Influence of the Internal Current on the Sintering Behavior of ZnO Ceramics Sintered by PCS Method

Tatsuya Misawa[1,a], Noboru Shikatani[1,b], Yuji Kawakami[2,c], Takashi Enjoji[2,d], Yasunori Ohtsu[1,e] and Hiroharu Fujita[1,f]

[1]Department of Electrical and Electronic Engineering, Faculty of Science and Engineering, Saga University, 1 Honjo-machi, Saga-shi, 840-8502 JAPAN.
[2]Department of Material and Environment, Industrial Technology Center of Saga, 114 Yaemizo, Nabeshima-machi, Saga-shi, 849-0932 JAPAN.
[a]misawa@ep.ee.saga-u.ac.jp, [b]sikatani@emat.fit.ac.jp, [c]kawakami@saga-itc.go.jp, [d]enjoji@saga-itc.go.jp, [e]ohtsu@ep.ee.saga-u.ac.jp, [f]fujita@ep.ee.saga-u.ac.jp

Abstract

The influence of the internal current for the ZnO ceramics on the sintering behavior by pulse current sintering (PCS) method was investigated. To clear the dependence of inner current on the sintering behavior of ZnO ceramics, direct measurement of electric resistance of ZnO specimen under sintering by SPS device was carried out. It was observed that electric resistance of specimen decreases with increase in the temperature. The electric resistance begins to decrease from the low temperature of 200 ℃. The internal structure of sintered ZnO ceramics changed by the control of the internal current in the specimen using Al_2O_3 plate.

Keywords : pulse current sintering method, ZnO ceramics, thermoelectric material

1. Introduction

Pulsed current sintering (PCS) method, which can produce the various materials in short time and high quality, is expected as the future manufacturing method of sintered materials and ceramics[1-5]. The sintering using the PCS method finish in the short time as several decade minutes or several hours, while the general sintering using the atmospheric oven needs several hours or a few days.

In the PCS method, the pulsed large current of several kA flows in the highly compressed carbon mold which enclosed the sample, and heats the specimen by Joule heating. In case of the electrical conducting material, it is considered that the part of the sintering current flows inside of specimen, and strongly affects the sintering behavior.

The electric resistance of the ZnO ceramics containing the small Al_2O_3 decreases in dependence on the sintering extent, so it is also anticipated that the Ohmic heating by the current flow in the sample strongly contributes to the desirable sintering on the ZnO materials. We are aiming at the clarifying of the influence of the internal current on the sintering behavior of ZnO ceramics by PCS method. Then, the direct measurement of electric resistance of ZnO specimen under PCS sintering and SEM observation of sintered specimen was carried out in order to clarify the current effect for the sintering behavior.

2. Experimental Setup

The experiments were carried out using the PCS device (SPS-3.20MK-IV, SPS Syntex Inc.). The carbon graphite mold which enclosed the specimen is installed in the experimental device, and applies the pressure of several decade MPa under the low gas pressure(Fig.1(a)). The pulsed current is applied to the carbon mold, and the specimen is heated by Ohmic heating of carbon mold. Typical experimental setup are Compressing pressure P_{press}~ 30MPa, gas pressure P_{gas}~ 20Pa, maximum temperature T_{max}~1100°C, maximum sintering current I_s~3kA. The temperature of carbon mold was made to rise to 1100℃ in 18 minutes, and fixed at 1100℃ during 10 minute.

The ZnO specimen powder contain small amount of Al_2O_3, TiO_2 and CoO, and composition mol ratio is ZnO:Al_2O_3: TiO_2:CoO = 100:2:2:2.

For measuring electric resistance and inner temperature of ZnO specimen, two Pt electrodes and Pt thermocouple covered by ZrO tube have been installed in the side of the ZnO specimen through the carbon die. DC power source and carbon film resistor are being connected with 2 platinum electrodes, and electric resistance is measured from current and voltage which flow between electrodes (Fig.1(b)). Electric resistance of ZnO specimen, temperature of the specimen and carbon mold were recorded by data logger.

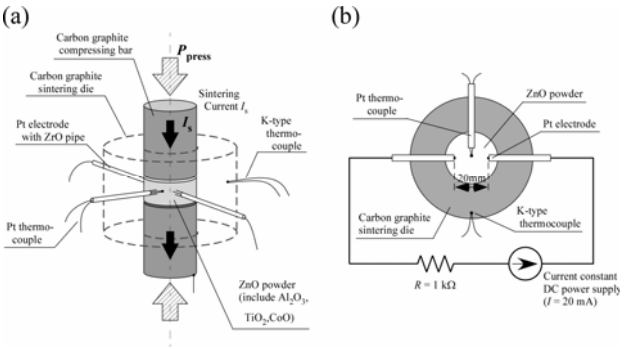

Fig. 1. Experimental setup.
(a): Carbon graphite mold and electrodes
(b): Circuit for measuring electric resistance

3. Result and Discussion

It was observed that the electric resistance of ZnO specimen decreases with the progress of the sintering (Fig.2). The dependence of inner temperature on electric resistance is different in four temperature regions. In the region of room temperature ~ 200°C (Fig.2(I)), the electric resistance is very large, and sample has not sintered. When the temperature rises over 200°C (Fig.2(II)), the electric resistance drastically decreases. Electric resistance changes about 10^{-5} times during 200°C and 350°C, and it is anticipated that the current begins to be flowing in inside of ZnO specimen in this temperature region. In the region of 350°C ~ 900°C (Fig.2(III)), the electric resistance slowly decreases with the change of the temperature. Because the specimen start to be contracting from ~ 600°C, it is considered that the sintering mainly develops in this temperature region. And over 900°C (Fig.2(IV)), it becomes that the electric resistance is almost constant. The contraction of the specimen has also finished about 1000℃. Therefore, it is considered that sintering finished.

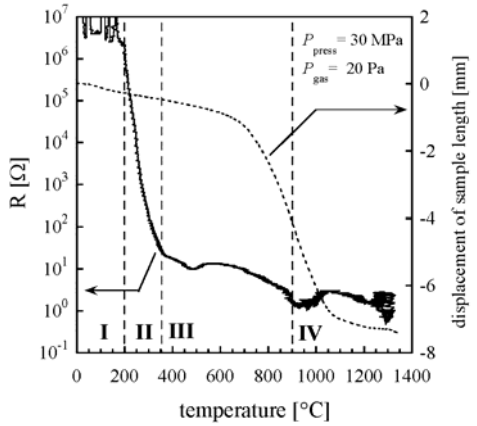

Fig. 2. Dependence of inside temperature on the electric resistance of ZnO specimen.

To clear the dependence of inner current on the sintering behavior of ZnO ceramics, sintering experiment limited the internal current by the Al_2O_3 plate was carried out. The internal structure of sintered ZnO ceramics changed by the control of the internal current. Under the current limited condition (Fig.3(a)), shape of the material powder has remained, and the sintering does not extents sufficiently.

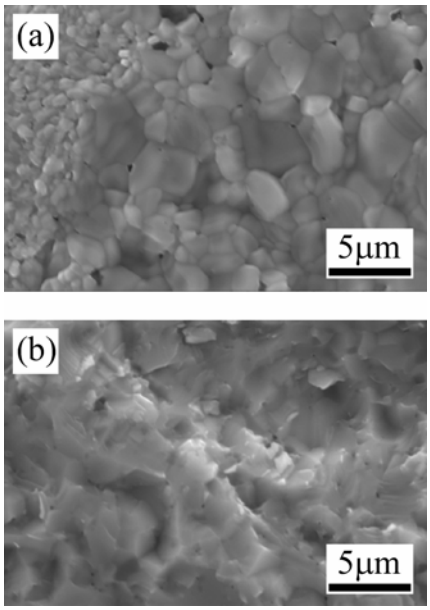

Fig. 3. Typical SEM image of cross section of sintered ZnO specimen.
(a): with Al_2O_3 plate (current limited condition)
(b): without Al_2O_3 plate

4. References

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