Materials Transactions, Vol. 42, No. 9 (2001) pp. 1846 to 1849

Effect of Interface Behavior between Particles on Properties of Pure Al Powder Compacts by Spark Plasma Sintering *1

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Pure Al powder was sintered by spark plasma sintering (SPS) process at various sintering temperatures and loading pressures. The density, electrical resistivity, tensile properties and microstructure of powder compacts were investigated. The powder compacts with the similar properties as base aluminum metal was obtained at sintering temperature above 873 K, loading pressure above 23.5 MPa. For the powder compacts with the similar density but with the large difference in the electrical resistivity and tensile properties, the interfaces between Al powder particles were investigated using high resolution transmission electron microscopy (HRTEM) and energy dispersive X-ray spectroscopy (EDS). Two types of interfaces, with metal/metal bonding and metal/oxide film layer/metal bonding, were observed in Al powder compacts. The properties of powder compacts were mainly subject to the behavior of oxide film between the powder particles.

(Received April 24, 2001; Accepted June 7, 2001)

Keywords: *spark plasma sintering, aluminum powder compact, electrical resistivity, tensile properties, interface, oxide film, high resolution transmission electron microscopy*

1. Introduction

Spark plasma sintering (SPS) has been drawing attention as a newly developed process in recent years because it possesses many advantages comparing with the other sintering methods such as hot press (HP) and hot isostatic press (HIP) processes.[1–3] In the SPS process, pulse electric current flows directly in the sintered materials and mold. A very high heating efficiency is offered. It can easily consolidate a high quality compact that is sintered at a lower sintering temperature and in a shorter time than the conventional processes, even to those materials that are very difficult to be sintered by the other processes.

Al powder particles are covered with aluminum oxide film, and the oxide film cannot be broken and/or removed by heat. It is difficult to be sintered by normal and hot press sintering processes,[4,5] but SPS process makes it easily. Recently, it has been reported that aluminum metal and its compounds powders[6–11] were sintered using SPS process. However, it is still unclear about the effects of SPS process conditions and oxide film behavior between powder particles on properties of powder compacts.

In the present study, pure Al powder was sintered by means of SPS process. The density, electrical resistivity, and tensile properties of the powder compacts were measured. Microstructure at the interfaces between Al powder particles in the compacts was observed using high resolution transmission electron microscopy (HRTEM). Effect of oxide film behavior between particles on properties of Al powder compacts was investigated.

2. Experimental Methods

Commercial pure Al powder (99.798 mass%) was used in this study. The particle sizes and distribution are as follows: below 44 μm, 42.5%; 44–62 μm, 28.1%; 62–74 μm, 14.0%; 74–104 μm, 15.4%. The sintering was carried out in a vacuum using SPS system (SPS-520 model; Izumi Technology Company, Ltd.). The heating model was 50 K/min (room temperature $\sim$ T_s–50 K), 12.5 K/min (T_s–50 K $\sim$ T_s). The holding time at sintering temperature was 5 min. The pulse frequency was 40 KHz. The uniaxial pressure model was conducted using the top and bottom graphite punches. The shape of the powder compacts obtained by SPS process was that of the tensile specimen, with length of 20 mm and width of 5 mm at the parallel part.

Density of the powder compacts was determined by measuring the weight and size. The electrical resistivity was obtained using four points probe method by measurement of the voltage between two probes when the certain direct current was flowed. Tensile test of the powder compacts was carried out by means of AG-250KNG autograph tester. The tensile velocity of 2 mm/min was used.

Microstructure at the interfaces between Al powder particles in the compacts was carried out using HRTEM. Thin foil specimens were cut out from the compacts by a diamond saw, ultrasonically cut into 3 mm diameter disks, mechanically thinned and dimpled and then ion milled to electron transparency for transmission electron microscopy (TEM) observations. Conventional and high resolution transmission electron microscopy (CTEM and HRTEM) experiments and energy dispersive X-ray spectroscopy (EDS) analysis were performed at room temperature using the JEM-ARM 1000 TEM equipped with an EDS system. The system was operated at 1000 KV for TEM observation and at 400 KV for EDS analysis.

*1 This Paper was Presented at the Spring Meeting of the Japan Institute of Metals, held in Narashino, on March 29, 2001.
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3. Results and Discussion

Pure Al powder compacts with tensile test plate were fabricated by SPS process with various sintering temperatures and loading pressures. The relative density, electrical resistivity and tensile properties of the compacts were investigated.

Figure 1 shows the results of the effect of sintering temperature and loading pressure on relative density of the Al powder compacts by SPS process, and Fig. 2 gives the results on electrical resistivity. Increasing in sintering temperature at the same loading pressure, or increasing in loading pressure at the same sintering temperature, the relative density of powder compacts increases and the electrical resistivity decreases. The relative density of powder compacts is above 97% and the electrical resistivity is almost same as that of base aluminum metal when the sintering temperature is above 823 K and the loading pressure is over 23.5 MPa.

Change of tensile strength of pure Al powder compacts with sintering temperature and loading pressure is shown in Fig. 3. The tensile strength of powder compacts increases when sintering temperature or loading pressure increases. At SPS process conditions with 823 K, 47.0 MPa, or with 873 K, 23.5 MPa, the tensile strength of powder compacts is consistent with that of base aluminum metal. However, for the powder compacts with the former SPS process condition, the elongation is much smaller than that of base aluminum metal, as shown in Fig. 4. The reduction of area is shown the same results as that of the elongation of the powder compacts.

From these investigated results, we can determine that pure Al powder compacts with the similar properties as base aluminum metal can be obtained by SPS process with sintering temperature above 873 K, loading pressure above 23.5 MPa.

Furthermore, effect of relative density on electrical resistivity and tensile strength of pure Al powder compacts were investigated. The results indicate that increasing in relative density of powder compacts at the same loading pressure, the electrical resistivity decreases and the tensile strength increases. It can also be seen that powder compacts at some sintering conditions have the similar relative density, namely, there is the similar contact area between powder particles, but the large difference in the electrical resistivity and in the tensile strength exist. For example, the powder compacts by SPS process conditions with 873 K, 9.4 MPa and with 623 K, 23.5 MPa have almost the same relative density, but the difference in electrical resistivity is about 100 times, and the difference in tensile strength is about 10 times.

In order to understand the cause of the difference in the electrical resistivity and in the tensile strength, pure Al powder compacts with 873 K, 9.4 MPa and with 623 K, 23.5 MPa were selected. Microstructure at bonding interface between Al powder particles in the compacts under two sintering conditions was investigated by means of HRTEM and EDS.

Figure 5 gives a set of TEM micrographs of pure Al powder compact by SPS process with 873 K, 9.4 MPa. Figure 5(a) shows a bright field (BF) TEM image of the compact, and Fig. 5(b) shows an HRTEM micrograph taken from the circle region in Fig. 5(a). The specimen was oriented such that the

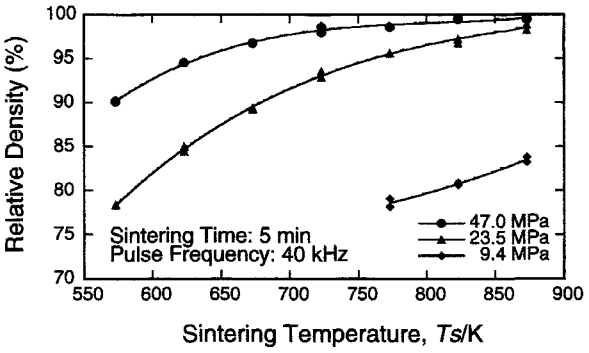

Fig. 1 Effect of sintering temperature and loading pressure on relative density of Al powder compacts by SPS process.

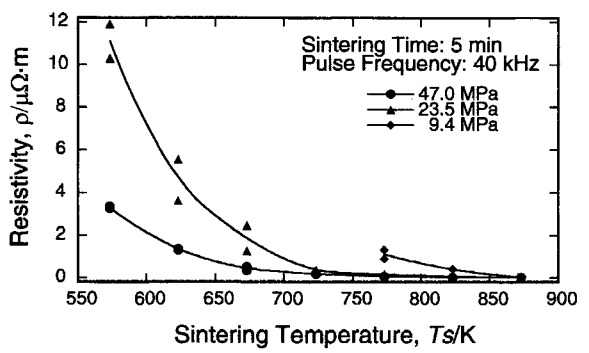

Fig. 2 Effect of sintering temperature and loading pressure on electrical resistivity of Al powder compacts by SPS process.

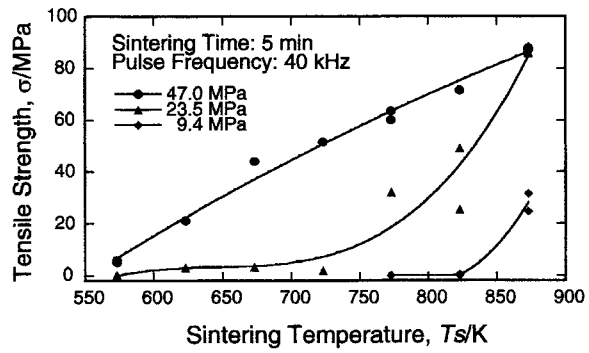

Fig. 3 Effect of sintering temperature and loading pressure on tensile strength of Al powder compacts by SPS process.

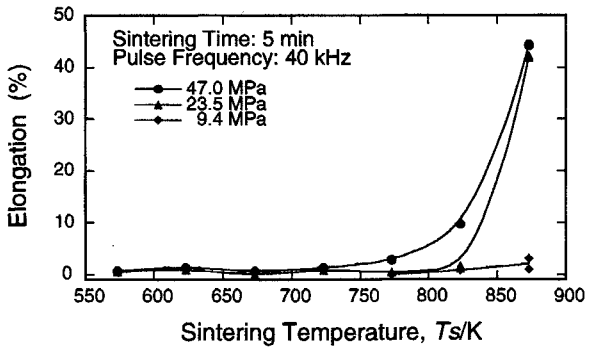

Fig. 4 Effect of sintering temperature and loading pressure on elongation of Al powder compacts by SPS process.

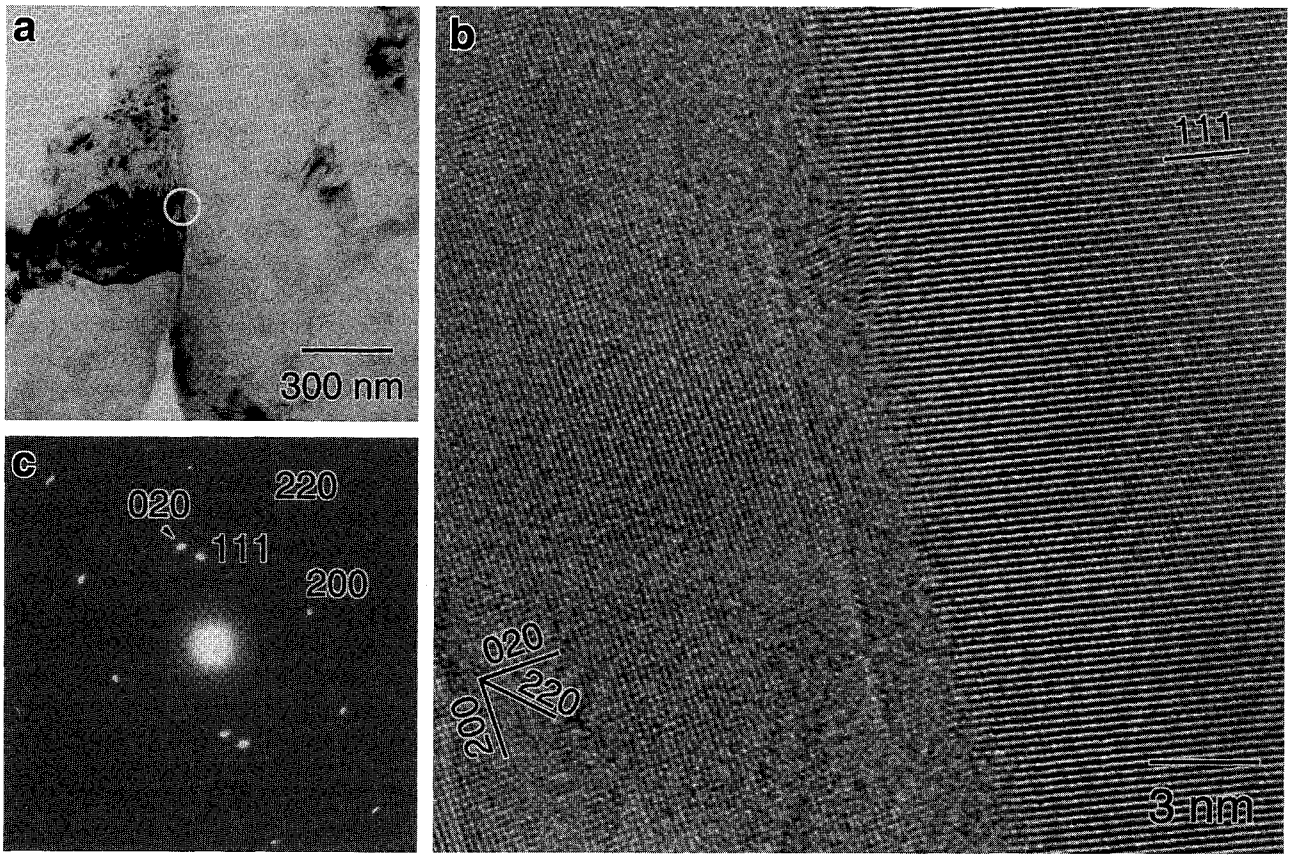

Fig. 5 TEM micrographs of Al powder compact using SPS process by 873 K, 9.4 MPa. (a) bright field TEM image; (b) an HRTEM micrograph of the typical metal/metal bonding taken from the region indicated by circle in (a); (c) the corresponding SAD pattern.

grain boundary plane was parallel to the electron beam direction. The boundary appears to be structurally clean and no evidence for oxide film layer or amorphous phases or the other impurity layers was observed. Direct bonding between two adjacent Al metal grains was seen. The left grain is in [001] zone-axis orientation and the right grain is in the (111) plane of Al fcc structure, as shown in the Fig. 5(b). Figure 5(c) gives the corresponding selected area diffraction (SAD) pattern. The results are consistent with those from HRTEM observation. Extra spot has not been observed expect for the diffraction spots from two Al metal particles. EDS analysis also indicates that there exists no oxide or the other impurity except for Al at the interface region between powder particles.

Figure 6 gives a set of TEM micrographs and EDS analyses results of powder compact by SPS process with 623 K, 23.5 MPa. Figure 6(a) shows a bright field (BF) TEM image, and Fig. 6(b) gives an HRTEM image taken from the circle region in Fig. 6(a). It can be seen that there are an amorphous layer and two Al metal grains in the image. The thickness of amorphous layer is about 10 nm. The left part is an Al grain of fcc structure in [111] direction, and the right part is in $(200)_{fcc}$ plane of Al structure, as shown in the image. EDS spectrum obtained from the interface region between powder particles indicates that there exists oxygen element, as shown in Fig. 6(c). But the EDS spectrum taken within the powder particle, has not given the evidence of the oxide existence, as shown in Fig. 6(d). Therefore, it can be deduced that there exists the oxide film at the interface between Al powder particles.

TEM observations and EDS analyses are carried on several interfaces between powder particles in these specimens. The analyses show that there are a lot of the metal/metal bonding and the relatively little of metal/oxide film layer/metal bonding for the powder compact by 873 K, 9.4 MPa. On the contrary, there is a lot of metal/oxide film layer/metal bonding for that by 623 K, 23.5 MPa. From the observed results, we can deduce that the cause of the difference in properties for powder compacts are mainly subject to the behavior of oxide film at the interface between Al powder particles. For breakdown of the oxide film at the Al powder particles surface, we think that it is mainly caused by plastic deformation of powder particles under loading pressure. The hardness ratio of alumina to aluminum metal is 1800 : 20 at room temperature. And increasing in the sintering temperature, the matrix aluminum is further softened. The hardness ratio becomes larger. The breakdown of oxide film at Al powder particles surface becomes easy. Therefore, the powder compact by 873 K, 9.4 MPa is provided with a lot of the metal/metal bonding than that by 623 K, 23.5 MPa. Although both have the similar relative density, the electrical resistivity of powder compact of the farmer is lower, and tensile strength, elongation, reduction of area are larger than those of the latter.

4. Conclusions

Pure Al powder was sintered by SPS process at various sintering temperatures and loading pressures. The properties of powder compacts and microstructure at interface between

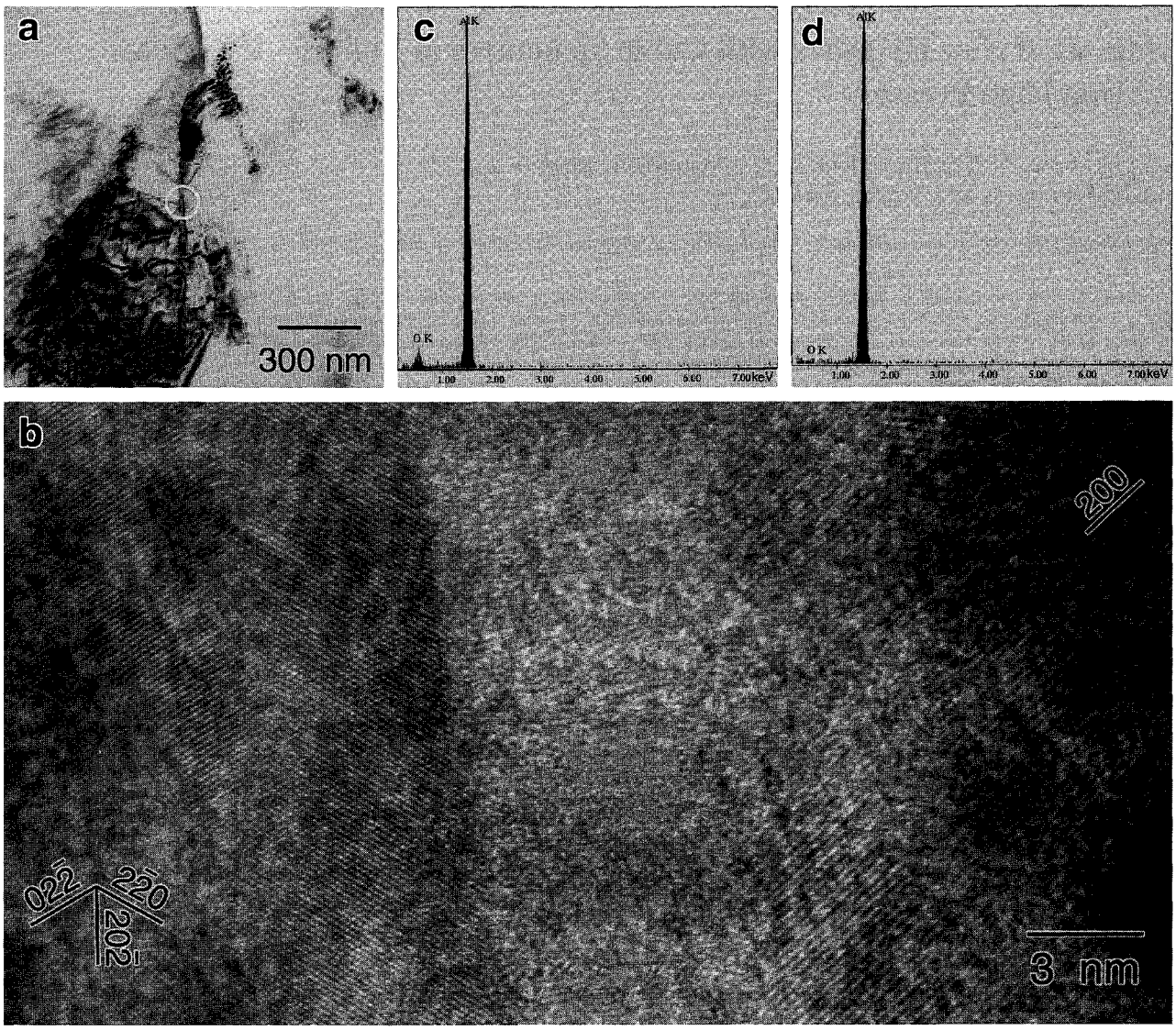

Fig. 6 TEM micrographs and EDS spectra of Al powder compact using SPS process by 623 K, 23.5 MPa. (a) bright field TEM image; (b) an HRTEM micrograph of the typical metal/oxide film layer/metal bonding taken from the region indicated by circle in (a); (c) EDS spectrum at the interface region between powder particles; (d) EDS spectrum within powder particles.

powder particles were investigated. The results obtained are summarized as follows.

(1) Two types of interfaces, with metal/metal bonding and metal/oxide film layer/metal bonding, were observed in powder compacts. The properties of powder compacts were mainly subject to the behavior of oxide film between the powder particles.

(2) Increasing in sintering temperature or loading pressure, relative density and tensile strength of powder compacts increase, and the electrical resistivity decreases. At SPS process conditions above 873 K, above 23.5 MPa, breakdown of oxide film at Al powder particles surface and metal/metal bonding is dominant in the compacts, the properties of powder compacts are similar to that of base aluminum metal.

Acknowledgements

The authors would like to express our gratitude to Mr. K. Chiba and Mr. T. Umeda of Niigata University for their helps in experiment. Authors gratefully also acknowledges the financial support from "2000 Niigata University Project Development Foundation" and "Sasaki Environment Technology Promotion Foundation".

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