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Synthesis of $Ba(Mg_{1/3}Ta_{2/3})O_3$ Powder by Hydrothermal Method and Characterization of Ceramics

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The characterization of barium magnesium tantalate ($BaMg_{1/3}Ta_{2/3}O_3$;BMT) ceramics using BMT powder prepared by hydrothermal method was reported from a view point of powder processing. Single phase BMT powde was obtained at 120℃ 5h. The dielectric constant and unloaded Q($1/\tan\delta$), were 28.0 and 1,900 at 7GHz, respectively.

1. INTRODUCTION

Recently, communication system using microwave frequency is an exciting new development. It is well-known that BMT ceramics has the highest value of unloaded Q in a range of microwave frequency. Hyuga et al. showed that the Q value strongly depended on the ordering of the B site ions ; $Mg_{1/3}Ta_{2/3}$. [1] Nomura et al. measured that the Q value of BMT ceramics was 3,000 and 1mol%Mn-doped was 16,800 at 10GHz.[2]

However, studies in regarding with the effect of chemical powder processing on the BMT ceramics have not been reported yet. In this study, we prepared BMT fine particles by a hydrothermal method, and showed the characteristics of the as-prepared powder and their sintered ceramics, e.g., crystal phase, specific surface area, density, grain size, and electric properties.

2. EXPERIMENTAL PROCEDURE

Barium chloride, $BaCl_2$ (>99%), magnesium nitrate, $Mg(NO_3)_2$ (>99%), and tantalum oxide, Ta_2O_5 (99%) were used as raw materials. $BaCl_2$ aqueous solution and $Mg(NO_3)_2$ aqueous solution were mixed with various hydroxide of alkaline metals aqueous solution. (Lithium hydroxide; LiOH, Sodium hydroxide; NaOH, Potassium hydroxide; KOH), for comparison, synthesis by conventional method is called CO. After addition of Ta_2O_5 powder, the mixed solution was stirred in an autoclave at various temperature from 120 to 180℃ for 3h, and BMT powder was precipitated. Alkaline metal ions and hydroxide ions were mostly removed by repeated filtering and washing of the precipitate by deionized water. The precipitate dried at 120℃ for 20 h. The powder was pressed to pellets with a 10mm in diameter and a 10 to 15 mm in thickness. The formed pellets was sintered at 1350 to 1600℃ for 2 to 30h. The cooling rate after sintering was 100℃/h. The powder and sintering pellets of BMT were analyzed with X-ray diffractometry (XRD) to study the crystal phase and the super lattice structure. The density of sinterd pellets was measured by water immersion method and the specific surface area of powders by B.E.T.

method. Microstructures were analyzed by scanning electron microscopy.(SEM) Electrical properties were measured with a networked analyzer in a range of microwave frequency.

3. RESULTS AND DISCUSSION

X-ray diffraction patterns of the as-prepared powders by LiOH,NaOH and KOH hydrothermal method is shown in Figure 1. As shown, all of them were single phase perovskite.

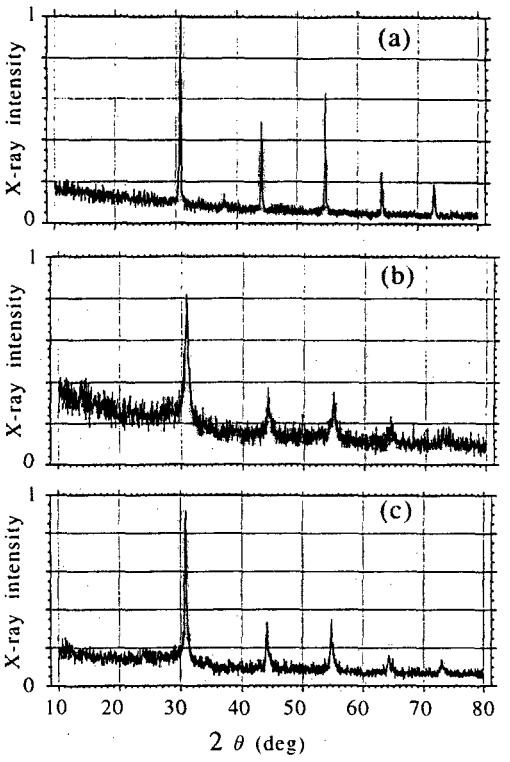

Figure 1. X-ray diffraction patterns of BMT powders by hydrothermal method. (a) LiOH (b) NaOH (c) KOH

Table 1 shows the relations among hydrothermal reaction temperature, specific surface area and the relative height of the major X-ray peak intensities. ($I/I_0 \times 100$; I_0 is the X-ray intensity in the powder reacted by 120℃) Unde rvarious experimental conditions, The specific surface area of as-prepared powder are obviously not affected by reaction temperature.However the relative height of the major X-ray peak intensity increased with increasing the reaction temperature, this is because high temperature accelerates the cristallization of powder.

Table 1. Effect of reaction temperature on specific surface area and relative height of X-ray intensity of as-prepared powders.

Reaction temp.(℃)	Specific surface area (m^2/g)	Relative height of X-ray intensity(%)
120	42	100
140	40	107
160	34	117
180	33	177

Figure 2 shows the sintering temperature dependence of density. The maximum density using hyderothermally produced powder is 7.56 g/cm^3 at 1450℃.On the other hand, conventional one is 7.58 g/cm^3 at 1600℃. The sintering temperature of the pellets using hydrothermally produced powder is low by 150℃ compared with those of conventional powder. This was mainly due to the fine particle size of the powder by hydrothermal processing.

The dependence of grain size of sintered pellets on KOH and NaOH hydrothermal processing is shown in Figure 3. Both grain size of sintered pellets increases with sintering temperature due to the growth of particles. With the same sintering temperature, the grain size of sintered pellets are almost equal. The kind of alkaline metal hydroxides using hydyothermal processing scarcely affects the change of grain size of sintered pellets.

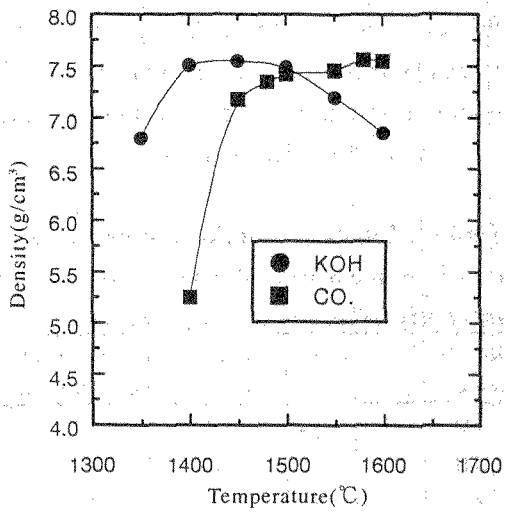

Figure 2. Sintering temperature dependence of density.

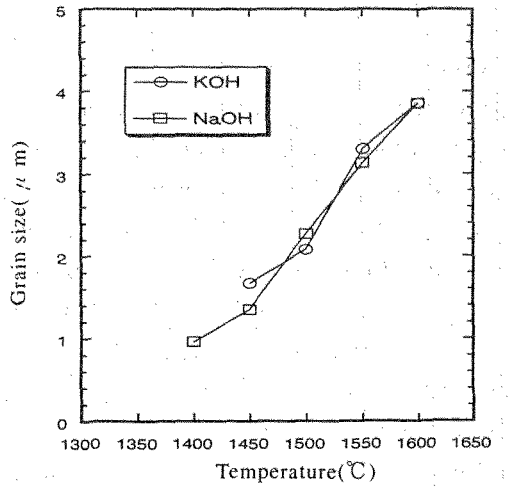

Figure 3. Sintering temperature dependence of grain size of BMT ceramics by hydrothermal processing.

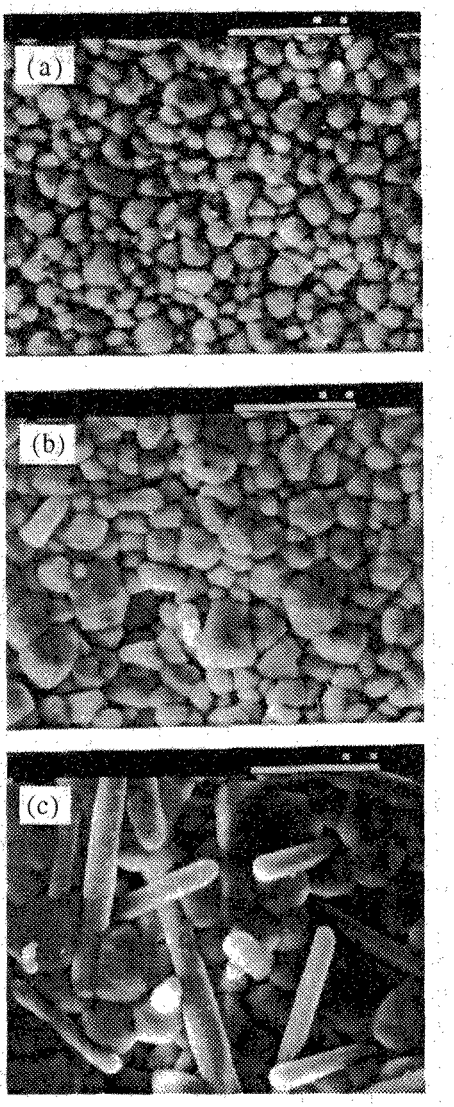

Figure 4 Scanning electron micrograph of BMT ceramics at various sintering temperature (a) 1450℃ (b) 1500℃ (c) 1600℃. (bar=10 μ m)

Figure 4 shows the SEM photograph of the as-fired surface of BMT ceramics using hydrothermal produced powder with KOH sintered at 1450,1500 and 1600 ℃. Microstructure at 1450 and 1500℃ is more dense than that at 1600℃. This agreed with the change of density of sintered pellets, as shown in Figure 2. The crystal form with needles appeared at 1600℃. It seem that this form is the abnormal grain growth of ultrafine powder. Figure 5 shows the sintering

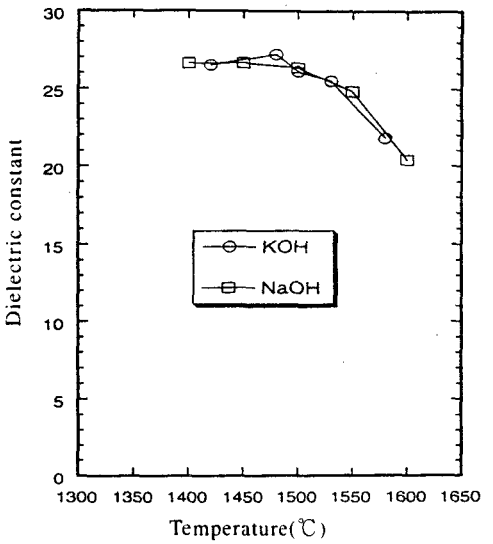

Figure 5. Sintering temperature dependence of dielectric constant, where powder were prepared by KOH and NaOH hydrothermal reaction.

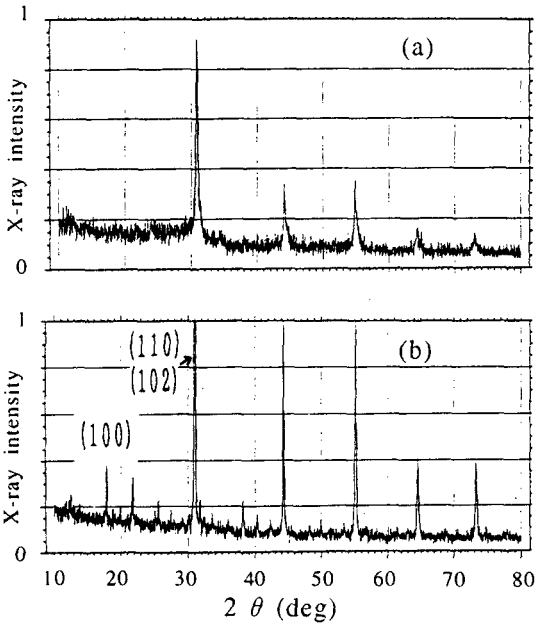

Figure 6. X-ray diffraction patterns of before and after sintering. (a) as-prepared powder (b) sintered pellet at 1480℃ for 8h.

temperature dependence of dielectric constant with KOH and NaOH. Dielectric constant decreased at sintering temperature above 1500 ℃. This also agreed with the decrease of density at high sintering temperature because of grain growth with needles partices. Maximum dielectric constant and unloade Q, were 28.0 and 1,900 at 7GHz, respectively.

Figure 6 shows the X-ray diffraction patterns sythesized powder and after sintering ceramics body. Super lattice structure appears in XRD pattern of after sintering at 1480℃ for 8h. This result was similar to the case of using conventional produced powder, however, we could not solve the relationship between the degree of ordering structure and dielectric properties.

4 .CONCLUSION

(1) Single phase perovskite BMT powder obtained at 120℃ for 5h by hydrothermal method.
(2) Properties of powder and ceramics were not affected by the kind of alkaline metal hydroxides in hydrothermal reaction.
(3) The sintering temperature of the pellet prepared by hydyothermal method is lower by 150 ℃ than that by conventional powder.
(4) Maximum dielectric constant and unloade Q , were 28.0 and 1,900 at 7GHz, respectively.

REFERENCES

1. T. Hyuga, K. Matsumoto and H. Ichimura, J. of the Ins. Electronics, Information and Communication Eng., CPM86-32 (1986).
2. S. Nomura, K. Toyama and K. Kaneta, Jpn. J. Appl. Phys., 21, L624-626 (1982)