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Structural, dielectric and electrical properties of $NaBa_2X_5O_{15}$ (X=Nb and Ta) ceramics

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Abstract

Polycrystalline samples of $NaBa_2X_5O_{15}$ (X=Nb and Ta), i.e., $NaBa_2Nb_5O_{15}$ (NBN) and $NaBa_2Ta_5O_{15}$ (NBT), members of tungsten bronze family, were prepared using a high-temperature solid-state reaction technique. A preliminary structural (XRD) analysis of these compounds shows the formation of a single-phase orthorhombic structure at room temperature. The scanning electron microscopy (SEM) provides information on the quality of the samples and uniform grain distribution over the surface of the sample. Detailed studies of the dielectric properties suggest that they have undergone a phase transition well above the room temperature. Measurement of electrical conductivity (dc)/resistivity as a function of temperature suggests that compounds have semiconducting properties above the room temperature with negative temperature coefficient of resistance (NTCR) behavior.
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Keywords: Solid-state reaction; TB structure; XRD; Microstructure; Dielectric; NTCR

1. Introduction

The discovery of ferroelectricity in $BaTiO_3$ [1,2] has attracted the attention of a large number of researchers to study ferroelectric properties of various oxides of different structural families (i.e., perovskites, tungsten bronze, pyrochlore, etc.) for device applications. Some ferroelectric oxides (usually called as electronic materials) with high ε, high Q-value, good stability of temperature coefficient of resonant frequency and low dielectric loss have extensively been studied because of their applications in discrete as well as multilayer capacitor [3–7] and microwave telecommunication system [8,9].

Among many ferroelectric oxides studied so far, some compounds with tungsten bronze (TB) structure such as barium sodium niobates (BNN), potassium lanthalum niobates (KLN) [10–13] etc. have been found to be very important because of their immense applications for solid-state devices. The TB-type compounds consist of a complex array of BO_6 octahedral sharing corners in such a way that a wide variety of cations can be accommodated in any or all different interstices (i.e., A, B and C) of a general formula $(A_1)_2(A_2)_4(C)_4(B_1)_2(B_2)_8O_{30}$,where A type of cations can be accommodated in any or all different interstices A_1 and A_2. The B types of cations are substituted at the octahedral sites B_1 and B_2 [14]. The A_1 and A_2 sites can be filled by monovalent cations, whereas B_1 and B_2 sites can be filled by tri to pentavalent cations and C site, being small, often remains vacant or may be filled by mono or divalent cations. The distribution of metal cations at different atomic sites of TB structures plays a significant role to tailor the physical properties of the materials for device applications. The extensive literature survey on TB structure reveals that though some interesting work on the binary system $NaNbO_3$-$BaNb_2O_6$ have been reported by Burns et al. [19], no work have

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been reported on the titled compounds $NaBa_2X_5O_{15}$ (X=Nb and Ta). Therefore, we have extensively studied structural, dielectric and electrical properties of these compounds.

2. Experimental

Polycrystalline samples of $NaBa_2Nb_5O_{15}$ (NBN) and $NaBa_2Ta_5O_{15}$ (NBT) were prepared by a high-temperature solid-state reaction technique using high purity ingredients; Na_2CO_3 (99.9%, M/S. Sarabhai Chemicals, India), $BaCO_3$ (99.9%, M/S. Sarabhai Chemicals, India), Nb_2O_5 (99.9%, M/S. BARC, India) and Ta_2O_5 (99.9%, M/S. E. Merck, Germany) in a suitable stoichiometry. The ingredient oxides and carbonates were mixed thoroughly; first in an air atmosphere for 1 h and then in alcohol (i.e., methanol) for 1 h so as to make a homogeneous fine powder. Then the powder was calcined in an alumina crucible at 1223 K for 12 h. The process of grinding and calcination was repeated several times until the desired material was obtained. The calcined fine powder was cold pressed into cylindrical pellets of 10 mm diameter and 1–2 mm of thickness at a pressure of 4×10^6 N/M^2 using a hydraulic press. Polyvinyl alcohol (PVA) was used as binder to reduce the brittleness of the pellets. The binder was burnt out during high temperature sintering. The pellets were sintered at 1523 K for 15 h in an air atmosphere using alumina crucibles. The formation and quality of the compounds were checked by X-ray diffraction (XRD) technique. The X-ray diffraction pattern of the compounds was recorded at room temperature using an X-ray powder diffractometer (PHILIPS, PW-1710) with CrKα radiation (λ=2.2909°A) in a wide range of Bragg's angles 2θ ($20° \leq \theta \leq 80°$) with a scanning rate of 3° per minute.

The surface morphology of the sintered pellets was studied at room temperature by a scanning electron microscopy (SEM) (JEOL-JSM-5800). To measure the electrical properties of the compounds, air-drying silver paints was applied on both flat surfaces of the pellets to serve as electrodes. Electrical impedance (Z), phase angle (θ), capacitance (C_p) and dielectric loss (D) were measured as a function of frequency (10 kHz–1 MHz) at different temperatures (323 K–773 K) using HIOKI 3532 LCR Hi-TESTER in conjunction with a laboratory-made sample holder and heating arrangement with an ac signal (voltage 1.3 V). A chromel–alumel thermocouple and digital multimeter (M/S Electronic of India, DM 6108) were used to measure the temperature. All the measurements were recorded within a small temperature interval (≈2 °C). The DC conductivity of the compounds was measured using a Keithley-617 programmable electrometer and a laboratory-fabricated experimental set up and heating arrangement. To overcome the effect of moisture, if any on electrical properties, the samples were preheated to 423 K for 4 h

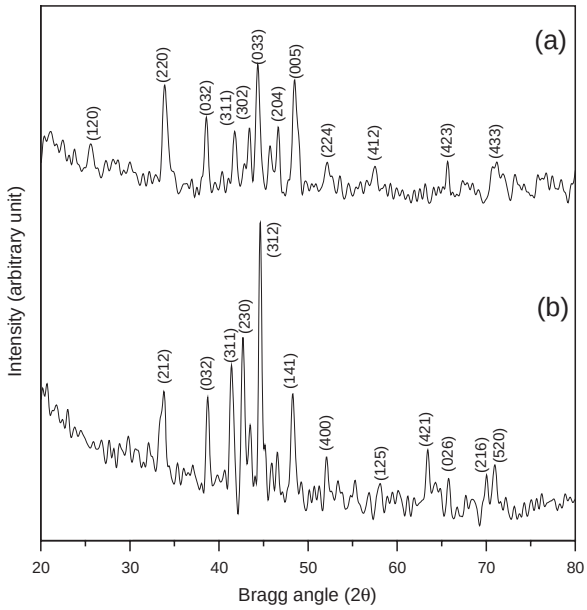

Fig. 1. (a) Comparison of room temperature X-ray diffraction profiles of (a) $NaBa_2Ta_5O_{15}$ (NBT) and (b) $NaBa_2Nb_5O_{15}$ (NBN) compounds.

Table 1
Comparison of observed and calculated d-values (°A) of some reflections for $NaBa_2Nb_5O_{15}$ and $NaBa_2Ta_5O_{15}$ compounds at room temperature

$NaBa_2Nb_5O_{15}$					$NaBa_2Ta_5O_{15}$				
h	k	l	d_{obs}	d_{cal}	h	k	l	d_{obs}	d_{cal}
2	1	2	3.9403 (48)	3.9500	1	2	0	5.1901 (62)	5.1872
0	3	2	3.4528 (47)	3.4504	2	2	0	3.9178 (81)	3.9174
3	1	1	3.2443 (58)	3.2450	0	3	2	3.4700 (84)	3.4698
2	3	0	3.1463 (59)	3.1525	3	1	1	3.2183 (73)	3.2212
3	1	2	3.0187 (100)	3.0137	3	0	2	3.0192 (77)	3.0889
1	4	1	2.8025 (55)	2.7985	0	3	3	3.0316 (99)	3.0330
4	0	0	2.6083 (32)	2.6060	2	0	4	2.8900 (91)	2.8925
1	2	5	2.4683 (26)	2.4713	0	0	5	2.7943 (100)	2.7927
4	2	1	2.3516 (24)	2.3527	2	2	4	2.6060 (61)	2.6053
0	2	6	2.1799 (34)	2.1819	4	1	2	2.3758 (57)	2.3747
2	1	6	2.1088 (28)	2.1054	4	2	3	2.1131 (68)	2.1138
5	2	0	1.9665 (27)	1.9671	4	3	3	1.9665 (58)	1.9666

The number in parenthesis is I/I_0.

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