Fabrication of Composite Material Using Alumina Agglomerated Sludge and Aluminum Powder by Spark Plasma Sintering

Isao FUKUMOTO**, Shunei MEKARU**, Shinichi SHIBATA**
and Kiyomitsu NAKAYAMA**

The composite material was fabricated using alumina sludge, industry waste, and aluminum powder by spark plasma sintering (SPS). Sludge of industry waste was treated to change the $\alpha$ alumina crystal structure at temperature 1573 K for 2 hours. The bending strength of the composite materials was investigated by changing the volume fraction of sludge 0–6% and forming conditions. As a result, it was found that the sludge content mainly affected on the bending strength. The bending strength showed the highest value at 2% sludge content. From the observation of crack propagation using optical microscope, it became clear that the sludge existed as agglomerated powder in the composite material, and this sludge prevented the crack to propagate.

Key Words: Powder, Aluminum, Alumina, Spark Plasma, Sintering, Composite, Sludge

1. Introduction

Spark plasma sintering (SPS) technique enables aluminum powder, which has the oxide film on the surface, to sinter at low temperature and short time in comparison with ordinary hot press. Significantly, as spark plasma can destroy the oxide film of aluminum powder, aluminum powder can be merged each other closely(1). In this study, we attempted the fabrication of composite material using aluminum powder and alumina ceramics material by SPS. $\alpha$ alumina ceramics was obtained by heat treatment of the alumite sludge, industry waste(2). $\alpha$ sludge particle existed as the state of agglomerated powder block. Then, the powder was considered to play an important role in the mechanical properties of the composite material. The composite materials were made by varying the content of $\alpha$ alumina sludge, 0–6%. From the analysis of variance on the bending strength of the sintered samples, it was found that the factors of sludge content, pressure and sintering temperature on SPS affected the bending strength. Especially, the bending strength at the 2% sludge content showed the highest value. From the comparison of the sludge content 0% and 2%, it was observed that the crack propagation was avoided by the agglomerated sludge at 2% sludge content.

2. Materials and Method

2.1 SPS

SPS also known as pulsed electric current sintering was carried out in vacuum using Dr sinter SPS-2050 apparatus (Sumitomo Coal Mining Co., Ltd., Japan). Figure 1 shows the schematic diagram of SPS. The prepared composite powder was carefully placed into a 50 mm diameter graphite die, and heated up to the sintering temperature at a heating program rate. During fabrication, a pressure of 10–40 MPa was kept. Sintered samples were approximately 50 mm in diameter and 5 mm thickness.
Table 1 Chemical compositions of aluminum powder (mass%)  

| Element | Cu | Fe | Si | Mn | Mg | Zn | Ni | Cr | Ti | Al |
|---------|----|----|----|----|----|----|----|----|----|----|
| Mass%   | 4.60 | 0.08 | 7.75 | 0.66 | 4.70 | TR | 0.04 | TR | TR | 80.1 |

Table 2 Chemical compositions of α alumina sludge (mass%)  

| Component | SiO₂ | Al₂O₃ | Fe₂O₃ | CaO | MgO | Na₂O | Ig. Loss |
|-----------|------|-------|-------|-----|-----|------|---------|
| Mass%     | 0.99 | 91.2  | 0.3   | 0.02 | 0.21 | 0.72 | 6.56    |

Table 3 Factor and level in orthogonal table  

| Factor | Level |
|--------|-------|
| A      | 0     |
| B      | 3     |
| C      | 5     |
| D      | 6     |
| E      | 1     |
| F      | 7     |
| G      | 10    |
| H      | 15    |

Table 4 Orthogonal table L18 for bending strength  

| A | B | C | D | E | F | G | H | I | J | K | L | M | N | O | P | Q | R | S | T | U | V | W | X | Y | Z |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | 1 | 2 | 1 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 |
| 2 | 1 | 2 | 1 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 |
| 3 | 1 | 2 | 1 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 |

Table 5 Analysis of variance on bending strength  

| Factor | f | S | V | ρ (%) |
|--------|---|---|---|-------|
| A      | 3.54 | 3.54 | 2.4 |
| B      | 5.81 | 2.90 | 3.4 |
| C      | 47.19 | 23.60 | 50.0 |
| D      | 9.37 | 4.68 | 7.4 |
| E      | 7.38 | 5.69 | 5.2 |
| F      | 9.47 | 4.74 | 7.6 |
| G      | 0.67 | 0.33 | 0.0 |
| A×B    | 0.93 | 0.47 | 0.0 |
| e⁴     | 4.58 | 2.29 | 2.1 |
| e⁵     | 5.51 | 1.38 | 21.9 |
| Total  | 88.94 | 100.0 |

\[
\eta = -10 \log \left( \frac{1}{n} \sum \frac{1}{y_1^2} + \frac{1}{y_2^2} + \cdots + \frac{1}{y_n^2} \right)
\]

\(\eta\): SN ratio (dB) \(y\): Bending strength

Table 5 shows the result of analysis of variance on bending strength. The symbol f, S, V and p mean freedom, scattering, variance and contribution ratio. The contribution ratio of sludge content factor showed the highest value of 50% and the setting temperature and the pressure showed the value of 7.6% and 7.4% respectively. Though we had expected the factors of milling time of sludge and bending time during aluminum powders and sludge have the relationship with bending strength, those factors have

2.2 Material

Table 1 shows the chemical compositions of aluminum powder supplied from Toyo Aluminum Company. Table 2 shows chemical compositions of α alumina sludge (α sludge) obtained by heat treating, alumite sludge at 1573 K for 2 hours. Figure 2 shows the microphotograph of aluminum powders and α sludge by using scanning electron microscope (SEM). These powder by varying the sludge content in the range of 2–6 volume % were blended by V-blender machine. Sintering temperature and pressure etc were selected as factors for SPS.

2.3 Mechanical properties and observation of microstructure

Sintered samples were cut into rectangular bar specimens (5×4×45 mm). Bending strength was measured with Shimazu AUTOGRAPH AG-50kND using three point bending test with a span length of 36 mm and cross head speed of 0.5 mm/min. Observation of microstructure were carried out using an optical microscope and SEM.

3. Results and Discussion

There are lots of factors which affect on mechanical properties of sintered samples. Because of many factors such as the ingredient ratio of sludge, blending and forming condition on fabrication of the composite material, we adopted the Taguchi Method for deciding the optimum condition. Table 3 shows the factors and levels. Table 4 shows the orthogonal table (L18) which means 18 kinds of combination in experimental conditions with 7 factors and 2 or 3 levels. The bending strength was transformed into SN ratio (η) in order to determine the effect of some kinds of factors. SN ratio was obtained from next formula.
no effect on bending strength. Figure 3 shows the relationship between sludge content and SN ratio. As the sludge content increasing, the bending strength shows the decreasing value, especially 2% sludge content showed the highest value. Therefore, we investigated the structure of composite material when varying the sludge content 2%, 4% and 6%. Figure 4 shows the observation of structure by microscope. When the sludge content increasing from 2% to 6%, the distance between the agglomerated sludge became shorter, because of the same agglomerated sludge size of about 30µm in diameter. Figure 5 shows the microphotographs of the fracture surfaces after bending test. The fracture surfaces were so rough and similar that the fracture pattern indicated the agglomerated sludge separated from the aluminum matrix. Hence, when the sludge content increasing from 2% to 6%, the crack easily occurred in boundary surface and connected each other.

Next, we investigated the effect of pressure and sintering temperature conditions. Though high pressure is desire for getting high density, the pressure was selected to set 30 MPa in order to avoid the failure of die and punch. Above 773 K, aluminum powders was found to melt and leak as liquid through the clearance of the punch and dies. Therefore, we compared the bending strength with and without sludge 2% on the setting pressure 30 MPa and sintering temperature 773 K. Figure 6 shows the comparison of the bending strength during aluminum only and 2% sludge content composite material. As compared with aluminum only 468 MPa, composite material showed 541 MPa. Figure 7 shows the SEM microphotograph of fracture surfaces after bending test. The flat surface was observed on aluminum only, while the surface of 2% sludge composite material is rough. From the observation of the cross sections after bending test as shown in Fig. 8, we could recognize the scattered black block in
Table 6 Micro vickers hardness of material

|                | HV1 | HV2 | HV3 | HV4 | HV5 | HV6 | Average |
|----------------|-----|-----|-----|-----|-----|-----|---------|
| composite      |     |     |     |     |     |     |         |
| Matrix         | 82.6| 85.9| 83.3| 87.9| 87.9| 87.2| 86.1    |
| Black block    | 252 | 441 | 444 | 480 | 272 | 380 | 365.0   |
| Aluminum only  | 84.4| 81.1| 83.8| 82.5| 84.9| 85.6| 83.7    |

Fig. 6 Comparison of bending strength between aluminum only and composite material

Fig. 7 Microphotograph of fracture surface on bending test

Aluminum only   Aluminum + 2% α sludge

2% sludge content composite material. As compared with the crack propagations, the crack in aluminum only propagated straightly, the crack in 2% sludge composite material bypassed the black block. Therefore, we measured the hardness of the area around the block. Table 6 shows the micro vickers hardness of material. The hardness of black blocks is higher than the aluminum matrix. Hence, it is confirmed that the black block was agglomerated α sludge particle. This phenomenon indicated that the agglomerated sludge plays a role for avoiding the crack propagation. Furthermore, this obstruction implies that 2% agglomerated α sludge particle performed as a reinforcement of the composite material.

4. Conclusion

By adding α alumina sludge obtained by heating aluminate sludge, industry waste, at 1573 K into the aluminum powder, composite materials were fabricated by spark plasma sintering technique. The mechanical properties were investigated by changing the α sludge content volume 0–6%. The obtained results are as follows.

1. From the analysis of variance on bending strength, it became obvious that the factors of α sludge content, pressure and heating temperature affected on the bending strength markedly.

2. α sludge particle existed as an agglomerated block in the composite material. At α sludge content 2%, the composite material showed the highest bending strength, because the agglomerated sludge obstructed the crack propagation.

3. From the observation of the fracture surface and cross section by SEM and an optical microscope, in aluminum only, the crack propagated straightly, and 2% sludge composite material, the crack bypassed the agglomerated sludge.

Acknowledgment

We greatly appreciate the help from the scholarship provided by the Light Metal Educational Foundation.

References

(1) Nagae, T., Yokota, M. and Nase, M., Sintering Process and Mechanical Properties of High-Si-Al Alloy Powder by Spark Plasma Sintering, Funtai Oyobi Funmat-suyakin, (in Japanese), Vol.44, No.10 (1997), pp.945–949.

(2) Fukumoto, I. and Yonaha, T., Determination of Forming Conditions for Injection Molding Products Using Aluminum Sludge as Material, LIGHT METALS REVIEW, Vol.5, July (1998), pp.7–12.