PREPARATION OF Al_2O_3/Pd PARTICULATE COMPOSITE USING AN ELECTROLESS PLATING TECHNIQUE

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ABSTRACT

Dispersed-phase metallic inclusions in ceramics are expected to toughen the ceramics. For this aim, Pd metal layers were formed on each surface of Al_2O_3 particles using an electroless plating technique. The coated powders were hot-pressed to form composites. Fracture toughness increased to 4.5MPa$m^{1/2}$ for the >10 vol% composites but flexural Strengths decreased somewhat. Low vol% composites were electrical insulators. The >15 vol% composites, however, showed Metallic conductivity. SEM observation and EDX analysis on the fracture surfaces of the composites revealed that fine Pd particulates were dispersed uniformly in matrix by this dispersion procedure.

INTRODUCTION

Recent studies have revealed that mechanical properties of ceramics can be fairly improved by incorporating metallic inclusions, for instance, SUS particulate reinforced PSZ ceramics (1) and SUS particulate reinforced MgO ceramics (2). These ceramics composites are expected to develop into so-called Functionaly Gradient Materials (FGM). However, studies of these metal-ceramics system are few. In our work previously reported (3), ductile and chemically non-reactive Pd metal particles were dispersed in Al_2O_3 matrix by ball mill mixing using ethanol as a solvent. The powder consisted of pure Al_2O_3 particles of 0.15μm in average diameter and Pd metal particles of average 0.3μm. The mixed powders were hot-pressed into a composite, which was mechanically characterized. A large agglomerate of Pd metal particles of about 5μm in size was observed in the fracture surface of the composite shown in Fig.1. This suggests that the large agglomerates were formed from the secondary particles of Pd metal during hot-pressing. Thus, Pd particles could not be dispersed sufficiently by ball mill mixing. Presence of Pd agglomerates resulted in weakening of the composite.

In order to avoid the formation of mechanical flaw centers resulted from segregation of Pd particles, it is necessary to disperse metal particulates as uniformly as possible. For this aim, a Pd metal layer was formed by chemical deposition on each surface of Al_2O_3 particle by utilizing an electroless plating technique. The Pd coated powders were hot-pressed in to a composite under a flowing nitrogen atmosphere, which was mechanically and electrically characterized. SEM observation and EDX analysis were conducted on the Pd coated particle and the fracture surface of the composite, to examine dispersion of Pd metal on the surface of the coated particle and in the composite.

EXPERIMENTAL PROCEDURE

(1) Electroless Plating on Al_2O_3 particle

Electroless Plating techniques are widely applied for applied the preparation of printing circuits or Hybrid IC. An application for structural materials was reported that Ni metal layers were deposited on carbon fibers by this technique to toughen ceramics (4). As mentioned already, this approach was used to disperse Pd metal particles in Pd/Al_2O_3 composites as uniformly as possible as follows (Fig.2).

Firstly, α-Al_2O_3 powders (Taimei Chem. Co,Ltd.) of 0.15 μm in average diameter were dispersed and suspended in deionized water. After then, necessary amount of aqueous solution of palladium chloride was added in the suspensin and sufficiently mixed. As a reducing agent, aqueous solution of hydrazine was added drop by drop to the suspension, stirring the suspension vigorously. Thus, Pd metal layers were slowly formed on Al_2O_3 particles. After the reaction, the coated powders were washed by repeated sedimentation, decantation and addition

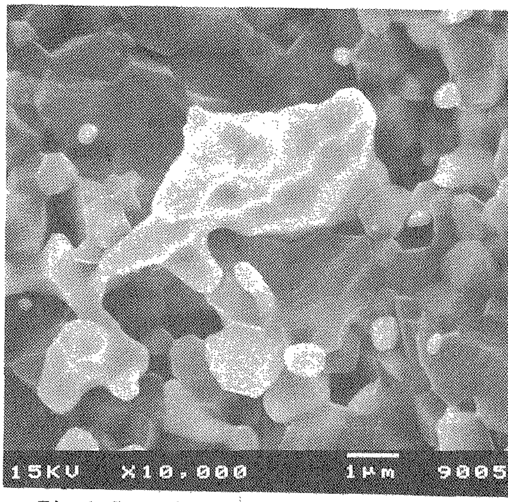


Fig. 1 Scanning electron micrograph on the fracture surface of Al_2O_3 composite dispersed by the ball milling method. Largest Pd-particle size is about 5 μm.

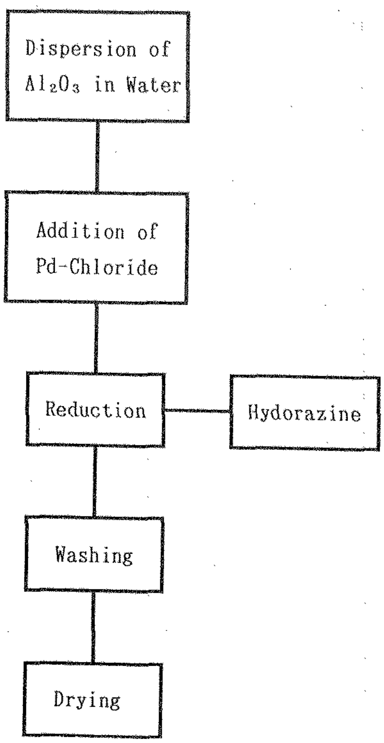


Fig. 2 Process of Pd electroless plating on Al_2O_3 powders.

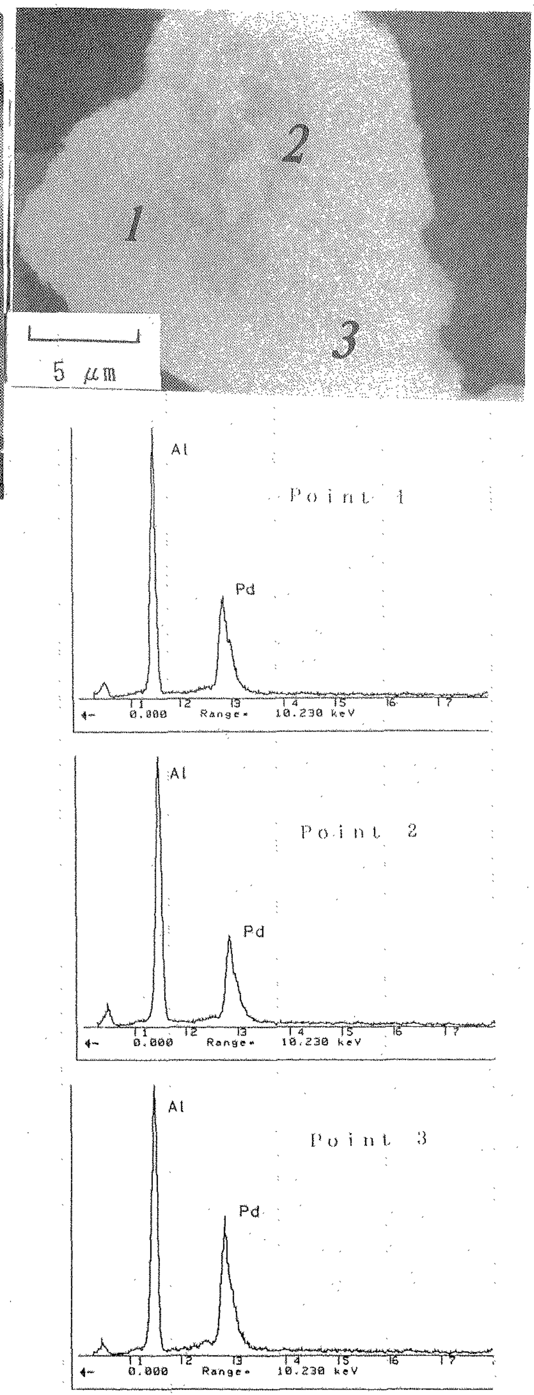


Fig. 3 Scanning electron micrograph and three points EDX analysis an agglomerated Al_2O_3 particle coated with Pd.

of deionized water and then dried. Pd contents were chosen in the range from 3 to 20vol% of composite. The color of the powder changed from white to black, showing the degree of Pd metal deposit on the particles. Hereafter, the particles thus prepared are called as EP particles. SEM observation of the particles showed that a secondary particle of about 20μm in diameter was formed by coagulation of finer primary particles (Fig. 3). EDX analysis on the three points of the agglomarated particle revealed that fairly uniform condensation of Pd metal was attained on the particle because the rate of Pd to Al peak height coincides fairly well at the three points.

(2) Preparation of Composites and Their Mechanical Characterization

In, addition to EP powders, mixed powders (BM powders) prepared by ball mill were used in comparison. α-Al_2O_3 particles (Taimei Co. Ltd) was mixed with Pd metal particles of 0.3 μm in average diameter using ethanol as a dispersion agent by ball milling. These mixed powders were hot-pressed in graphite dies into an EP composite or a BM composite under a pressure of 30MPa in a flowing nitrogen gas atmosphere at 1350°C for 45 minutes. Specimens for mechanical tests were machined from the hot-pressed composites and surface-ground with 1000-grit diamond wheel into rectangular bars of 4mm by 3mm by 40mm. Relative density was measured by Archimedes principle and flexural strength was determined in three point flexure for these specimen. Values of fracture toughness for the 0, 3, and 10 vol% composites and Vickers hardness for all composites were obtained by introducing cracks in the polished surfaces of the tips of the flexure-tested bars by Vickers indentations method (5). For 15 and 20 vol% composites, value of fracture toughness was obtained by the Chevron notched beam method (6), since extreme difficulties occurred for obtaining polished surfaces of these composites to observe propagation of indentation cracks. These mechanical tests were carried out on the surface vertical to hot-pressing direction. Electrical resistivity of the composites were measured by the standard 4-probe tchnique. The microstructures and metal particle dispersion of the composite sample were characterized by scanning electron microscope(SEM) observations and EDX analysis on the fracture surfaces of the tested bars.

RESULTS AND DISCUSSION

The relative density of composite is almost constant (Fig. 4). On the other hand, the hardness of the composite substantially decreases with Pd content, probably because of metallic ductility of Pd particulate (Fig. 5). Moreover, the flexural strength of the composite decreases to a half of the

Al_2O_3 (Fig. 6). The fracture toughness of the composite increases by addition of Pd to 5 MPam$^{1/2}$ as expected, however, not linearly with Pd content (Fig. 7). Difference in mechanical properties between the EP composite and the BM composite is obscure. It suggests that effect of improved particulate dispersion on the mechanical properties of the composite is not so remarkable as expected.

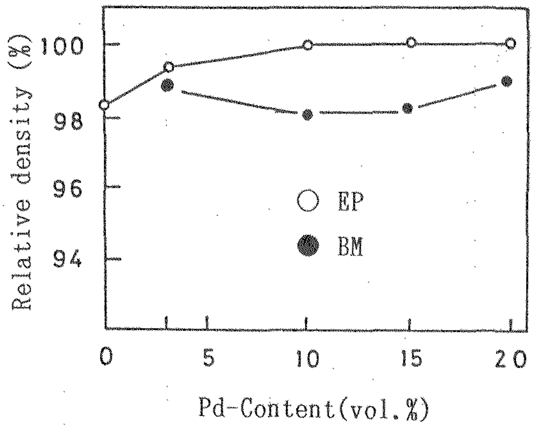


Fig.4 Relative density vs. Pd content of composites.

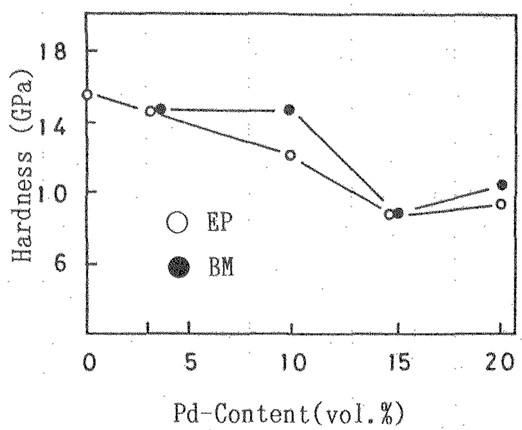
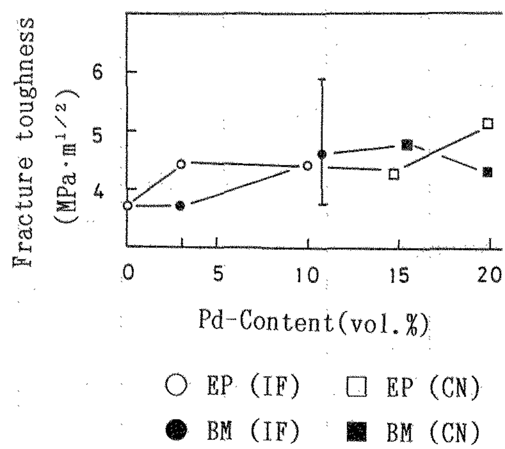


Fig.5 Hardness vs. Pd content of composites.

EP: Electroless Plating
BM: Ball Mill



IF: Indentation method
CN: Chevron notch method

Fig.7 Fracture toughness vs. Pd content of composites.

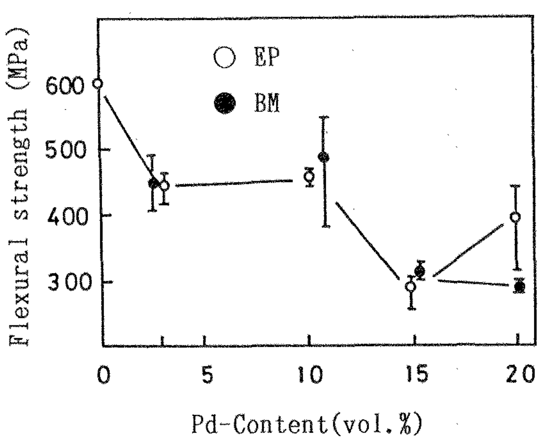


Fig.6 Flexural strength Pd content of composites.

Figure 8 shows SEM observation of the fracture surface of the EP composite, and EDX analysis of Al_2O_3 matrix (A) and dispersed Pd particle (B). Uniform distribution of spherical Pd particles are observed. This indicates that uniform metal particle dispersion is attained by the EP method.

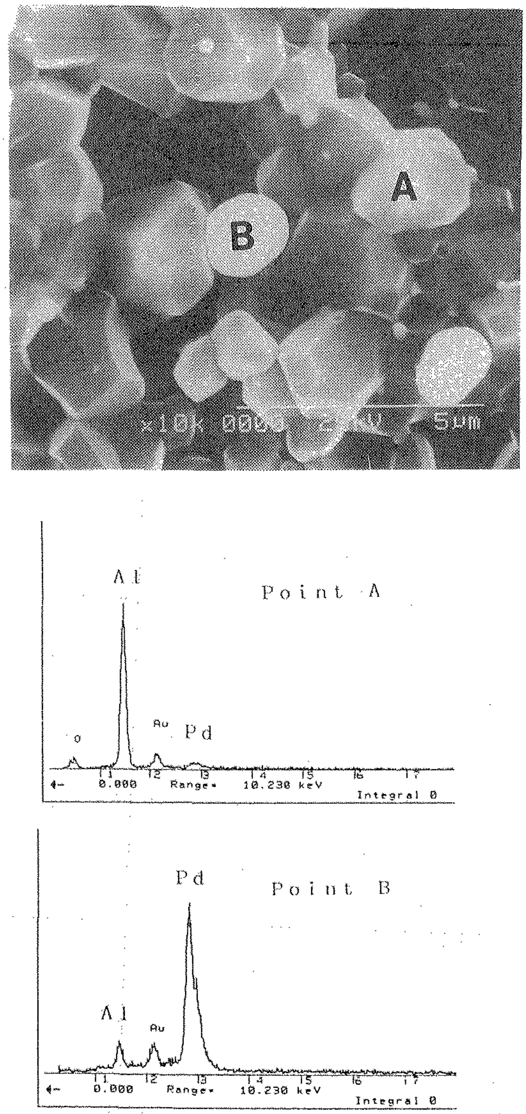


Fig.8 Scanning electron micrograph and two points EDX analysis on the fracture surface Pd/Al_2O_3 composites.

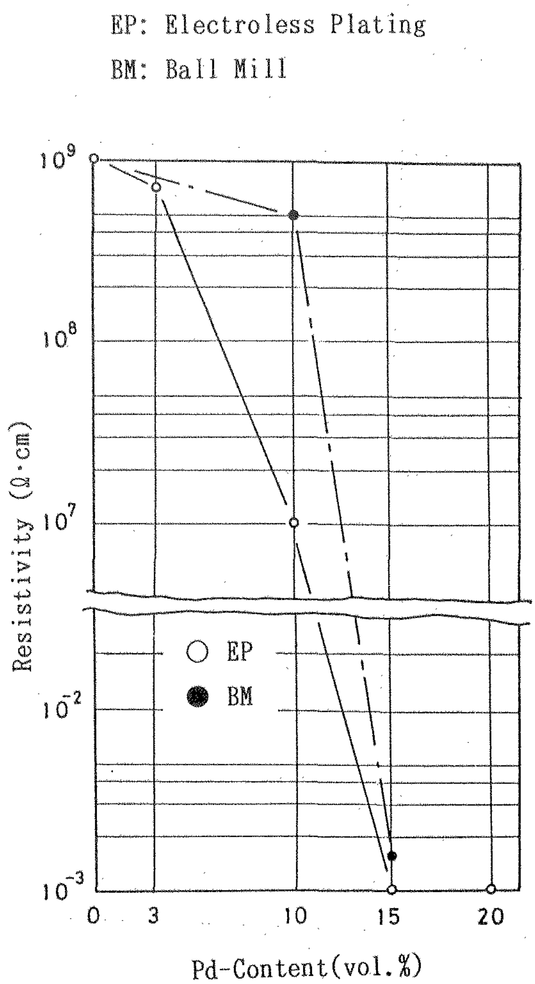


Fig.9 Resistivity vs. Pd content of composites.

On the other hand, small amounts of Pd metal are found on Al_2O_3 matrix, probably due to diffusion of Pd metal into Al_2O_3 matrix at high temperature may account for abrupt increase in electrical resistivity mentioned below.

The electrical resistivity of the composite decreases abruptly for 10vol% addition by the EP method and to almost metallic values for more than 15% addition by both methods (Fig. 9). Hence, both the EP composite and the BM composite show almost similar mechanical and electrical behaviours for more than 15 vol% addition of Pd metal particles.

CONCLUSION

The fracture toughness of the Pd metal particulate reinforced Al_2O_3 composite increased a little , while its hardness and flexural strength decreased. No substantial differences in mechanical and electrical properties were observed between the EP composite and the BM composite, though the Pd particles were fairly well dispersed in the former compared to the latter. Thus, other metal/Al_2O_3 composite system should be studied to find another possibility for toughening the ceramics by metal particulate incorporation.

Acknowledgment: We thank Miss. T. Fukuyama at Shonan Institute of Technology for her assistance in SEM observations and EDX analysis.

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