Solid State Ionics xxx (2014) xxx–xxx

Contents lists available at ScienceDirect

Solid State Ionics

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

Nonlinear and linear impedance of bismuth vanadate ceramics and its relation to structural properties

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ARTICLE INFO

Article history:
Received 15 July 2014
Received in revised form 16 September 2014
Accepted 23 October 2014
Available online xxxx

Keywords:
ferroelectric ceramic
nonlinear impedance
bismuth vanadate
impedance spectroscopy

ABSTRACT

The nonlinear and linear electrical properties, topography, and microstructure of bismuth-vanadate ceramics, were studied. The structure was observed with the use of X-ray diffraction (XRD), scanning electron microscopy (SEM), atomic force microscopy (AFM) and confocal microscopy methods. The obtained results showed that ceramic is porous. Two phase transitions were determined with the use of DSC measurements. The linear and nonlinear ac complex conductivity was studied as the function of frequency, temperature, and ac voltage. The activation energy of dc conduction processes was evaluated. The temperature dependence of conductance and permittivity showed a specific behaviour in two temperatures which were in agreement with phase transition temperatures. The nonlinear impedance studies showed that the ratio of third harmonic to base frequency conductivity reached more than 0.20. Obtained nonlinearities achieved two maxima near phase transition temperatures.

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1. Introduction

1.1. Structure of $Bi_2VO_{5.5}$

Bismuth vanadate ceramic $Bi_2VO_{5.5}$ (BiV) is ferroelectric at room temperature [1]. As thin film it shows nonlinear optical properties [2,3]. Because of its promising properties, this material have been studied for few decades. Several works were also devoted to doped variation of this material [4–8]. Its composition may be described as $(Bi_2O_2)^{2+}(VO_{3.5}\square_{0.5})^{2-}$ where □ are oxide ion vacancies. BiV can be considered to be analogous to α-Bi_2WO_6, the n = 1 member of Aurivillius family of oxides with intrinsic oxygen vacancies in the perovskite layer [9–11]. Bismuth vanadate crystallizes in a non-centrosymmetric, polar orthorhombic class [1,11]. It exhibits three main polymorphs: a non-centrosymmetric α-phase at room temperature, a centrosymmetric β-phase at 730 K and a centrosymmetric γ-phase stable above 835 K. BiV melts at 1153 K [10,12]. The α-phase and β-phase has an orthorhombic symmetry but the γ-phase has tetragonal symmetry [13]. The distortions of crystal cell are small therefore these phases can be described as mean orthorhombic cell with parameters: $a_m = 5.53$, $b_m = 5.61$, $c_m = 15.26$ Å [9,11]. The lattice type of α-phase is orthorhombic (space group *B2cb*) and the lattice parameters $a = 5.543$, $b = 5.615$ and $c = 15.321$ Å [2]. However, the literature presents also different values. In Sooryanarayana et al. [13] report, a space group *Aba2* is suggested with lattice parameters $a = 5.602(2)$, $b = 15.269(3)$ and $c = 5.5250(8)$ Å and Mairesse et al. [14] found $a = 5.6106(1)$, $b = 15.2707(3)$ and $c = 5.5316(3)$ Å.

The γ-phase exhibits a high ionic conductivity which is attributed to the presence of oxide ion vacancies in the perovskite layer. The oxygen vacancies in α and β-phases of BiV also give rise to non-negligible ionic conductivity of the order of 10^{-6} to 10^{-3} $\Omega^{-1} \cdot cm^{-1}$ [9,11,15,16]. Moreover, vanadium oxide is a transition metal oxide which may exist in two different valence states V^{4+} and V^{5+}. Materials containing these type of oxides may exhibit electronic conductivity usually described by the mechanism of polaron hopping between such ions [17–19].

1.2. Nonlinear impedance

Usually only linear measurements of impedance are performed. The linear measurements are sufficient for conventional situations and it is a small excitation amplitude what is used to guarantee linearity. For untypical systems with intrinsic nonlinearities and nonstationary processes it is better to use nonlinear method as well. It may provide us with more complete information [20,21]. The magnitude and the frequency dependence of the nonlinear conductivity may be used for instance to provide additional clue about the mechanisms of ion transport (the apparent jump distance of ion hopping) [21–23]. However, this procedure was performed in high electric field.

Nonlinear electrical properties may be described by the dependence of current density on the electric field [21]:

$$j = \sigma_1 E + \sigma_3 E^3 + \sigma_5 E^5 + \ldots \quad (1)$$

where σ_1 denotes linear conductivity, while σ_3, σ_5 etc. are higher order conductivity coefficients. An application of a sinusoidal electric field

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http://dx.doi.org/10.1016/j.ssi.2014.10.028
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Please cite this article as: N.A. Szreder, et al., Nonlinear and linear impedance of bismuth vanadate ceramics and its relation to structural properties, Solid State Ionics (2014), http://dx.doi.org/10.1016/j.ssi.2014.10.028

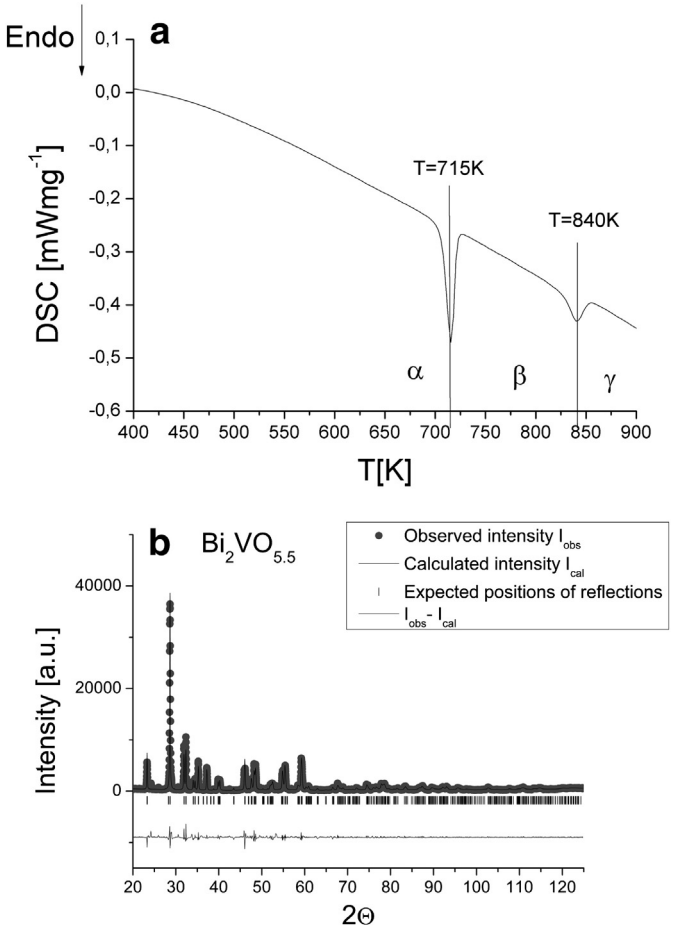

Fig. 1. (a) DSC curve of Bi $_2$VO$_{5.5}$; (b) fit (solid line) of the refined structural model (Rietveld method) to the room-temperature x-ray powder diffraction data (circles). Upper part – circles, present observed data, solid line show calculated intensities. The lower part (line) shows on the same scale the differences between the observed and calculated pattern. The bars correspond to $Bi_2VO_{5.5}$.

$E(t) = E_0 \cdot \sin(\omega t)$ leads to the following expression for the current density being in phase with the electric field j' [21]:

$$j' = \sigma'_1 E_0 \sin(\omega t) + \sigma'_3 E_0^3 \sin^3(\omega t) + \sigma'_5 E_0^5 \sin^5(\omega t) + \ldots = \sigma'_1(\omega) E_0 \sin(\omega t) + \frac{3}{4}\sigma'_3(\omega) E_0^3 \sin(\omega t) - \frac{1}{4}\sigma'_3(3\omega) E_0^3 \sin(3\omega t) + \frac{10}{16}\sigma'_5(\omega) E_0^5 \sin(\omega t) - \frac{5}{16}\sigma'_5(3\omega) E_0^5 \sin(3\omega t) + \frac{1}{16}\sigma'_5(5\omega) E_0^5 \sin(5\omega t) \ldots \quad (2)$$

As one can see from this equation, higher order conductivity coefficients may be determined with the aid of the higher harmonic currents [20,21]. Since BiV material exhibits ferroelectric and nonlinear optical properties, therefore it is worth to study nonlinear electrical properties also, what is the main aim of the presented work. We also intend to determine influence of neglecting nonlinear effects for precision of linear impedance analysis.

2. Experimental

Polycrystalline $Bi_2VO_{5.5}$ ceramic was synthesised via a conventional solid state reaction route [24]. The stoichiometric mixture of initial powders of Bi_2O_3 and V_2O_5 were ball-milled in pure acetone for 6 h. The milling was performed in steps of 1 h with rest intervals of 10 min. The mixture was initially heated up to 770 K and then to 1020 K in air. It was kept at this temperature for 24 h and grinded next. The formation of the compound was confirmed by X-ray powder diffraction studies. The calcined powder was mixed with a small amount of ethyl alcohol binder and cold-pressed into pellets (12 mm in diameter and 2–3 mm in thickness) under a compacting pressure of 26 kNcm^{-2}. The obtained pellets were sintered at 1070 K for 24 h with heating and cooling rates of 50 Kh^{-1}.

Differential scanning calorimetry (DSC) measurements were performed on powder samples in a nitrogen flow of 50 cm^{3}min^{-1} using Netzsch STA 449 F1. The heating rate was maintained at 15 Kmin^{-1} in the temperature range of 323–923 K. The structure has been studied by the X-ray diffraction method with the use of a Philips X 'Pert Pro MPD system with the CuKα radiation. The topography of the samples was investigated by Atomic Force Microscope NT-MDT Ntegra, Scanning Electron Microscope FEI Company Quanta FEG250 and Confocal Microscope Olympus OLS 4000 Lext with a CCD camera. Density of sintered pellets was measured by the Archimedes method in kerosene.

For the electrical measurements gold electrodes were evaporated at the polished samples. Impedance measurements were carried out in the frequency range from 10 mHz to 1 MHz, the ac voltage range from 0.01 to 3 V_{rms} and the temperature range from 153 to 923 K with the Novocontrol Concept 40 broadband dielectric spectrometer in nitrogen atmosphere and high temperature Novotherm HT 1600 in air atmosphere. Dc measurements were carried out with the use of Hioki 3522–50 LCR HiTester.

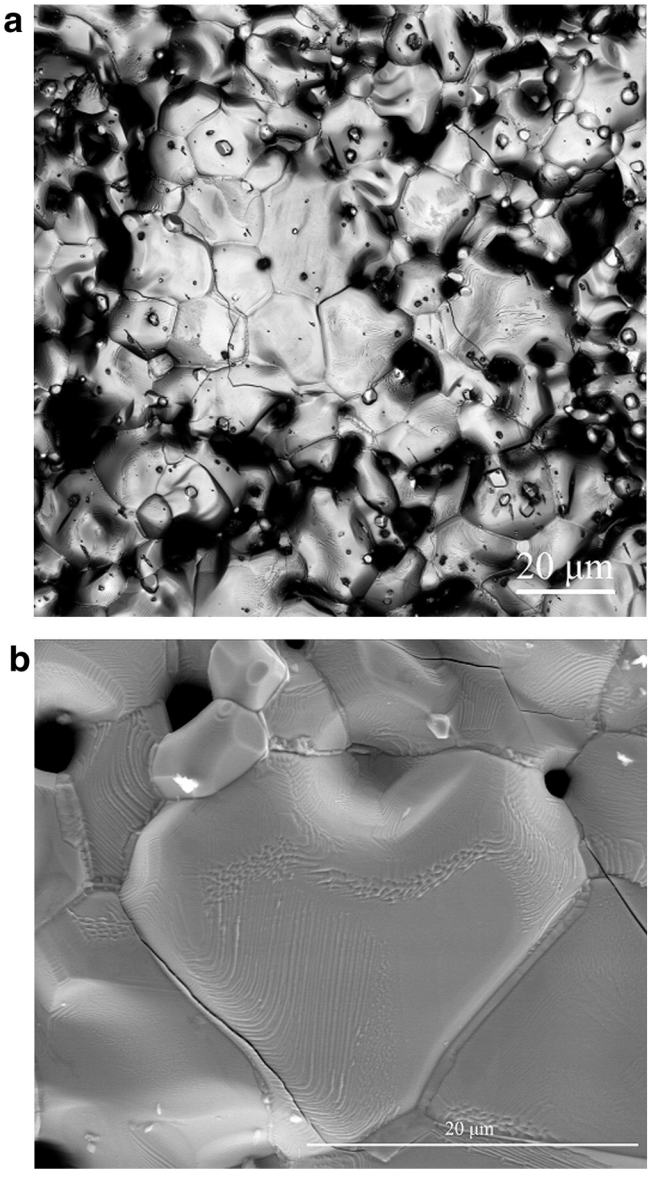

Fig. 2. (a) The confocal microscopy image; (b) SEM microscopy image.

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