Synthesis and Electric Properties of M₂BiNb₅O₁₅ (M = Na, K) Tetragonal Tungsten Bronze Type Oxides

IWAI Yutaka and TAKUCHI Hisao

Department of Materials Engineering, Nagaoka National College of Technology, 888 Nishikatakai-machi,Nagaoka-shi 940-8532, Japan Fax: 81-258-34-9700, e-mail: iwai@nagaoka-ct.ac.jp

Oxide system with nominal composition of $M_2BiNb_5O_{15}$ (M=Na, K) were studied. $(K_{1-x}Na_x)_2BiNb_5O_{15}$ ($0.8 \ge x \ge 0$) show the tetragonal tungsten bronze(TTB) type structure. Dielectric property was investigated in these compounds. Key words: tetragonal tungsten bronze structure, ferroelectric property, $(K_{1-x}Na_x)_2BiNb_5O_{15}$

1. INTRODUCTION

Needs for Pb-free electric materials are growing from the concern about environmental protection. Pb($Zr_{1,x}Ti_x$)O₃ (PZT) extensively used as outstanding ferroelectric and piezoelectric material cannot become an exception, and the Pb-free material which can be replaced with these in the future is called for. Many of Pb-free ferroelectric materials having tetragonal tungsten bronze structure were known. For example, K₂BiNb₅O₁₅ has been studied by several authors[1,2]. In the present research, we investigate synthesis of Na containing system (K_{1-x}Na_x)₂BiNb₅O₁₅ and these electric property.

2. EXPERIMENTAL

Potassium carbonate (purity 99.9%), sodium carbonate (purity 99.9%), bismuth oxide (99.99%) and niobium oxide (99.9%) were used as raw materials. They were mixed thoroughly and calcined at 850° C for 5 hrs. They were pressed into pellets and were heated at 1150°C for 20h in air. Measurement of dielectric constant was carried out at the temperature from 25°C to 750 °C by a LCR mater (HP4284A). Phases were identified from X-ray powder diffraction data. Crystal structure analysis was carried out by a pattern fitting method.

3. RESULTS AND DISCUSSIONS

Tetragonal tungsten bronze (TTB) phase wwa obtained in a composition range of $0.8 \ge x \ge 0$ in $(K_{1-x}Na_x)_2BiNb_5O_{15}$. The relation between temperature and the dielectric constants in $(K_{1-x}Na_x)_2BiNb_5O_{15}$ are shown in Fig.1. $K_2BiNb_5O_{15}$ shows the maximum value of the dielectric constant ($\varepsilon_r = 1148$) at 345°C and confirm the Ismalzade's result [1]. Such maximums are also observed in $(K_{0.6}Na_{0.4})_2BiNb_5O_{15}$ and $(K_{0.2}Na_{0.8})_2BiNb_5O_{15}$ at about 55°C. But, it is in an examination stage about the relation of a Curie point. Clear temperature dependency was not observed about the permittivity of x = 0.6 composition sample.

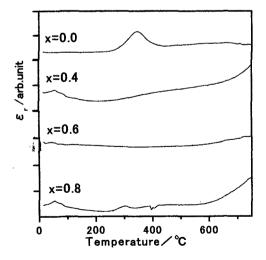


Fig.1 Temperature dependence of dielectric constants in $(K_{1,x}Na_x)_2BiNb_5O_{15}$

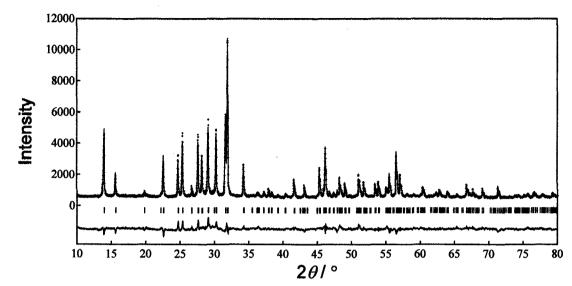


Fig.2 X-ray powder diffraction pattern of K₂BiNb₅O₁₅

Crystal structure of $K_2BiNb_5O_{15}$ was analyzed using RIETAN program [3] (Fig.2). It became clear that $K_2BiNb_5O_{15}$ showed the tetragonal tungsten bronze structure where bismuth atoms were occupied by in A1 position and potassium atoms were occupied in A2 position, respectively, shown in Fig.3.

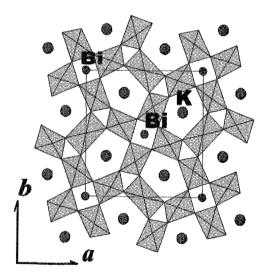


Fig.3 Crystal structure of K2BiNb5O15

4. SUMMARY

Synthesis and characterization of the oxide system $(K_{1,x}Na_x)_2BiNb_5O_{15}$ was performed. This investigation was summarized followingly.

- (1) TTB phase was obtained in the composition range of $0.8 \ge x \ge 0$.
- (2) The Maximum value of the dielectric constant at 345° C was observed in K₂BiNb₅O₁₅.
- (3) Crystal structure of K₂BiNb₅O₁₅ was analyzed and the tetragonal tungsten bronze type structure of the compound was confirmed.

REFERENCES

[1] I. G. Ismalzade: Sov Phys Crystallography, 8, 274 (1963)

[2] T. Sugai and M. Wada: Jpn. J. Appl. Phys. 11(2972) 1863

[3] F. Izumi, "The Rietveld Method", Ed. by R. A. Young, Oxford University press, Oxford (1993) pp. 121-24.

(Received October 11, 2003; Accepted March 10, 2004)