Journal of Alloys and Compounds 505 (2010) 168–171

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

Investigations on the effect of preparation conditions on $AgNbO_3$ ceramics

Om Prakash Nautiyal[a], S.C. Bhatt[a], K.S. Bartwal[b,*]

[a] Department of Physics, HNB Garhwal University, Srinagar 246174, India
[b] Laser Materials Development & Devices Division, RRCAT, Indore 452013, India

ARTICLE INFO

Article history:
Received 5 May 2010
Accepted 30 May 2010
Available online 11 June 2010

PACS:
81.05.Mh
61.05.cp
68.37.Hk
78.20.Ci

Keywords:
Ceramics
Perovskite
XRD
Scanning electron microscopy
Dielectrics

ABSTRACT

$AgNbO_3$ ceramic was found to be one of the candidate materials for lead-free piezoelectric materials. The processing and sintering procedures have detrimental effect on the characteristics of the ceramics. Ceramic pellets of silver niobate, $AgNbO_3$ were prepared by solid-state reaction technique. To investigate the effect on the ceramic characteristics, two different sintering procedures were adopted. The crystallinity and surface morphology of the prepared samples were investigated by XRD and SEM techniques. The samples prepared by both the methods show orthorhombic structure at room temperature. Comparisons of lattice parameters, crystallite and grain sizes of $AgNbO_3$ are reported. Frequency variations of dielectric constant (K), tangent loss ($\tan\delta$) and electrical conductivity (σ) were measured at room temperature in the frequency range 1 Hz to 10 MHz.

1. Introduction

The piezoelectric ceramics have been used for various electronic products. The most used piezoelectric ceramics are lead compounds e.g. PZT due to their higher electromechanical coupling coefficient (k) and higher piezoelectric constant (d). However, these heavy metals are considered as one of the origins of environmental pollutions. Therefore, the lead-free piezoelectric materials with high piezoelectric performance are of current interest to materials researchers. The ferroelectric $AgNbO_3$ ceramic was found to be one of the candidate materials for lead-free piezoelectric materials. Preparation of such lead-free ceramics is of great interest today. The variables such as purity, stoichiometry and particle size, aggregation state, processing and sintering procedures have a marked effect on their characteristics. The crystalline, physical and electromechanical properties of ferroelectric material greatly depend on the preparation method. For studies of fundamental properties of materials, large homogeneous single crystals are usually desirable to minimize the effects of surfaces and imperfections. However, single crystals are very expensive and difficult to grow, whereas ceramics have the advantage of being a great deal easier to prepare. In addition, in ceramics, there are possibilities of forming new phases, polymorphic transformations, and decomposition of crystalline compounds. Silver niobate ($AgNbO_3$) is a member of the perovskite niobate. Due to technological importance and tremendous technological potential silver niobate [1,2] and its mixed systems: silver tantalum niobate [3–5], silver sodium niobate [6–8], silver potassium niobate [7,9] and silver lithium niobate [10–14] have been studied by several researchers. The development and improvement of electronic systems must be preceded by the designing of new miniaturized and suitable components. Although existing commercial components meet the engineering criteria, other new systems can be developed or existing ones can be perfected by introducing new components with improved characteristics. This development encourages the investigations of electronic materials, in particular the development of dielectric materials with specific dielectric properties that are required for specific applications. The processing and sintering procedures plays an important role in the characteristics of ceramics with and without additives. Valant et al. [3] studied the micro-structural and dielectric properties of $Ag(Nb_{1-x}Ta_x)O_3$ compositions made by three different processing ways: milling for 30 min at 150 rpm in a ZrO_2 mill; hand-grinding; and granulating to obtain three different particle size distributions.

In the present study, ceramic pellets of silver niobate, $AgNbO_3$ were prepared by two different sintering methods. The values of

* Corresponding author at: Raja Ramanna Centre for Advanced Technology, Department of Atomic Energy, Laser Materials Division, Indore 452013, M.P., India. Tel.: +91 731 248 8656; fax: +91 731 248 8650.
E-mail address: bartwalks@yahoo.co.in (K.S. Bartwal).

0925-8388/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.jallcom.2010.05.171

Table 1
Lattice constants and average crystallite/grain sizes for $AgNbO_3$ prepared by two different methods at room temperature.

S. no.	Compositions	Lattice parameters			Crystallite size (XRD) (μm)	Grain sizes (SEM) (μm)
		a (Å)	*b* (Å)	*c* (Å)		
1	$AgNbO_3$ (Ist method)	5.5972	5.5423	3.9100	0.0212	5.127
2	$AgNbO_3$ (Ist method)	5.6035	5.5473	3.9109	0.0223	7.472

lattice parameters, crystallite and grain sizes of $AgNbO_3$ were investigated. Frequency variations of dielectric constant (K), tangent loss ($\tan\delta$) and electrical conductivity (σ) were also measured and compared.

2. Experimental details

The ceramic samples of silver niobate ($AgNbO_3$) were prepared by solid-state reaction and sintering method. The starting materials used for preparing $AgNbO_3$ were silver oxide (Ag_2O), purity 99% [*Qualigens fine chemicals*], and niobium pentaoxide (Nb_2O_5), purity 99.9% [*Loba Chemie*]. The starting materials were initially dried at 200 °C for 2 h to remove the absorbed moisture and then quantities of the reagent were weighed in stoichiometric proportion. After weighing and mixing the starting chemicals Ag_2O and Nb_2O_5, the samples of silver niobate ($AgNbO_3$) were prepared by the following two sintering methods.

2.1. Ist method (calcination method)

The ground and mixed powder was again wet-mixed using methyl alcohol in a pestle and mortar for 2 h. The grounded powder was calcined at 850 °C for 3 h for solid-state reaction. The powder was grounded and pellets were made using a pelletizer of 11 mm diameter applying pressure of 3 tons. The prepared pellets were placed on a silica crucible and sintered in air atmosphere at 1050 °C for 12 h. These light yellow color pallets were used for the experiments.

2.2. IInd method (three-step sintering method)

In this method the mixed fine powder was wet-mixed using methyl alcohol in a pestle and mortar for 2 h. The powder was grounded and pellets were made as above in the Ist method. These pellets were sintered for 3 h at 850 °C in a silica crucible. The sintered pellets were crushed, milled and mixed for 2 h and again pressed into pellets. The pressed pellets were sintered at 1040 °C for 2 h. Again, the above sintered pellets were crushed, milled, and mixed for 1 h and pressed into pellets and finally sintered at 1100 °C for 3 h. A light yellow color and good quality ceramics have been obtained.

The sintered pellets obtained by both the methods were used for SEM characterization and dielectric measurements. The prepared ceramic samples were characterized for phase and crystallinity by powder X-ray diffraction technique using *D-8 ADVANCE* X-ray diffractometer (Bruker). Surface topography of the prepared samples was studied by scanning electron microscopy (SEM) using LEO-440 scanning electron microscope. Crystallite and grain sizes were determined using XRD and SEM data. Capacitance, resistance, and dissipation factor (tangent loss) measurements were carried out in metal–insulator–metal (MIM) configuration using Solarton 1260 impedance gain phase analyzer. The variations in dielectric constant (K), tangent loss ($\tan\delta$) and electrical conductivity (σ) were measured for $AgNbO_3$ prepared by both the methods in the different frequency ranges.

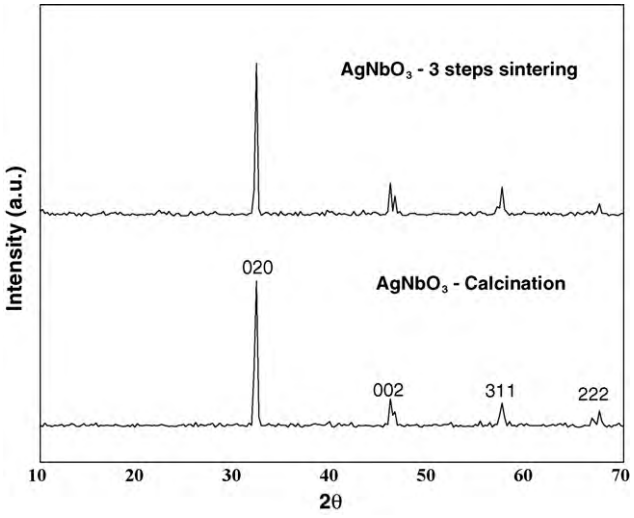

Fig. 1. Powder XRD pattern for $AgNbO_3$ ceramics prepared by both the methods.

3. Results and discussion

The light yellow color pallets of $AgNbO_3$ were obtained by two different sintering methods. The prepared ceramic samples were analyzed for phase and crystallinity by powder X-ray diffraction technique. X-ray diffraction pattern for all the samples were recorded at room temperature. The diffraction data were collected in the 2θ ranges of 10–70° with a scan step of 0.02°. Fig. 1 shows the X-ray diffraction patterns for $AgNbO_3$ ceramic prepared by two different methods. From the observed diffraction patterns the lattice spacing was determined which was used to determining the unit-cell parameters. The unit-cell parameters were determined using the '*WinPLOTR*' computer software (2005 version), which includes CRYSFIRE and FULLPROF software. From X-ray patterns, it was found that at room temperature pattern of silver niobate prepared by both the methods are same. The lattice constants of $AgNbO_3$ prepared by both methods are shown in Table 1. The calculated lattice parameters shows that at room temperature $AgNbO_3$ system is in orthorhombic phase. The lattice parameters for $AgNbO_3$ are nearly same but the peak intensity of reflected X-ray beam for the sample made by second method i.e. three-step sintering is high. It is understood that the crystallite sizes for the samples made by IInd method

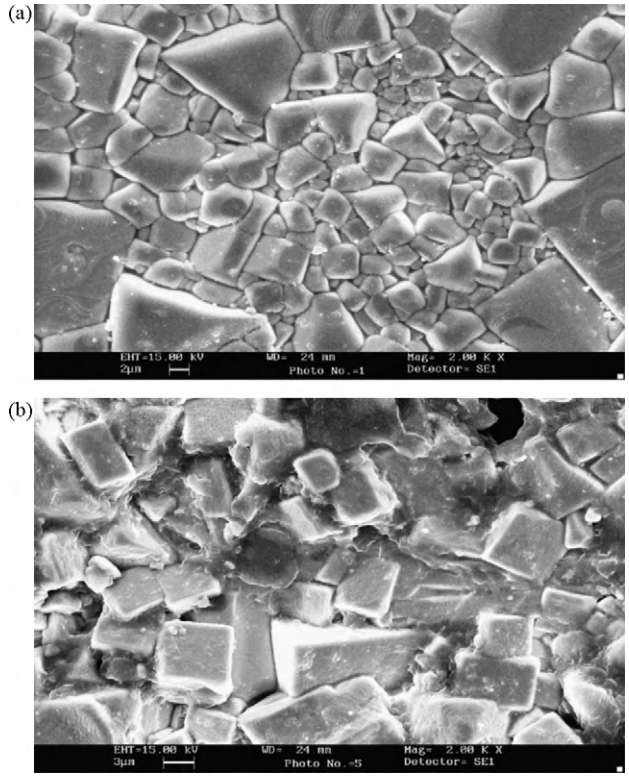

Fig. 2. Scanning electron micrographs for $AgNbO_3$ prepared by Ist method (a) and IInd method (b).