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Effect of sintering profile on densification of nano-sized Ni/Al_2O_3 composite

Dong-Guk Cho, Seung-Kyu Yang, Joon-Chul Yun, Hyung-Sub Kim, Jai-Sung Lee, Caroline Sunyong Lee *

Department of Materials Engineering, Hanyang University, Ansan, Gyeonggi 426-791, Republic of Korea

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ABSTRACT

The effect of sintering profiles on densification of nanosized Ni/Al_2O_3 composite was investigated. Ni powder (100-nm average grain diameter) was combined with Al_2O_3 powder in a ratio of 60:40 by weight (40.1:59.9 by volume) and sintered for 3 h at 1350 °C. To observe the variation in sintering behavior of the Ni powder in the composite, two different H_2 reduction profiles were used: H_2 gas added at 700 °C for 2 h (Profile #1) during sintering and H_2 gas added from room temperature to 1000 °C with no holding time (Profile #2). The microstructural changes in the Ni/Al_2O_3 composite, which consisted mainly of Ni dispersed in an Al_2O_3 matrix, were observed for these two sintering profiles. For Profile #1, incomplete sintering of the Al_2O_3 matrix was observed due to the extensive growth of Ni dispersants, and the overall dispersion of nanosized Ni powder was poor but coarsened. In contrast, in the composite sintered using Profile #2, the overgrowth of Ni dispersants was reduced, the sintering of the Al_2O_3 matrix was much more complete, and the density was 10% greater. In addition, a large difference in the Vickers' hardness values between those two samples was measured. The significant difference in density differences for those two composites is possibly due to the rearrangement of Al_2O_3 powders caused by the overgrowth of Ni powders, and the increase in the non-contact area of the Al_2O_3 powders during the coarsening of Ni dispersion that resulted in a less dispersed and more porous sample. This illustrates that the sintering profile dramatically controls the density and the hardness of composites formed from nanosized metal powders.

1. Introduction

Various kinds of metal–ceramic composite materials are used in high-temperature applications such as automobiles, aerospace vehicles, and fuel cells. These materials must be able to withstand high temperatures, and thus their thermal resistance, chemical durability, abrasion resistance, and heat resistance must be quite high. However, ceramics are formed through covalent and ionic bonding among atoms, which can result in low resistance to fracturing and low plastic deformation. Several studies have investigated methods of overcoming these drawbacks and improving the fracture resistance using particle-dispersion toughening [1–4]. Al_2O_3 is widely used as a ceramic material because of its low cost, high strength, and relatively large coefficient of thermal expansion (CTE) that makes it suitable for use with metals. Various types of dispersant powder such as Ni, Fe, Al, Ag, Cu, W, Mo, Pt, and Nb have been used with Al_2O_3 to improve its ductility [5–13]. Ni is a good choice for metal–ceramic composite fabrication because it is highly ductile, thermally stable at high temperature, and corrosion resistant.

Metal–ceramic composites generally contain a low percentage of metal by weight; ceramics with high proportions of metal have not been widely reported. In cases where the metal content is high, the composite is usually used as a functionally graded material (FGM) or as a joining material between dissimilar materials [14]. However, such FGM or joining materials with a high percentage of metal have problems such as a tendency to fracture and poor dispersion due to density differences, and these characteristics pose serious problems for the fabrication and use of these materials [15]. Previous studies showed that the weakest point of Ni/Al_2O_3 FGM occurs at a weight ratio of 60:40 Ni/Al_2O_3 [16], which is accompanied by a significant amount of porosity. To overcome these problems, Ni/Al_2O_3 composite for the application of FGM was studied in this study in order to solve poor density and agglomeration problems, occurring in the composite with high metal-contents.

Conventionally, HIP (Hot Isostatic Pressing) and SPS (Spark Plasma Sintering) have been used to obtain samples with high density. However, many industries and research facilities do not have these equipments due to its cost and complex sintering processes requiring many controls and technology. Therefore, the pressureless sintering furnace was used in this study to fabricate composite with high density by controlling porosity during diffusion of Ni particles, which is the main factor influencing its density. The formation of

* Corresponding author. Tel.: +82 31 400 5221; fax: +82 31 417 3701.
E-mail address: Sunyonglee@hanyang.ac.kr (C.S. Lee).

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porosity during diffusion of materials in a composite becomes apparent as the size of particles becomes nanosized as this is a common problem in sintering process. During sintering of composite materials, porosity formation due to diffusion of metal particles was observed in fabricating composite materials between graphene and gold [20].

Therefore, diffusional growth of particles during fabrication of composite materials is an important factor to consider to reduce formation of porosities. This problem of porosity formation caused by diffusion of metal particles can be resolved by simultaneous sintering among dissimilar materials by adjusting sintering profiles and particle size.

A large difference in the CTE between two materials can lead to poor density and increased porosity. Moreover, the difference in density between dissimilar materials results in non-uniform dispersion between the matrix and dispersants. Nanosized Ni powders were used in this study because nanosized powder improves densification due to large surface area. However, because nanosized Ni powder is easily oxidized, H_2 gas was added to create a reducing environment. This study examined the relationship between the sintering profiles and the densification of 60:40 Ni/Al_2O_3 composites produced with different H_2 gas temperature profiles.

2. Experimental procedures

Ni powders with its size of 100 nm in diameter, were obtained by ball milling 1-μm sized NiO powders in the bead-mill (Nanointech, Korea) followed by reduction in sintering. During ball milling process, NiO powders, methyl alcohol, and ZrO_2 balls with its size of 0.65 mm in diameter, were mixed vigorously at 2400 rpm in the bead-mill for 10 h to obtain nanosized NiO powders. These nanosized NiO powders was reduced in a H_2 atmosphere at 350 °C for 2 h to obtain Ni powders with an average diameter of 100 nm. These powders were then mixed with Al_2O_3 powders (0.16 μm, Tamicrom, TM-DAR) in ethanol in a ratio of 60:40 by weight. To break up this powder agglomeration, the powder mixture was ball-milled with methyl alcohol and Al_2O_3 balls with its size of 5 mm in diameter at 300 rpm in ball mill (Samwoo Scientific Co., Korea) for 4 h and then dried in an oven at 50 °C for 24 h. The mixture was then ground, sieved in a 325 mesh, placed in WC mold with its size of 12 mm in diameter, and uniaxially cold-pressed (CP) at 177, 354, and 531 MPa. These green bodies were pressed in a cold isostatic press at 100 MPa and finally sintered in a pressureless sintering furnace (CTF-A80A Hydrogen Atmosphere Tube Furnace, Cermotech Co., Ltd., Korea) using the two different profiles shown in Fig. 1.

The Vickers' hardness was tested at 200 gf using Vicker's indenter (DUROMETRI MICRO VICKERS, Affri system, Italy), and field-emission scanning electron microscopy (SEM; Hitachi-S4800, Hitachi Ltd., Japan) and field-emission transmission electron microscopy (TEM; JEM-2100F, Jeol Ltd., Japan), were used to observe the differences in microstructure and mechanical properties for the material produced using different sintering profiles. The densities were compared using Archimedes' method.

3. Results

3.1. Microstructural observation

Fig. 2 shows the fracture surface of the green body and a cross-sectional image of the sintered body to illustrate powder growth.

Fig. 2c shows the fractured surface of the green body for 60 wt.% Ni composite; the Ni powder is bright and irregularly shaped, whereas the Al_2O_3 powder is generally dark and round. The Ni and Al_2O_3 were evenly mixed and distributed. As shown in Fig. 2d, after sintering, the Ni powders clustered, and its grain size increased to greater than 1 μm, whereas the grain size of Al_2O_3 was approximately 0.2 μm, not increasing as much. The samples processed under both profiles were similar.

The TEM images in Fig. 3 show that Ni grains processed under sintering Profile #1 with 177 MPa of CP (Cold Press) pressure, increased to greater than 1 μm in diameter, whereas the Al_2O_3 grains did not grow significantly, but did exhibit necking among individual grains. The Ni grains remained relatively round.

The 3-h sintering process at 1350 °C thus caused the Al_2O_3 powder to experience limited grain growth including necking, whereas the Ni grew extensively, resulting in volume diffusion.

3.2. Microstructural change under different sintering profiles

The sintering profile and CP pressures were varied as described above to investigate the effect of controlling the microstructural changes in Ni/Al_2O_3 composites.

Fig. 4 shows the cross-sectional SEM images of the sintered body processed with different CP pressures and using different sintering profiles. The dark matrix is the Al_2O_3, and the bright spots are Ni grains. Overall, the Ni grains grew to a maximum size of 1–3 μm. The grains of Ni powders sintered under Profile #2 ranged from 0.1 to 1 μm, regardless of the CP pressure. Moreover, the growth of the Ni grains increased as the CP pressure increased. Thus, heating for 2 h at 700 °C in a H_2 atmosphere which is part of Profile #1 promoted the growth of Ni grains to 3 μm as shown in Fig. 4.

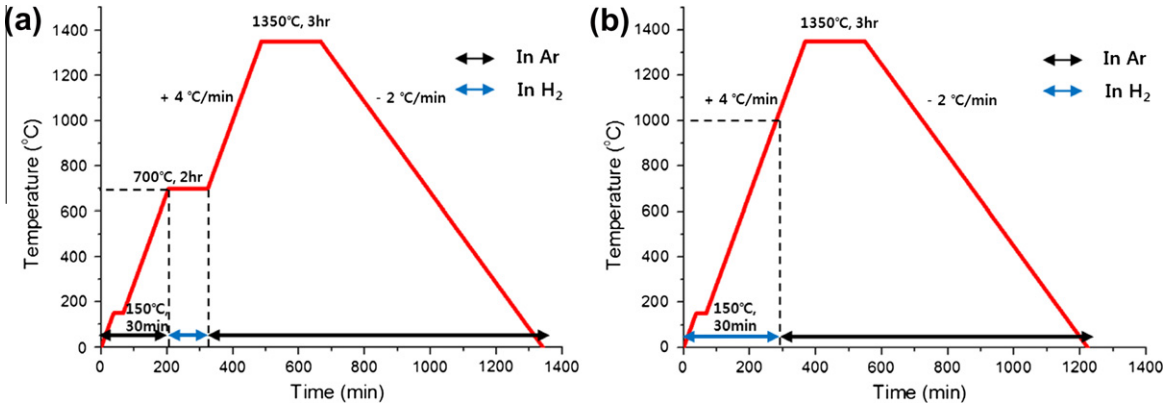

Fig. 1. Sintering conditions used to fabricate Ni–Al_2O_3 composites.