Low temperature sintering of Al_2O_3 according to the amount of Bi_2O_3 additive considering economics

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Structural ceramics are generally superior in strength to metal at high temperatures. However, they must be sintered at high temperatures to achieve this high strength. Therefore, lowering the sintering temperature would have economic benefits, including in terms of energy consumption. This study investigated the possibility of reducing the sintering temperature by adding bismuth oxide(Bi_2O_3) to the sintering of aluminium oxide(Al_2O_3). The amount of Bi_2O_3 additive was changed to 12~19.1 wt.% and the sintering temperature was in the range of 850~1200 ºC, and the compressive strength was evaluated under these conditions. The addition of Bi_2O_3 can decrease the necessary sintering temperature by about 400 ºC. Ceramic structures that can be low temperature sintering can save costs due to reduced manufacturing costs, replacement costs, shutdowns, etc., which will ensure enormous economic efficiency.

Key words: Al_2O_3, Bi_2O_3, Sintering temperature, Compressive strength, Economic effect.

Introduction

Aluminium oxide (Al_2O_3) is an advanced ceramic material that is used in a wide variety of applications due to its excellent fire resistance, chemical stability, and wear and deformation resistance. It is also a relatively abundant and low-cost resource, making it particularly attractive for commercial applications. Al_2O_3, like other ceramics, has excellent high temperature stability and high temperature strength. Al_2O_3 is known to have a direct effect on the sintering process because it changes the microstructure and interfacial energy of the sintered material with the use of additives. Silicon oxide (SiO_2) suppresses densification but promotes grain growth [1]. Magnesium oxide (MgO) can suppress the abnormal grain growth of Al_2O_3, and it can densify it to near theoretical density [2, 3]. The addition of titanium oxide(TiO_2) promotes not only the densification of Al_2O_3 but also grain growth [4, 5]. meanwhile, the addition of yttrium oxide(Y_2O_3) increases the strength and sintering properties [6-11]. However, the authors was sintered Al_2O_3 at 1,600 ºC and studied it [6]. Ando et al was sintered Mullite/SiC by hot-pressing at 1,650 ºC [11]. The addition of manganese oxide (MnO) has been shown to increase the density and lower the sintering temperature [12-14]. Bi_2O_3 has a low melting point of 825 ºC, and is therefore considered to be a suitable material for use as a ceramic sintering additive.

De Marco V. et al. reduced the normal sintering temperature by more than 400 ºC by using Bi_2O_3 as an additive of Gd: cerium oxide (CeO_2). [15] In another study, barium zirconate ($BaZrO_3$) was contracted by 19.0% at 1,480 ºC with the use of 3 mol% Bi_2O_3 additives, and the mixture reduced pore volume at sintering for 24 hours of 1,400 ºC [16].

This study investigated the effect of using sintering additive Bi_2O_3 on compressive strength by varying the amount of Bi_2O_3 as well as the sintering temperature in the sintering of Al_2O_3. From this study, the addition of Bi_2O_3 can lower the sintering temperature of alumina by about 400 ºC.

Materials and Experiment Methods

The Al_2O_3 powder used in this experiment had an average particle diameter of 0.5 μm (Korea CIS Co., Ltd.), the SiC powder had an average particle diameter of 0.27 μm (Betarundum UF, Ibiden, Japan), and the Y_2O_3 powder was 0.27 μm fine grade from Nippon Yttrium. Bi_2O_3 was obtained from Daejung chemicals & metals Co., Ltd (Korea).

The mixing ratio of the powder is shown in Table 1. A mixture of Al_2O_3, SiC, Y_2O_3, and Bi_2O_3 was mixed for 24 hours along with Al_2O_3 balls and alcohol. The mixed powder was then dried in a 100 ºC. furnace (Jisco, Model: J-300M) for 3 h before being filtered in a 100 μm sieve. next, the dry powder was placed into a cylindrical mold with a diameter of 10mm and molded to a pressure of 30 MPa. atmospheric sintering proceeded in a tubular furnace (Lenton, Model: LTF-180) of 1,200

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**Table 1.** Batch composition of each specimen (wt.%).

	Al_2O_3	SiC	Y_2O_3	Bi_2O_3
A	81.7	13.1	4.0	1.2
B	79.9	12.8	3.9	3.4
C	78.1	12.5	3.8	5.6
D	74.0	11.9	3.6	10.5
E	70.3	11.3	3.4	15.0
F	66.9	10.7	3.3	19.1

°C for 1 h and 30 min. The temperature increased at a rate of 5 °C/min. The properties of the sintering material were determined based on compressive strength and density. The fracture surface of the sintering material was observed by SEM (Scanning Electron Microscope: Hitachi (Japan), S-2700). The components of the sintering material were analyzed by EDX (Energy Dispersive X-Ray Spectrometer: Horiba (Japan)).

Results and Discussion

Fig. 1 shows the relative density of the Al_2O_3 sintering materials according to the amount of Bi_2O_3 added. This represents the percentage of Al_2O_3 theoretical density. The theoretical density of Al_2O_3 is 3.95 g/cm^3. It can be seen that relative density increases as the sintering temperature is increased when the amount of Bi_2O_3 is constant. In addition, at a low sintering temperature of 850 °C, the "A" specimen containing the least amount of Bi_2O_3 showed a relative density of about 85% while the "E" specimen containing the highest amount of Bi_2O_3 showed a relative density of about 86%. The relative densities of the "B~E" specimens showed almost similar relative densities at each temperature. The relative density of the "B" specimen at 1,100 °C was the highest at about 94.9%. The relative density of the specimen sintered at 1,200 °C was slightly lower than that of 1,100 °C. The Al_2O_3 sintering material with Bi_2O_3 additive shows excellent relative density when the sintering temperature is 1,100 °C and 1,200 °C, and its compressive strength is high. However, the "A" specimen with low Bi_2O_3 content and the "E" and "F" specimens with high Bi_2O_3 contents were small at 850 °C and 1,200 °C, and the relative density also decreased.

Fig. 2 shows the compressive strength of the Al_2O_3 sintering material according to the amount of Bi_2O_3 added. The figure also shows the standard deviation. The compressive strength was divided into two groups regardless of the amount of Bi_2O_3 added: 1,000 °C or less and 1,100 °C or more. In the case of "A" specimens with a small amount of Bi_2O_3, the compressive strength increased as the sintering temperature increased. The compressive strengths of the "B~E" specimens showed almost the same compressive strength at each sintering temperature. In the "F" specimen, there was a slight decrease in compressive strength. The compressive strength of the "B" specimen was the highest at 1,100
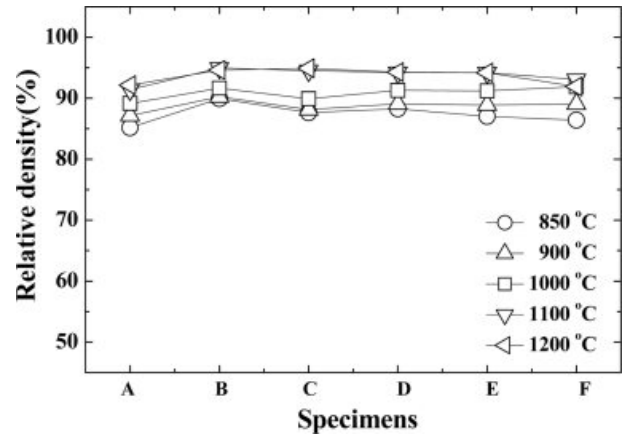
Fig. 1. Relative density of the sintered Al_2O_3 according to the amount of Bi_2O_3.
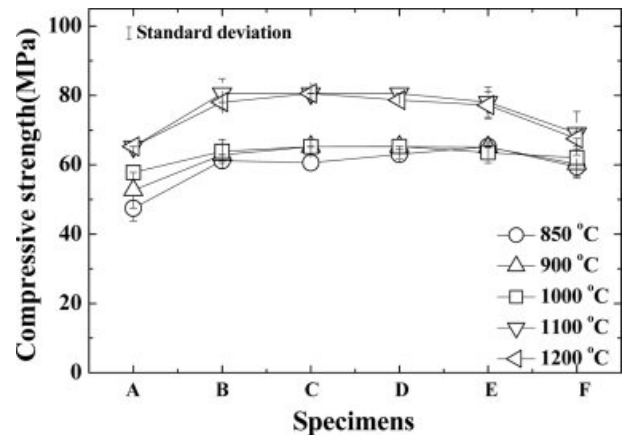
Fig. 2. Compressive strength of the sintered Al_2O_3 according to the amount of Bi_2O_3.

°C and 1,200 °C, but its compressive strength decreased as the content of Bi_2O_3 increased. This indicates that the addition of Bi_2O_3 can lead to a decrease of about 400~500 °C compared to the sintering temperature of 1,600 °C of Al_2O_3 with Y_2O_3 additive [17-20]. This is because the sintering material became dense as the sintering temperature increased.

Fig. 3 shows the SEM results of the fracture surface. (a) shows a "B" specimen sintered at 900 °C and (b) shows a "B" specimen sintered at 1,100 °C. (c) and (d) are "A" and "F" specimens sintered at 1,200 °C. When the same amount of Bi_2O_3 was added, it was confirmed that the relative density and the compressive strength increased as the sintering temperature increased. At each sintering temperature, the "A" and the "F" specimens appeared small while the "B~E" specimens appeared to be almost similar. (a) confirms that sintering was not completed because the temperature was low, and as a result the particles were not sintered. (b) shows the best condition of relative density and compressive strength. This means that the particles are sintered, and that many micro pores formed between the particles. Many
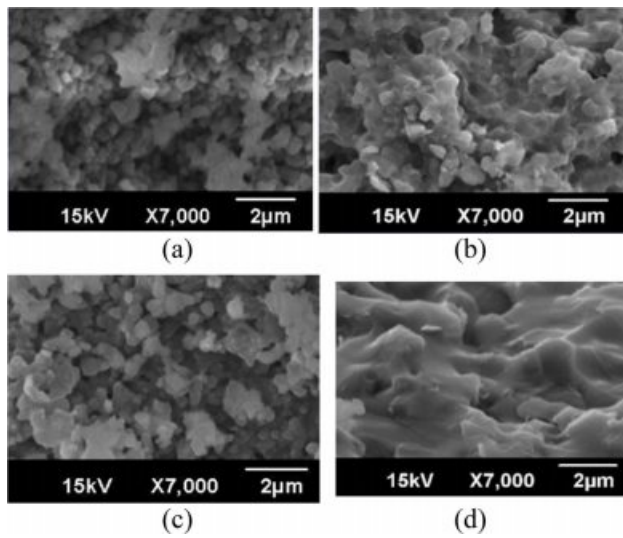
Fig. 3. SEM observation of fracture surface. (a) "B" specimen sintered at 900 °C, (b) "B" specimen sintered at 1,100 °C, (c) "A" specimen sintered at 1,200 °C, and (d) "F" specimen sintered at 1,200 °C.

of these pores disturb crack propagation, leading to it having the highest compressive strength. Although (c) had a sintering material of 1,200 °C, the fracture surface was similar to the result of (a). (d) shows that Bi_2O_3 melted and penetrated between particles due to the large amount of Bi_2O_3, resulting in low sintering power between particles. As the brittleness is strong and cracks rapidly propagate, the compressive strength is considered to be reduced.

Fig. 4 shows the EDX analysis result of the fracture surface. (a) and (b) show "B" specimens sintered at 900 °C and 1,100 °C, respectively, and (c) and (d) respectively show "D" specimens and "F" specimens at 1,200 °C. (a) and (b) show "B" specimens with the same amount of Bi_2O_3. More Bi_2O_3 was detected at 1,100 °C than 900 °C. This is because Bi_2O_3 melts and covers the surface of the sintered ceramic particles. Naturally, the sintering material of high temperature detected a lot of oxygen. (c) and (d) were sintered at the same temperature, but the amount of Bi_2O_3 was different. (c) indicates that it is subject to the same factors as (b), but since (d) measured the quantity of the surface, it is judged that not all of the addition amount was detected. Therefore, the results shown in Figs. 1 to 3 indicate that the addition of Bi_2O_3 can lower the sintering temperature, but the optimal amount of additive is considered to be around 3.4 wt.%.

This study proved the lower sintering temperature at 1,100 °C or 1,200 °C can achieve similar performance by adding Bi_2O_3 to Al_2O_3 compared with Al_2O_3 sintered at 1,600 °C in the previous study [6]. By decreasing the sintering temperature, it is possible to reduce the time for sintering which lead to energy saving. It was calculated that the sintering temperature at 1,100 °C or
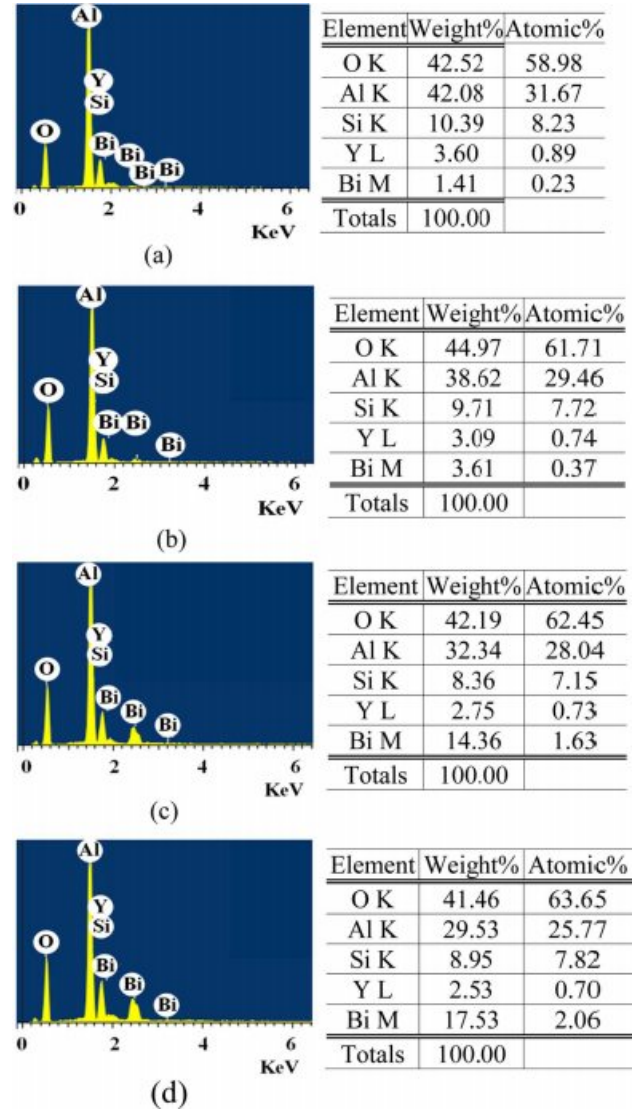

Element	Weight%	Atomic%
O K	42.52	58.98
Al K	42.08	31.67
Si K	10.39	8.23
Y L	3.60	0.89
Bi M	1.41	0.23
Totals	100.00	

(a)

Element	Weight%	Atomic%
O K	44.97	61.71
Al K	38.62	29.46
Si K	9.71	7.72
Y L	3.09	0.74
Bi M	3.61	0.37
Totals	100.00	

(b)

Element	Weight%	Atomic%
O K	42.19	62.45
Al K	32.34	28.04
Si K	8.36	7.15
Y L	2.75	0.73
Bi M	14.36	1.63
Totals	100.00	

(c)

Element	Weight%	Atomic%
O K	41.46	63.65
Al K	29.53	25.77
Si K	8.95	7.82
Y L	2.53	0.70
Bi M	17.53	2.06
Totals	100.00	

(d)

Fig. 4. EDX analysis of fracture surface. (a) "B" specimen sintered at 900 °C, (b) "B" specimen sintered at 1,100 °C, (c) "D" specimen sintered at 1,200 °C, and (d) "F" specimen sintered at 1,200 °C.

1,200 °C can save energy use by 9.1% and 4.5% respectively compared with that of 1,600 °C.

Conclusions

In this study, Bi_2O_3 was added to the sintering of Al_2O_3 ceramics to evaluate the sintering characteristics according to the addition amount. The obtained results are as follows.

As the sintering temperature increases, the compressive strength and relative density of the sintering material tend to increase, but the compressive strength is decreased when more than 15 wt.% of Bi_2O_3 is added.

The specimens containing more than 15 wt.% of Bi_2O_3 shows decreased compressive strength and relative density when sintered at 1,200 °C.

It is judged that the compressive strength of 1,100 °C

and 1,200 $^{\circ}$C is excellent by adding 3.4~10.5 wt.% of Bi_2O_3. The optimal condition was strictly obtained from 3.4 wt.% of Bi_2O_3 at 1,100 $^{\circ}$C. The addition of Bi_2O_3 can lower the sintering temperature of alumina by about 400 $^{\circ}$C.

Sintering process of Bi_2O_3-added Al_2O_3 at 1,100 $^{\circ}$C and 1,200 $^{\circ}$C can reduced energy use by 9.1% and 4.5% respectively compared with that of sintered Al_2O_3 at 1,600 $^{\circ}$C. Ceramic structures that can be low temperature sintering can save costs due to reduced manufacturing costs, replacement costs, shutdowns, etc., which will ensure enormous economic efficiency.

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