Effect of Internal Structure on Thermal Properties of Alumina/Aluminum Composites Fabricated by Gelate-Freezing and Partial-Sintering Process, Respectively

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The purpose of the present work was to clarify the effects of parameters of porous ceramics on the characteristics of composites fabricated by high-pressure infiltration. Alumina with different porosity (15–70 vol%) and pore morphology was successfully fabricated by gelate-freezing and the partial-sintering, respectively. Alumina/aluminum composites were made by squeeze casting, the aluminum into the alumina sintered bodies. By evaluating the coefficient of thermal expansion (CTE) and thermal conductivity of the composites, it became obvious that variation in the structure and fraction of porous alumina sintered bodies influenced CTE, but not thermal conductivity.

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1. Introduction

Insulated Gate Bipolar Transistors (IGBTs) are widely used in various fields of industry from production of home electrical appliances such as freezers and air-conditioners to large transport machinery such as electric and hybrid cars and electric trains. IGBTs are at the heart of the electric systems in electric and hybrid cars, which have attracted much attention as being environmentally conscious, and this is one example of their highly efficient, practical use in key technologies. Because it is difficult to maintain the strict conditions required in engine compartments using the traditional heat-spreaders (copper foil-lined ceramic) used in IGBTs, new types of ceramic/metal composites are being researched as possible future heat-spreaders.

High-pressure infiltration is one of the easiest ways to fabricate ceramic/metal composites for use as new types of heat-spreaders. These materials are more advantageous than traditional lining-type heat-spreaders and their economical advantages make them attractive in automobile industries.

Ceramic/aluminum composites can be fabricated by infiltrating aluminum melt into porous ceramic by high-pressure infiltration, that is, squeeze casting.1-3 Therefore, the ceramics and aluminum form a continuous composite, the thermal and mechanical characteristics of which are expected to be affected by both the porous ceramic as well as the structure of the aluminum. No experimental studies have examined the properties of these composites, and to do so it is necessary to consider the porosity, pore size, pore distribution and pore continuity of the initial porous ceramic. The purpose of this study was to clarify the effects of the above ceramic parameters on the characteristics of composites fabricated by high-pressure infiltration with the aid of an existing composite theory, leading to the development of new, advanced heat-spreaders for use in power devices.

2. Experimented Procedures

2.1 Fabrication of porous ceramic by the gelate-freezing

For IGBT applications, composites need to possess high thermal conductivity and low thermal expansion. Because, in general, metallic materials such as Cu and Al have better thermal conductivity than ceramics, porous ceramics must have high porosity, highly continuous pores and a dense framework structure to enable incorporation of the metallic phase.

Freeze-dry processes have attracted much attention in recent years in the fabrication of ceramic filters and porous bodies for catalysts and so on.4-7 This method has also been examined in functional ceramics, but not in ceramic/metal composites. In this study, ceramic forms with various characteristics can be successfully fabricated by the freeze-dry process with water-based polymer/alumina slurry (gelate-freezing process). This process allows introduction of a large volume fraction of pores into the alumina sintered bodies. A flow chart of the fabrication of porous alumina sintered bodies by the gelate-freezing process is shown in Fig. 1.

2.1.1 Mixing and gelling

Fine-grained alumina powder (Sumitomo Chem. Co. Ltd., AKP30: grain size 0.3 μm) and water-soluble polymer solution (polyethyleneimine 3 to 10 mass% solution) were prepared. The weight ratio of alumina powder to polymer solution was varied from 4 g: 2 g, 4 g: 3 g and 4 g: 4 g; the slurry concentrations were 33, 25 and 20 vol%, respectively.

Alumina powder and polymer solution were poured into a mixing vessel 35 mm in diameter, mixed for 30 s then de-aired for 30 s using a stirrer. A gelling agent (di-glycerol-glycergel-ethel) was added to the slurry at a slurry to agent ratio of 10 g: 1.21 g; it was then mixed for a further 30 s and de-aired for 30 s using a stirrer. The slurry was kept in air for 30 min–3 h depending on the slurry concentration to allow gelling.

2.1.2 Freeze and drying

After confirming the gel state, the slurry was placed in a
freezer at –20°C for 3 h. The frozen slurry was then removed from the vessel, placed in a drying oven and dried at 35°C in a controlled atmosphere. Drying level was assumed by weighing the samples; the drying time required ranged from 3–12 h.  

### 2.1.3 Burning and sintering

To remove the polymer by burning, dried samples were kept at 600°C for 1 h in an electric furnace then cooled slowly. Finally, the samples were placed in an electric furnace and sintered at 1500°C for 3 h. The gelate-freezing resulted in the following:

i) Use of a water-based polymer in the slurry allowed porosity to be controlled to up to 70% and pore size up to 200 μm.

ii) By cohesion of the polymer during drying, the structure of the alumina became denser.

iii) Reduced pressure treatment was not needed in the drying process.

### 2.2 Fabrication of small-pored alumina sintered bodies by the partial-sintering

Another small-pored ceramic was fabricated by a traditional method, so called the partial-sintering, to compare the effects of micro pore structure. Three alumina powders of different grain size (Sumitomo Chem. Co. Ltd., AKP15, ALM43 and AL43: grain size 0.7 μm, 3 μm and 30 μm respectively) were prepared then packed into a vessel (30 mm × 40 mm) and pressed at 20 MPa into a preform. In this case, the sizes of the pores in the alumina sintered bodies depended upon the grain size of the alumina powders. The alumina preforms were sintered at 1500°C for 3 h in an electric furnace resulting in fabrication of small-pored alumina sintered bodies. In the case of course-grained ceramic powders, alumina did not promote a dense structure during sintering and, as a result, non-sintered parts remained.

### 2.3 Fabrication of aluminum/alumina composites

Aluminum/alumina composites were fabricated using the porous sintered alumina bodies by high-pressure infiltration (the squeeze casting method). The sintered alumina was pre-heated at 700°C in an electric furnace, and a casting mold was pre-heated at 500°C. Pure aluminum melt (99.9 mass%) was kept at 800°C in a crucible. Next, the sintered alumina was placed in the mold into which aluminum melt was then poured. Immediately following this, a pressure of 40 MPa was applied by a hydraulic piston for infiltration and solidification. After confirming solidification, the pressure was removed and the sample was cooled to room temperature in a furnace.

### 2.4 Measurement and observation

Specimens for measuring thermal diffusivity, specific heat and the coefficient of thermal expansion (CTE) were cut from the aluminum/alumina composites using a diamond cutter. The surfaces of the specimens were polished by sand paper #2000, and then with 3-μm diamond slurry.

For evaluation of micro framework structure, the following characteristics were measured:

i) Surfaces of specimens were observed by optical microscopy and scanning electron microscopy. The pores of the alumina sintered bodies were filled with polystyrene to allow observation of the microstructures in the sample wells.

ii) Porosity was measured by mercury porosimetry.

iii) Thermal diffusivity was measured from at room temperature to 300°C by the laser flash method.

iv) Specific heat was measured from at room temperature to 310°C by differential scanning calorimeter (DSC).

v) CTE was measured from at room temperature to 400°C using a thermal mechanical analyzer (TMA).

### 3. Results and Discussion

#### 3.1 Microstructure

##### 3.1.1 Microstructure of the porous alumina sintered bodies fabricated by the gelate-freezing

Optical microscopic photographs of the porous alumina sintered bodies fabricated by the gelate-freezing are shown in Figs. 2(a), (b) and (c). Table 1 shows the slurry and polymer concentrations of the solution and porosity of the porous alumina sintered bodies fabricated by the gelate-freezing. The porosity, pore size and pore shape of the alumina sintered bodies varied according to the various slurry and polymer content. Porosity was affected by both slurry and polymer content and pore size showed a tendency to increase with decreasing slurry content. Pore shape depended upon both slurry and polymer content. The pore aspect ratio decreased with decreasing slurry content while the pore shapes showed a tendency to sharpen with decreasing polymer content. Pore size distribution was measured by mercury porosimetry; pore sizes of less than 10 μm were not observed. Therefore, it was confirmed that the porous
3.1.2 Microstructure of the porous alumina sintered bodies fabricated by the partial-sintering

The porosity of the porous alumina sintered bodies fabricated by the partial-sintering is shown in Table 2, and the relationship between porosity and pore size cumulatively is shown in Fig. 3. Porosity increased according to grain size of the alumina powder, and pore size increased with increasing porosity. A pore size of more than 6 \( \mu m \) was not observed; therefore it was confirmed that alumina sintered bodies fabricated by the partial-sintering had a small pore size.

3.1.3 Microscopic photographs of the composites

Microscopic photographs of the composites fabricated by the gelate-freezing are shown in Fig. 4(a) and those fabricated by the partial-sintering in Fig. 4(b). The structure of the composites was strongly reflected by the shape of the porous ceramic. Cracks and separation were not observed on the interface of the aluminum and alumina. The porosity of the porous alumina sintered bodies and aluminum infiltration ratio of the composites corresponded with each other, and among different sized specimens; pores not filled with aluminum were not observed.

3.2 Coefficient of thermal expansion (CTE)

Differences in CTE between the heat-spreader and substrate cause thermal stress in circuit boards; therefore, CTE is an important parameter for ceramic substrates. CTE depended upon the volume fraction of alumina and shape of alumina sintered bodies fabricated by the gelate-freezing had larger pores and micro framework structure.

### Table 2 Porosity of porous alumina sintered fabricated by partial-sintering.

<table>
<thead>
<tr>
<th>Powder diameter (( \mu m ))</th>
<th>Porosity (vol%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>AKP15</td>
<td>0.7</td>
</tr>
<tr>
<td>ALM43</td>
<td>3.0</td>
</tr>
<tr>
<td>AL43</td>
<td>30</td>
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Fig. 3 Cumulatively of pore size for porous alumina sintered body by the partial-sintering.
the infiltrated aluminum. But the volume fraction and shape of aluminum depended upon the shape and structure of the porous alumina sintered bodies. Therefore in substances, the CTE of composites is determined by the porosity and pore distribution of the porous alumina sintered bodies.

3.2.1 Thermal expansion of the composites

The relationship between the thermal expansion and heating temperatures of composites fabricated by the gelate-freezing and the partial-sintering, respectively, are shown in Fig. 5. The thermal expansion of composites fabricated by the gelate-freezing varied linearly from 50 to 300°C; above 300°C the varying rate decreased with softening of the aluminum, and at a constant temperature of 400°C expansion decreased with relieved stress. The thermal expansion of composites fabricated by the partial-sintering varied linearly from 300 to 400°C.

Though the aluminum volume fraction of both composites differed by 8%, thermal expansion was nearly equal from 50 to 300°C; but at 400°C expansion of composites fabricated by the gelate-freezing was lower than that of composites fabricated by the partial-sintering. These differences were caused by differences in the structure of the alumina part. Inside the composite body, alumina suppressed the expansion of aluminum as a result of the difference in thermal expansion between alumina and aluminum. Therefore, thermal expansion of the composite was determined by the balance between thermal expansion of the alumina and aluminum. The porous alumina fabricated by the gelate-freezing had a dense structure and large pores; therefore, the thermal expansion of the resultant composites was lower than that of composites fabricated by the partial-sintering.

3.2.2 CTE of the composites

CTE varied with aluminum volume fraction as shown in Fig. 6. For comparison, the CTE of porous alumina formed by the partial-sintering and pure metal aluminum measured under identical conditions are shown in addition to the CTE of both composites. The CTE of composites fabricated by the partial-sintering was almost unchanged with increasing and decreasing temperature. In contrast, the CTE of composites fabricated by the gelate-freezing decreased with decreasing temperature, because expansion decreased at a constant temperature of 400°C. The CTE of the latter composites was lower than that of the former composites at all temperature and all aluminum contents; this was especially obvious at temperatures over 300°C with aluminum softening. Thus, in
composites fabricated by the gelate-freezing, it was confirmed that the CTE decreased as a result of the framework structure.

3.2.3 Comparison of the CTE to the rules of mixture

To clarify the difference in CTE with differing structure, the above results were compared to the common rules of mixture using the equations of (1) Turner and (2) Kerner.\(^{(1)}\)

\[
(1) \alpha_c = \frac{(\alpha_m \times V_m + \alpha_d \times V_d)}{(V_m \times \alpha_m + V_d \times \alpha_d)} \\
(2) \alpha_c = \frac{(\alpha_m + \alpha_d \times (V_m \times K_m + V_d \times K_d))}{(V_m \times (\alpha_m + K_m) + V_d \times K_d)}
\]

where, \(\alpha_c\) is CTE of composites, \(\alpha_m\) and \(\alpha_d\) is matrix and dispersion medium, \(V_m\) and \(V_d\) is volume fraction of matrix and dispersion medium, \(K_m\) and \(K_d\) is modulus of transverse elasticity, \(\rho_m\) and \(\rho_d\) is density, and \(G\) is CTE, volume elasticity, density, mass fraction, volume fraction and modulus of transverse elasticity. Index of \(m\) and \(d\) is matrix and dispersion medium respectively. The internal structure of the composites is not considered in Turner’s equation, so it is thought to show the lowest value of the rules of mixture. In Kerner’s equation, the shear stress at the interface of the grain and boundary layer is considered, and components of the composite are divided into matrix and dispersion medium. By reversing the volumes of alumina and aluminum, the highest and lowest values of the Kerner’s equation are obtained.

The average CTE was measured from 50 to 400°C, and the results of Turner’s and Kerner’s equations are shown in Fig. 6. The results of composites fabricated by the partial-sintering were in the highest and lowest limits of Kerner’s equation. Composites fabricated by high-pressure infiltration have an interpenetrating network with both the dispersion medium and matrix. The structure of the composites is therefore homogenized, and with regards to the structure of the dispersion medium, is very small. In this study, the CTE of composites fabricated by the partial-sintering were near the values of the grain dispersion model; that is, Kerner’s model. The results of composites fabricated by the gelate-freezing were nearly equal to or below those of Turner’s equation. This result proves that the framework of alumina has adequate strength to suppress the expansion of aluminum. It is considered that the values below those of Turner’s equation showed anisotropy as result of freezing the slurry.

3.3 Thermal conductivity

Thermal conductivity is an important parameter when determining the characteristics of heat-spreader material. Thermal conductivity of composites is determined mainly by the volume fraction or distribution of the dispersion medium.

3.3.1 Results of thermal conductivity

The temperature dependency of thermal conductivity is shown in Fig. 7. Thermal conductivity decreased gradually from 50 to 200°C, but was almost unchanged from 200 to 300°C. There was no difference in the thermal conductivity of composites fabricated by the gelate-freezing and those by the partial-sintering with the same aluminum content.

3.3.2 Comparison of the results of thermal conductivity to the rules of mixture

The influence of aluminum content on thermal conductivity at 46°C is shown in Fig. 8. Thermal conductivity was compared to three models of the rules of mixture: (3) the perpendicular model, (4) parallel model, and grain dispersion model. As the grain dispersion model, (5) Maxwell’s equation\(^{(10)}\) was used.

\[
(3) \lambda_c = \lambda_m \times V_m + \lambda_d \times V_d \\
(4) \lambda_c = (\lambda_m \times V_m + \lambda_d \times V_d) / (V_m + V_d) \\
\]

where, \(\lambda\) and \(V\) is thermal conductivity and volume fraction. Index \(c\), \(m\) and \(d\) is composite, matrix and dispersion medium respectively. Results of this comparison showed good correlation with the grain dispersion model. The perpendicular model and parallel model consider the structure of the composite, but the effect of structure and diameter of the dispersion medium is not considered in the grain dispersion model. This suggests that when the diameter of the dispersion medium was sufficiently smaller than the conduction distance, and especially when the dispersion medium was not anisotropic and dispersed at random, the conduction pass was leveled and dependency of conductivity on the internal structure was largely decreased. The diameter of the dispersion medium was 30–200μm in the gelate-freezed
composites and 1–6 μm in the partial sintered composites. Conduction distance was 2 mm (equal to the thickness of the specimens).

4. Conclusions

In this study, various kinds of porous alumina sintered bodies were fabricated by the gelate-freezing and the partial-sintering, respectively, and composites were fabricated by high-pressure infiltration using these alumina sintered bodies. The effects of the parameters of porous alumina sintered bodies on the characteristics of the composites were clarified. The results can be summarized as follows:

- The gelate-freezing of water-based polymer/alumina slurry resulted in successful fabrication of porous ceramics with a dense framework structure and large pores.
- Pore size and porosity could be controlled by polymer and slurry concentrations.
- The fabricated porous alumina sintered bodies had highly continuous pores and a dense alumina part.
- The porous alumina sintered bodies fabricated by the partial-sintering had small pores.
- After infiltration of aluminum, comparisons of composites fabricated by the gelate-freezing and the partial-sintering, respectively, revealed that the CTE of the former was lower than that of the latter, while the thermal conductivity of both composites was nearly equal.
- It became obvious that varying the structure of the porous alumina sintered bodies in the composite material affected the CTE, but not thermal conductivity.

REFERENCES

4) T. Fukusawa and H. Goto: Japanese patent no. 3124274.