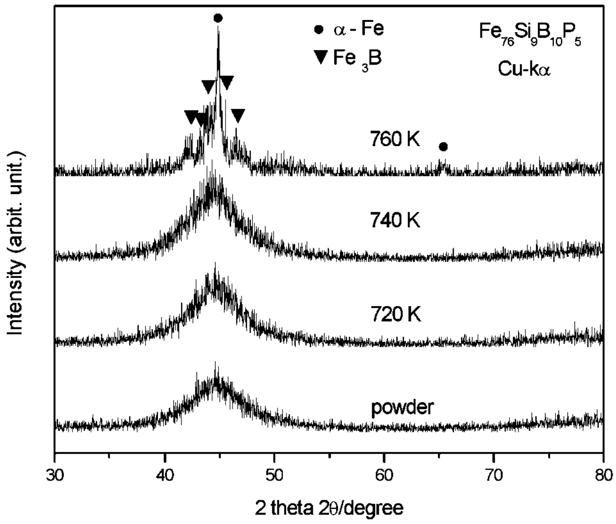

Fig. 3 XRD patterns of the bulk $Fe_{76}Si_9B_{10}P_5$ alloy sintered at various temperatures. The XRD pattern of the powders is also shown for comparison.

transition peak and two crystallization peaks. T_g and the crystallization temperature (T_x) of the $Fe_{76}Si_9B_{10}P_5$ powders are 780 K and 832 K, respectively. The largest ΔT_x is 52 K for $Fe_{76}Si_9B_{10}P_5$ powders where $\Delta T_x = T_x - T_g$, which is in agreement with that of the casting samples with a diameter (D_{cr}) of 2.5 mm[5] where T_g and T_x are almost the same. The endothermic and exothermic reactions are also observed in the DSC curve of the sample sintered at 720 K although the intensity of endothermic peak of the sintered samples is slightly lower than that of the powders. Curie temperature of the sintered sample also shifts to higher temperature.

Figure 3 shows the XRD patterns of these as-sintered compacts together with the pattern of the glassy powders for comparison. The diffraction patterns of the samples sintered at the of 720 K and 740 K consist of a halo pattern while no detectable diffraction peaks of crystalline phases is found, being in agreement with that of the original glassy powders. However, when $T_s = 760$ K, crystallization is present with the formation of Fe_3B and α-Fe phases identified by the XRD patterns.

Figure 4 shows the relative densities of the sintered samples at different T_s. The relative densities are expressed in percentage against the glassy ribbon density. The relative density of the compacts increases with increasing T_s. The relative density is 98.5% at $T_s = 720$ K, 98.7% at $T_s = 740$ K and reached 99.5% at $T_s = 760$ K which is just below T_g. Since the hot-pressing technique needs a much higher temperature, SPS technique should be the unique one to compact powers at $T_s < T_g$ while in our case $T_s = 740\,K < T_g = 780$ K. It is reported that the imposition

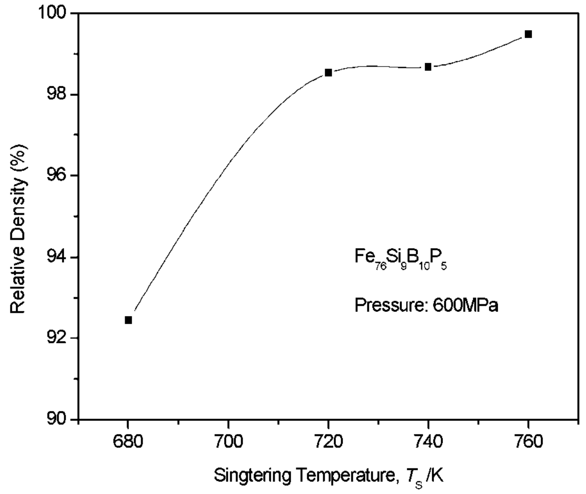

Fig. 4 Changes in relative density for the bulk $Fe_{76}Si_9B_{10}P_5$ alloy as a function of sintering temperature.

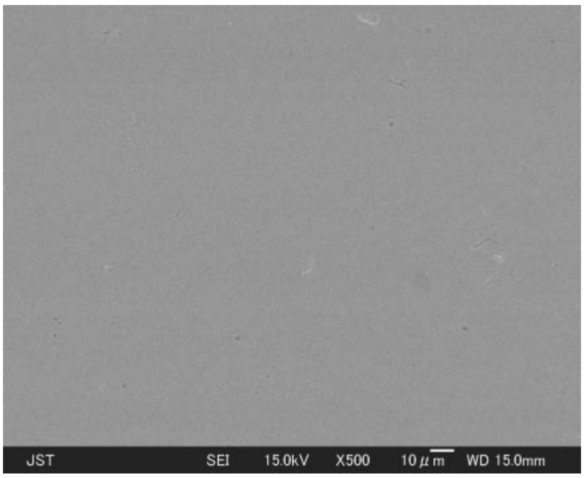

Fig. 5 Scanning electron micrograph of polished surface for the bulk $Fe_{76}Si_9B_{10}P_5$ alloy sintered at 720 K.

of the pressure of 500 MPa reduces the glass transition temperature by 40 K.[13] We used in the present study the pressure of 600 MPa. Although this value is slightly larger than Ref. 13), we consider that the highest value of T_s was within observed T_g.

Figure 5 shows a micrograph of polished $Fe_{76}Si_9B_{10}P_5$ sample sintered at 720 K. Only a few small pores are observed and raw powders are consolidated precisely. The increase of the relative density of in sintered $Fe_{76}Si_9B_{10}P_5$ alloy is due to the pressing of undercooled liquid at the sample surface into the pore zones due to plunging the powder with the surface in supercooled liquid state into the pores zones[14,15] which was caused by the rise of temperature during SPS operation.[16]

Figure 6 shows dc *B-H* hysteresis loop curve of the bulk $Fe_{76}Si_9B_{10}P_5$ alloy sintered at 720 K. The samples sintered at 720 K show high flux density with soft magnetic characteristics. The flux density under applied field of 800 A/m (B_{800}), maximum permeability (μ_{max}) and coercive force (H_c) are 1.45 T, 3471 and 17.6 A/m, respectively, being correspondent to the as-atomized $Fe_{76}Si_9B_{10}P_5$ alloy powders. These

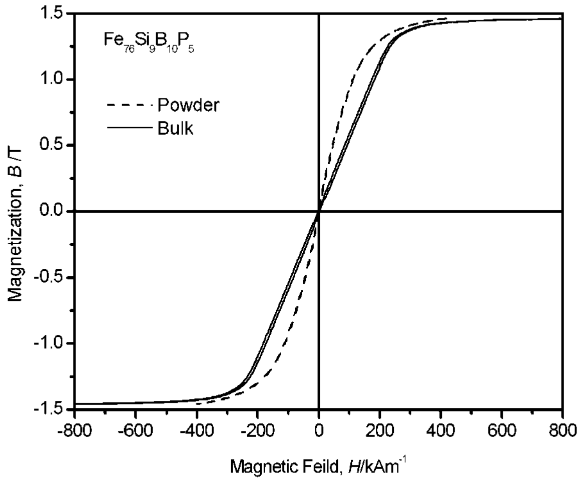

Fig. 6 The dc *B-H* curve of the bulk $Fe_{76}Si_9B_{10}P_5$ alloy sintered at 720 K. The data of the powders is also shown for comparison.

good soft magnetic properties should be related to the high density of our alloys.

4. Conclusions

We have fabricated bulk $Fe_{76}Si_9B_{10}P_5$ glassy alloys with a diameter of 15 mm using SPS technique just below T_g with excellent soft magnetic properties. Under the conditions of pressure of 600 MPa, holding time of 6 min and $T_s = 740$ K, the relative densities reach 98.7% while the glassy state remains. B_{800}, μ_{max} and H_c with $T_s = 720$ K are 1.45 T, 3471 and 17.6 A/m, respectively.

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