Journal of Ceramic Processing Research. Vol. 2, No. 2, pp. 87~91 (2001)

JOURNAL OF
Ceramic Processing Research

Characteristics of PLPP and infiltration of attrition-milled waste-Al_2O_3 substrates with cullet

Jeong-Hyun Park*, Myung-Ick Hwang, Kyu-Ho Song and Chul-Ku Cho[a]
Dept. of Ceramic Eng., Yonsei University
[a]Ceramic Materials Research Institute, Hanyang University

In order to recycle Al_2O_3 substrates from waste ceramics, the Al_2O_3 substrate was crushed by attrition and an alumina compact with cullet was made by infiltration. Another alumina compact with cullet was made by PLPP (pressureless powder packing forming method) and firing. During crushing, the particle size depended on speed and time of attrition. Crushing efficiency decreased with time. To confirm the behavior of cullet infiltration using the crushed powder, an infiltration device was prepared. The use of cullet as a flux proved to be difficult at temperature in the range of 1100~1300°C because of it's high viscosity. Therefore, borax was added and the infiltration distance increased many dozens times at over 15 wt.% of borax and 1150°C, and alumina compact was prepared by a mixing preform method and PLPP. After making an alumina compact, densification behaviors and characteristics were observed at a sintering temperature of 1150°C. For PLPP, the binder solution of 4 wt.% PVA solution was infiltrated into the packed powder to obtain a suitable strength of compacts, and silicon rubber was used as a mold because of its good releasing property. For the alumina compact formed with cullet by PLPP at 1150°C-2hr, the structure of the alumina bonded by an amorphous phase was observed and as the content of cullet increased in the alumina compact, so the porosity decreased. Addition of 10~40 wt.% of cullet gave 30~40% of the porosity.

Key words : Al_2O_3 substrate, preform, PLPP, flux, cullet, Borax.

Introduction

There are various ceramic industries within a country and ceramic waste is becoming a growing problem to industry and the environment. Most ceramic wastes from industries have been disposed of by means of burying not withstanding the high costs. Among the wastes, ceramic substrates make up a high portion dumping into the ground. Most of these substrates are fabricated from alumina. Because of aluminas excellent electrical and mechanical characteristics, it is widely used as substrates for semiconductor and other electrical device fabrications. Waste-alumina substrates have only about 4 wt.% flux as impurities. Also the sharp edge form after milling will enable new applications. For example, waste alumina can be used in refractories, sound proof materials, and so on. Therefore, the utilization of waste substrates will help prevent environmental contamination. In addition, using recycled products will reduce costs. The composition, particle size and shape of starting powder in fabrications of alumina ceramics are important. For the control of particle size and shape attrition milling was used in this experiment because of its high efficiency and low operation time.

*Corresponding author:
Tel : +82-2-2123-2848
Fax: +82-2-2123-2848
E-mail: sinter@yonsei.ac.kr

The PLPP (Pressureless Powder Packing) forming method is the following; powders were packed into the mold by mechanical vibration without any pressing, the binder solution was infiltrated into the packed powder and after drying, a green body was obtained. In this method, the capillary produced by pores between packed powders was the driving force for binder infiltration, and, the PLPP process was capable of demolding the green body easily and forming a complex shape.

In this study, alumina was milled by an attrition milling machine, and the milling efficiency was investigated. Then alumina powders were mixed with borax and waste-cullet were applied to the PLPP process. The Infiltration behavior of cullet and PLPP properties was investigated.

Experiments

Figure 1 shows the overall process. Waste-alumina substrates and cullet were used as starting materials. A cullet was waste glass from a glass factories. They are milled by attrition mill (KMD-1B, Korea). The milling media were alumina balls (φ3 mm, total weight: 1 kg). The milling velocity was controlled to 500, 600, 700 and 800 rpm. Milling time was 1, 2 and 3 hours. The milled powders were sieved by 25 mesh (210 μm) sieve. Also after 5 and 10 hour at 800 rpm, efficiencies as a function of holding time were observed. The particle size was observed by means of particle size

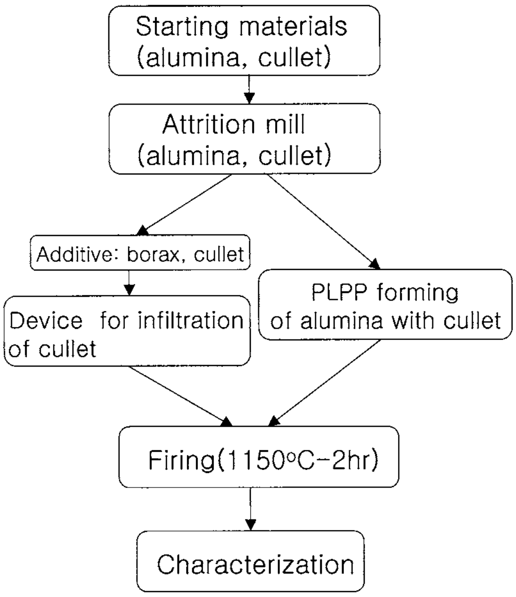

Fig. 1. Flow chart of overall process.

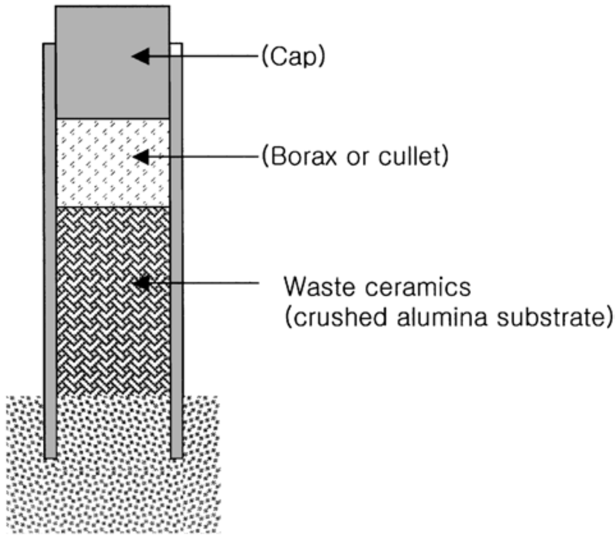

Fig. 2. Schematic illustrating of infiltration device.

analysis (PAR-III, Otsuka, Japan). The phase components of the starting materials were measured by XRD (Rint-2000, Rigaku, Japan). The milled alumina powders were sieved between 25 mesh~10 mesh sieve. The infiltration length of cullet and borax in alumina were observed as a function of content and firing temperature. Figure 2 shows the infiltration device. Also alumina with cullet and borax were formed by a PLPP forming method. Figure 3 shows the PLPP Process. The powder compacts were sintered at 1150°C-2 hour. The heating rate was 4°C/min. The structures of sintered alumina were observed by optical microscopy.

Results and Discussions

Figure 4 shows the XRD results from waste alumina substrates. Any alumina peaks were observed, so the

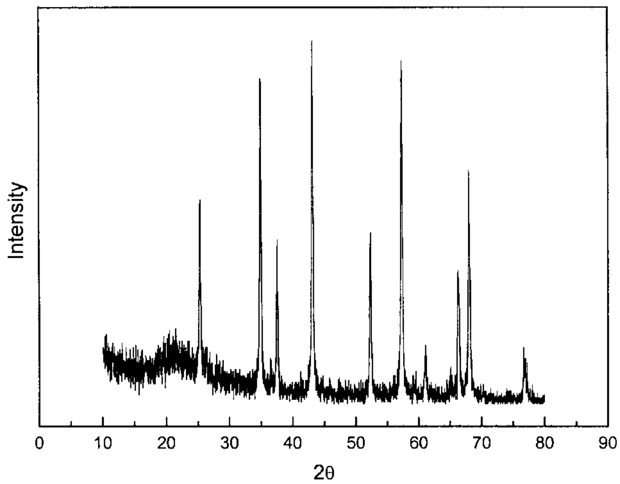

Fig. 4. XRD patterns of starting material.

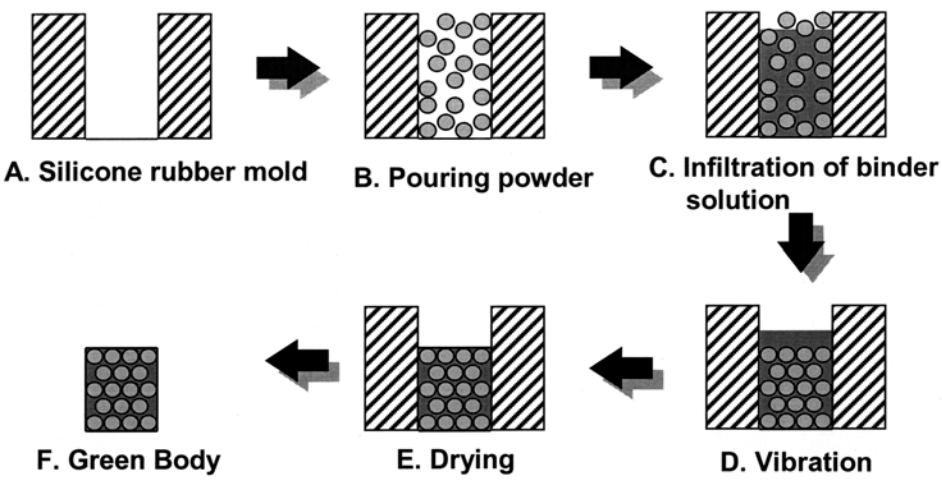

Fig. 3. Schematic illustrating pressureless powder packing (PLPP) forming method.

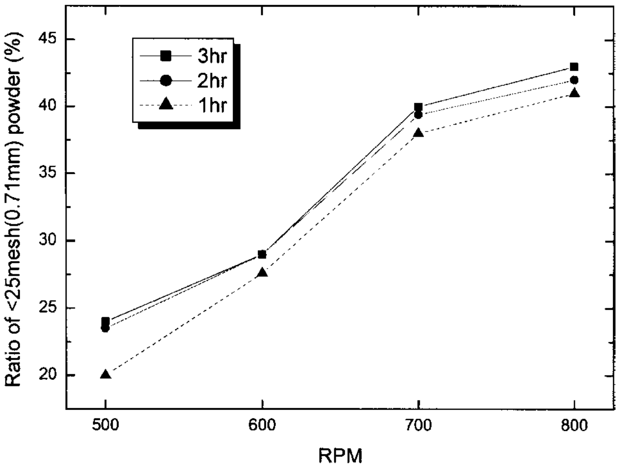

Fig. 5. Ratio of powder smaller than 25 mesh to total powder as a function of rpm and milling time.

alumina substrates were high purity. After attrition milling, milling efficiencies are shown in Figure 5. Particle sizes depended on rpm. In the range 500~700 rpm, particles amounts of sizes less than 25 mesh increased rapidly, but In the range 700~800 rpm, they have a slowly increasing pattern. The efficiencies as a function of time showed that increasing the time gave higher efficiencies. In detail, Fig. 6. shows the milling rate at 800 rpm after 1, 2, 3, 5 and 10 hour. Over 5 hour, the efficiencies incresed to 50% linearly, but after 5 hour, they increased very slowly. Therefore the dependence of rpm was larger than that of time. After attrition milling, the particle size of powder was observed. That results showed that powders mostly have particles larger than 20 μm. Figure 7 shows SEM photographs of milled powder. When the substrates of square shape were milled, square shape and irregular

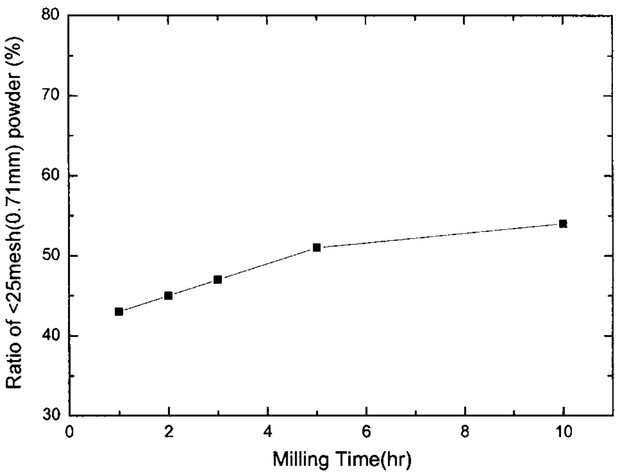

Fig. 6. Ratio of powder smaller than 25 mesh to total powder as a function of milling time at 800 rpm.

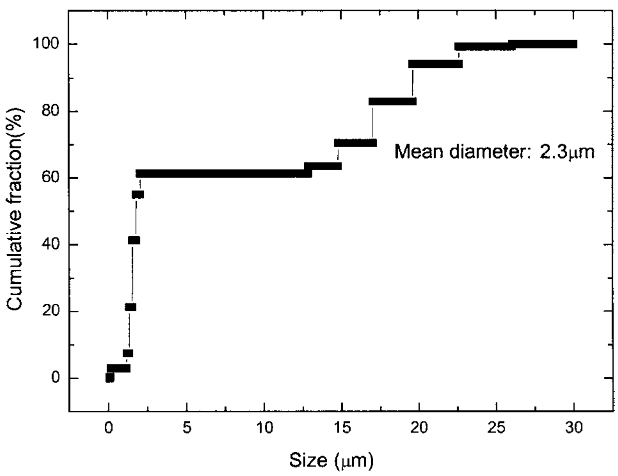

Fig. 8. Particle size distribution of cullet after attrition milling at 500 rpm=0.5 hr.

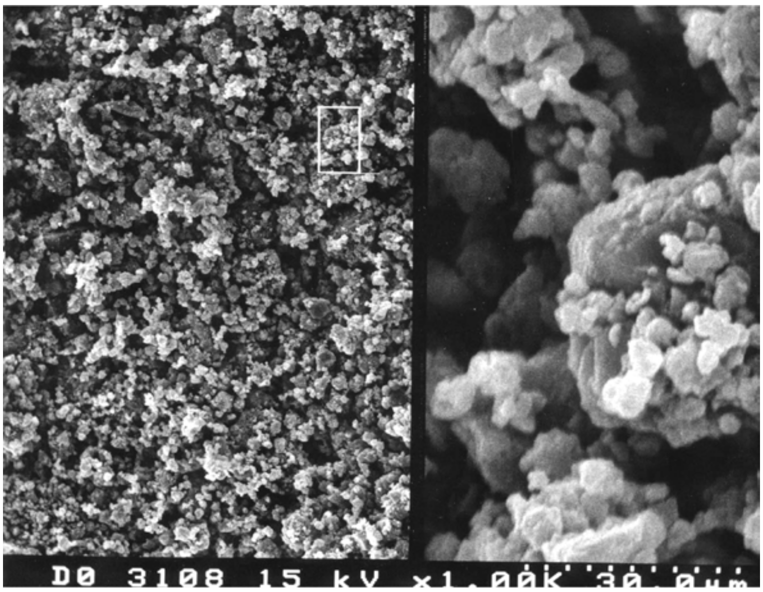

Fig. 7. SEM photograph alumina powder after attrition milling at 800 rpm-10 hr (<25 mesh).

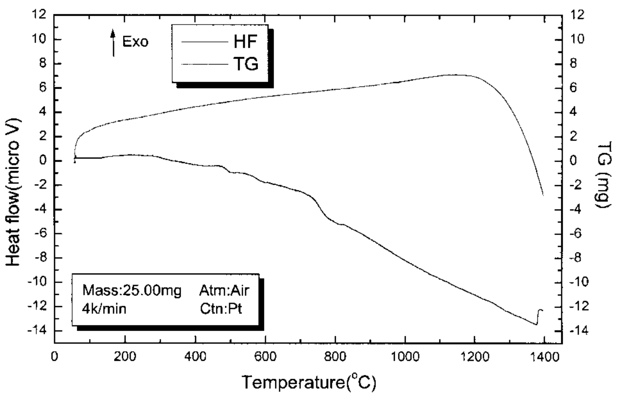

Fig. 9. DTA graph of cullet powder.

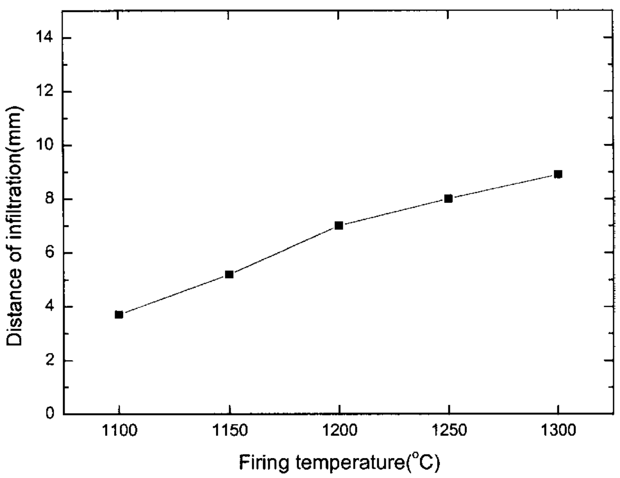

Fig. 10. Distance of infiltration of cullet as a function of firing temperature.

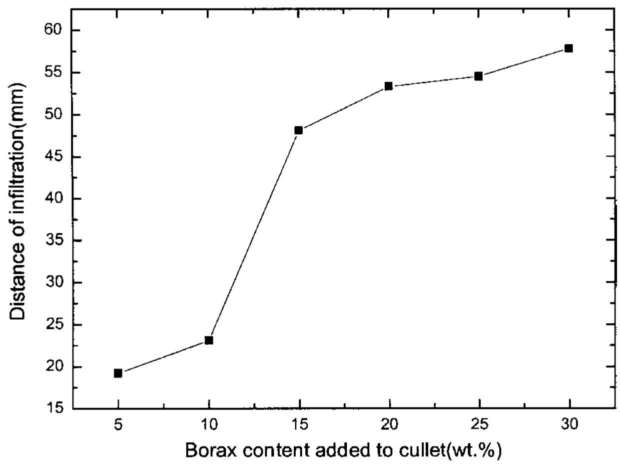

Fig. 11. Distance of flux infiltration as a function of borax contents added to cullet at 1150°C-2hr.

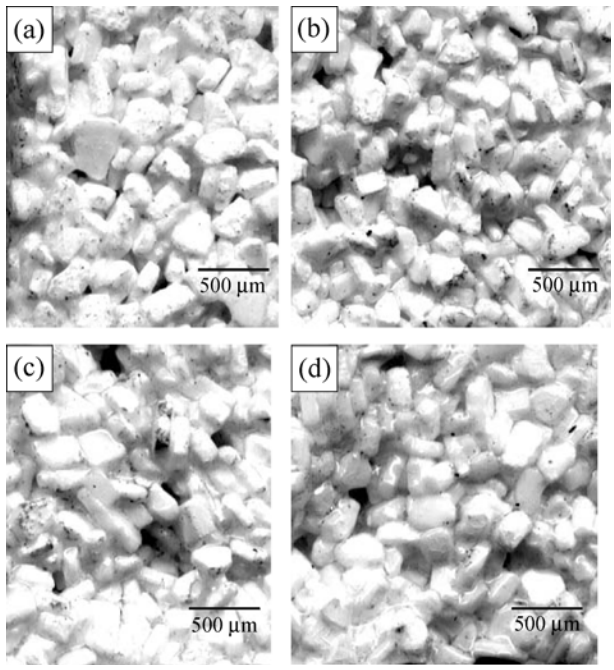

Fig. 12. Structures of PLPP specimens fired at 1150°C-2hr by PLPP method as a function of cullet content (x3); (a) 10 wt.%, (b) 20 wt.%, (c) 30 wt.% and (d) 40 wt.%.

shape particles were observed. Because the particles with a square shape slid of each other, a relatively larger number of particle of square shape remained. The cullet used as a flux in the sintering was milled by an attrition mill at 500 rpm for 30 min. Figure 8 shows the particle size analysis. The mean particle size was 2.3 µm.

When the infiltration device shown in Fig. 2 was fired at 1150°C, the cullet became a glass phase. However, its viscosity was so high that the cullet infiltrated very little. Figure 9 shows the DTA curve of cullet. A specific peak was not found, so previous the cullet reached its melting point without a rapid phase transition. Therefore the relationship between infiltration length and viscosity was measured. Some results are given in Fig. 10. Infiltration length of the cullet was as short as 4 mm. That was due to the high viscosity of the cullet. So, instead of cullet, borax with a low viscosity was used. The contents of added borax were 5, 10, 15, 20, 25 and 30 wt.%. Figure 11 shows infiltration lengths of borax into alumina at 1150°C for 2 hour. For low contents of borax, these lengths were short. But in contents of more than 15 wt.%, rapid infiltration occurred.

Alumina powders with a particle size milled economically and easily, 25 mesh~10 mesh were applied to the PLPP forming process, and various contents of fluxs were added. Then these powders were packed by a mechanical vibrator, and a polymer binder was infiltrated into the alumina powder in the mold. The binder was a PVA (poly vinyl alcohol) solution. The higher the concentration of PVA was, the higher strength of the compact was. But when PVA with concentration higher than 4 wt.% was used, infiltration almost did not happen. Mold materials were silicone rubber which has excellent separating property [7]. For low temperature sintering of alumina compact formed

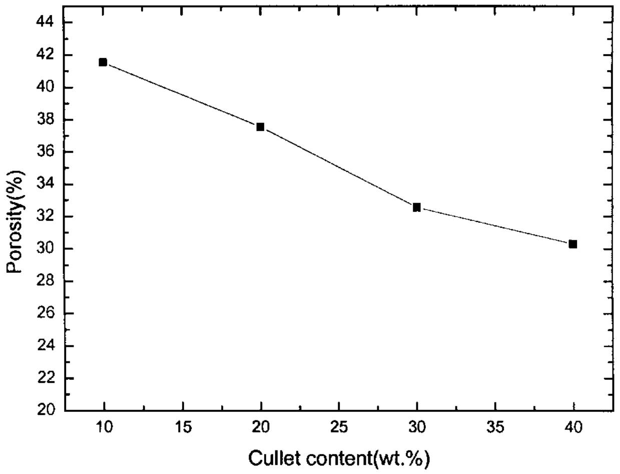

Fig. 13. Porosity of Pressureless Powder Packing (PLPP) specimens fired at 1150°C-2hr as a function of cullet contents.

by PLPP, cullet and borax were used as the flux. But when the PVA solution was infiltrated for PLPP, borax reacted with PVA solution and it changed into a gel. These borax could not be applied in the PLPP method. After sintering, alumina specimens with cullet are shown in Fig. 12. As cullet reacts with the flux, a more dense structure was seen in specimens with higher cullet contents. Figure 13 shows the porosity as a function of cullet content. The higher was the contents of cullet, the lower was the porosity.

Conclusions

To confirm the usage of recycled waste Al_2O_3 substrates, a preform into which cullet was infiltrated and a sample fabricated by PLPP were heat-treated with the following results.

1. The crushing efficiency of using an attrition mill depended on the rpm and the time of attrition. When it was crushed for more 2 hours, the crushing efficiency decreased. In the range 500~800 rpm, the crushing efficiency increased clearly with the crushing speed. The crushing efficiency increment decreased at 800 rpm and it depended on the speed of crushing rather than the time of crushing. Most of the particles didn't retain a square shape.

2. The cullet was used to achieve a suitable strength for the alumina. As a result, it was difficult to infiltrate because of the high viscosity of cullet. So borax was added because of its low viscosity. The infiltration length was dramatically improved at over 15 wt.% borax. And ceramics of suitable strength could be obtained.

3. The alumina particle of 25~10 mesh and cullet of several micrometers were mixed together, and this was formed by a PLPP method. Infiltrating the binding solution of 4 wt.% of PVA into packed powder resulted in obtaining a suitable forming strength and the infiltration speed. When it was heat-treated at 1150°C-2 hr, it showed decreased porosity and when 10~40 wt.% of cullet was added, 30~40 wt.% of porosity was measured.

References

1. James, F. Zievers and Paul Eggerstedt, Am. Ceram. Soc. Bull. 70[1] (1991) 108-111.
2. J. S. Reed, "Principles of Ceramics Processing", 2nd. Ed., J. Wiley & Sons, Inc. (1995) 313-337.
3. T. M. Cook and T. H. Courtney, Metall & Mater. Trans. A 26A[9] (1995) 2389-2397.
4. Richard, M., Fulrath, J. of the American Ceramic Society 42[9] (1959) 423-429.
5. David, W. Richerson, "Shape Forming Process", in Modern Ceramic Engineering, 2nd ed., Marcel dekker, ind. (1990) 148-518.
6. H. Thurnauer, "Controled Required and Problems Encounded in Production Dry Pressing", in Ceramic Fabrication Process, edited by W.D. Kingery, MIT press, Cambridge, Mass. (1963) 62-70.