Microwave-induced sintering of NiNbTiPt metallic glass blended with Sn powders using a single-mode applicator

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Abstract. Microwave (MW) processing is emerging as an innovative and highly effective material processing method offering many advantages over conventional methods especially for sintering application. In the present study, using a mixed powder of the Ni55Nb25Ti15Pt5 metallic glassy powder prepared by an argon gas atomization process blended with 50 vol.% Sn powder, the MW-induced heating and sintering were carried out by a single-mode 2.45 GHz MW applicator in a magnetic field maximum. The structure and thermal stability of the sintered glassy composite specimens were investigated. The addition of Sn particles enhanced densification of the sintered Ni-based metallic glassy alloy powders.

1. Introduction
Microwave (MW)-induced heating and sintering processes have attracted increasing attention due to its significant advantages in material processing compared to conventional powder sintering processes. It is recognized for its various advantages, such as: time and energy saving, rapid heating rates, considerably reduced processing cycle time and temperature, fine microstructures and improved mechanical properties leading to better product performance [1]. However, until recently, MW processing was mostly restricted to ceramics, cemented carbides and ferrites. Applicability of MW sintering to metals was ignored due to the fact that they reflect MWs. Roy et al. [2] reported that powdered metals can be heated rapidly in MW, which has led to the use of MW to consolidate various powdered metals and alloys [3-5].

Ni-based bulk metallic glasses (BMGs) exhibit high thermal stability, ultra-high strength and excellent corrosion resistance [6]. The combination of superior properties and low material cost enhances Ni-based BMGs to have promising applications as engineering materials. Recently, BMG rods of Ni-based alloys up to 2~5 mm in diameter have been produced using a Cu mold casting technique in various Ni-based metal-metal alloy systems [7-10]. However, the critical size of Ni-based BMGs obtained is much smaller than those of Zr-, Pd-, and Cu-based BMGs with critical sizes of over 20 mm. This limits the extensive practical applications of Ni-based BMGs. In order to overcome the limitations, development of appropriate techniques is an important research subject.
Recently, we attempted to sinter Ni$_{52.5}$Nb$_{10}$Zr$_{15}$Ti$_{15}$Pt$_{7.5}$ metallic glass alloy powder and its composite powders by the MW-induced heating process [11,12]. It was shown that the heating was better carried out in magnetic (H-) field. The sintered compacts retaining the glassy structure were achieved. However, powder particles were very weakly bonded and quite a low relative density was obtained. In the present study, we intend to investigate the sintering behavior of the gas atomized Ni$_{55}$Nb$_{25}$Ti$_{15}$Pt$_{5}$ metallic glassy alloy powder blended with Sn powder in order to increase relative density of the sintered samples by transient liquid phase because Sn has low melting point (505 K).

2. Experimental procedures
Master ingots of the Ni$_{55}$Nb$_{25}$Ti$_{15}$Pt$_{5}$ (composition is given in nominal at.%) alloy were prepared by arc melting a mixture of high purity Ni, Nb, Ti and Pt in an argon atmosphere purified using Ti getter. The Ni$_{55}$Nb$_{25}$Ti$_{15}$Pt$_{5}$ metallic glassy alloy powders were produced by a high pressure argon gas atomization method. The details of the processing procedure and characterization of the prepared powders have been given elsewhere [13]. The glassy alloy powders with the size below 63 μm were used in this study. Then a designated proportion of the glassy alloy powder and Sn powder was uniformly blended in a mixer. The Sn powders having a size below 37 μm and a purity of 99.9% were used.

The sintering of the mixed powders was carried out using a single-mode MW applicator (Nikoha MKN-152-359, 2.45 GHz, maximum power 1.5kW, Yokohama, Japan) in separated magnetic (H-) field or electric (E-) field. Figure 1 shows the schematic illustration of this MW apparatus [4]. The powder specimens were placed in a holder made of silica glass with the dimensions of 5 mm in diameter and 7 mm in height. The sample holder was set in a position of either H-field or E-field maximum area in the wave guide applicator. Heating was carried out at a constant input power (P$_{i}$) with tuning the power of the reflected wave (P$_{r}$) to be minimized by manipulating the three-stubs, manually. Movable plunger is set at the end of the wave-guide, which acts as a metal wall and the wave is reflected. The holding time at the sintering temperature was 600 s. The temperature measurement was performed in-situ by an optical pyrometer (PhotoriX system, Luxtron, Santa Clara, CA, USA) using a sapphire rod on a light guide, and thus the temperature reading above 653 K was possible. The temperature was calibrated comparing with the thermocouple measurement. All experiments were carried out in a flowing nitrogen gas to avoid oxidation of metallic samples at high temperature.

The density of the sintered specimens was determined by measuring their mass and dimension of the samples (mass per volume). The structures of the powders and the sintered specimens were examined by X-ray diffractometry (XRD) in reflection with a monochromatic Cu Kα radiation, and the microstructure was observed with a scanning electron microscope (SEM). Thermal stability associated with the glass transition temperature (T$_{g}$), crystallization temperature (T$_{X}$), and supercooled liquid region (ΔT=T$_{X}$-T$_{g}$) was examined by differential scanning calorimetry (DSC) at a heating rate of 0.67 K s$^{-1}$.

3. Results and discussion
The Ni$_{55}$Nb$_{25}$Ti$_{15}$Pt$_{5}$ alloy powders were prepared by the argon gas atomization method. The as-prepared powders were classified. The particles of the gas-atomized powders in size below 63 μm had a fully glassy structure. The SEM observation indicated that, irrespective of size, spherical morphology as well as clean surfaces is seen in all sizes of as-prepared alloy powders. Figure 2 presents a SEM micrograph of the as-prepared Ni$_{55}$Nb$_{25}$Ti$_{15}$Pt$_{5}$ powder with a particle size below 63 μm.
μm. No appreciable contrast revealing the formation of a crystalline phase is observed on the outer surface of the particles. Figure 3 shows a typical XRD pattern of the as-atomized powders. Only a diffuse diffraction pattern typical for a glassy phase is seen. Neither diffraction peaks corresponding to crystalline phases are observed, nor any apparent effect of powder size on the glassy phase stability is observed. This indicates that a single glassy phase is formed.

Using the prepared Ni₅₅Nb₂₅Ti₁₅Pt₅ glassy powder or its mixed powders blended with 50 vol.% Sn powders, the sintering was performed by the MW-induced heating using a single-mode 2.45 GHz MW applicator in a separated H-field. Figure 4 shows the XRD patterns obtained from the sintered bulk composite samples at various sintering temperatures as well as that of the original mixed powder for the Ni₅₅Nb₂₅Ti₁₅Pt₅ glassy+50 vol.% Sn powders. It is observed that the sintered bulk samples can be obtained at a low sintering temperature of 673 K. When the sintering was carried out at the temperature below 723 K, the XRD patterns of the sintered specimens were similar to that of the original mixed powder, indicating that the specimens sintered below 723 K mainly were composed of a glassy phase, except for the additional crystalline Sn phase. Comparing to 843 K for the monolithic Ni-based bulk sample with full glassy structure in H-field [11,12], the temperature for obtaining sintered bulk samples was lowered. The DSC analyses of the sintered specimens obtained at various sintering temperatures also exhibited the similar results.

The microstructure of the sintered bulk composite samples was characterized by SEM. Figure 5 shows SEM micrographs of the transverse cross section of the sintered bulk composite sample. The good bonding state among powder particles was obtained. The relative density of the sintered bulk composite samples was 52.6%, 53.3% and 56.2% at the sintering temperatures of 673 K, 703 K, and 723 K, respectively. The density is higher than that of the monolithic Ni-based bulk sample in 843 K, which is 42.0% [12]. This is because the addition of Sn particles can lead to liquid phase sintering. It is hypothesized that time for diffusion of Sn into Ni-based metallic glass is rather short upon rapid heating in the MW process. Therefore, most of the diffusion might have happened at the sintering temperature. As metallic glasses exhibit low viscosity in the supercooled liquid region, MW sintering causes quick diffusion kinetics of Sn in Ni-based metallic glass. Sn has a very low melting point and
the viscosity of the liquid metal has an Arrhenius dependence on temperature and thus decreases exponentially with temperature above its melting point [14]. Its viscosity at the sintering temperature is low, and thus liquid Sn can penetrate the fine voids and interstices between the metallic glassy particles due to the capillary action, leading to the promotion of shrinkage and densification.

4. Conclusions
We have fabricated Ni55Nb25Ti15Pt5 metallic glassy alloy powder by an argon gas atomization process. Using the prepared Ni55Nb25Ti15Pt5 glassy powder or its mixed powders blended with 50 vol.% Sn powder, the MW-induced heating and sintering were carried out by a single-mode 2.45 GHz MW applicator in a H-field maximum. The structure and thermal stability of the sintered glassy composite specimens were investigated. The addition of Sn particles enhanced densification of the sintered Ni-based metallic glassy alloy powders.

Acknowledgements
The authors sincerely thank H. Kimura and T. Kanomata from Institute for Materials Research for their kind assistance in powder preparation. The work was supported by a Grant-In-Aid for Science Research in a Priority Area on “Science and Technology of Microwave-Induced, thermally Non-Equilibrium Reaction” (No.18070001), and by Grant-In-Aid for Scientific Research (C) (No. 20560639), as well as by Grant-In-Aid on “Research and Development Project on Advanced Metallic Glasses, Inorganic Materials, and Joining Technology” from the Ministry of Education, Sports, Culture, Science and Technology, Japan. A part of this work was carried out at Advanced Research Center of Metallic Glasses, Institute for Materials Research, Tohoku University, Japan.

References
[1] D. Agrawal, Topical Reviews, 2006, 65(3), 129.
[2] R. Roy, D. Agrawal, J.P. Cheng, S. Gedevanishvili, Nature, 1999, 399, 668.
[3] M. Gupta, W.L.E. Wong, Scripta Mater., 2005, 52, 479.
[4] N. Yoshikawa, E. Ishizuka, S. Taniguchi, Mater. Trans., 2006, 47, 898.
[5] A. Upadhyaya, S.K. Tiwari, P. Mishra, Scripta Mater., 2007, 56, 5.
[6] A. Inoue, Acta Mater., 2000, 48, 279.
[7] W. Zhang, A. Inoue, Mater. Trans., 2002, 43, 2342.
[8] T.C. Hufnagel, C. Fan, R.T. Ott, J. Li, S. Brennan, Intermetallics, 2002, 10, 1163.
[9] D.V. Louzguine-Luzgin, T. Shimada, A. Inoue, Intermetallics, 2005, 13, 1166.
[10] L.Y. Chen, H.T. Hu, G.Q. Zhang, J. Z. Jiang, J. Alloys Compd., 2007, 443, 109.
[11] N. Yoshikawa, D.V. Louzguine-Luzgin, K. Mashiko, G.Q. Xie, M. Sato, A. Inoue, S. Taniguchi, Mater. Trans., 2007, 48, 632.
[12] G.Q. Xie, S. Li, D.V. Louzguine-Luzgin, Z.P. Cao, N. Yoshikawa, M. Sato, A. Inoue, Intermetallics, 2008, 16, in press.
[13] G.Q. Xie, D.V. Louzguine-Luzgin, H. Kimura, A. Inoue, Appl. Phys. Lett., 2007, 90, 241902.
[14] W.D. Kingery, M.D. Narasimhan, J. Appl. Phys., 1959, 30, 307.