Journal of Alloys and Compounds 656 (2016) 470–475

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

New low-dielectric-loss $NiZrNb_2O_8$ ceramics for microwave application

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ARTICLE INFO

Article history:
Received 9 November 2014
Received in revised form
19 September 2015
Accepted 1 October 2015

Keywords:
Ceramics
Microwave dielectric properties
Microstructure
Packing fraction
$NiZrNb_2O_8$

ABSTRACT

New low-loss $NiZrNb_2O_8$ microwave dielectric ceramics were synthesized via conventional mixed oxide route. The morphology, crystal structure and microwave dielectric properties were investigated. It exhibited a monoclinic wolframite crystal structure, with the space group of *P2/c* (C_{2h}^4). The lattice energy was carried out to evaluate the structural stability and sintering characteristics. Two kinds of grain shapes in dense samples, with similar elements ratios, were observed. Variations in the dielectric constant (ε_r) were analyzed by relative density and porosity-corrected polarizability. The quality factor ($Q \times f$) was correlated with packing fraction and grain growth. The temperature coefficient of resonant (τ_f) was depended on the dielectric constant. The typical microwave dielectric properties of $NiZrNb_2O_8$ were $\varepsilon_r = 23.77$, $Q \times f = 40280$ GHz, $\tau_f = -27.5$ ppm/°C, sintered at 1200 °C.

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1. Introduction

Microwave dielectric ceramics play an important role in the development of wireless communications devices, such as global positioning systems (GPS), microwave transmitting circuits, cell phones, and intelligent transportation systems (ITS) [1]. Recent revolution in telecommunications requires developing a variety of microwave dielectric ceramics [2,3]. Dielectric ceramics have been regarded as the basic materials in modern communication technology, for those in microwave application, three requirements need to be satisfied: (1) high dielectric constant for miniaturization ($\lambda = \lambda_0/\sqrt{\varepsilon_r}$, λ is the wavelength of electromagnetic wave in dielectric material and λ_0 is the wavelength of electromagnetic wave in free space), (2) low dielectric loss (or high quality factor) for enhancing the signal-to-noise ratio, (3) the near-zero temperature coefficient of resonant frequency for good performance of the devices under different atmospheric conditions.

Ceramics of $ABNb_2O_8$ form have attracted a great deal of interests for their predominant microwave dielectric properties. Numerous investigations have been reported on $ATiNb_2O_8$with different cations substituted at A-site [4–7]. Recently, much attention has been paid to investigations on Ti-site substitution of these compositions [8–12]. Liao et al. reported that the microwave dielectric properties of $ZnZrNb_2O_8$ ceramics were dependent on the relative density [9]. Then, Cheng et al. reported the $MgZrNb_2O_8$ ceramics with high $Q \times f$ values [10]. Especially, Ramarao et al., reported a class of $AZrNb_2O_8$ (A = Mn, Mg, Zn and Co) materials last year [11]. However, it was noted that the $NiZrNb_2O_8$ had been ignored out of the $AZrNb_2O_8$ system. Furthermore, the Ni ion was an important member in bivalent ions, which was frequently used as a substitution [13–15].

In the present study, we reported the microwave dielectric properties of $NiZrNb_2O_8$ ceramics, which exhibited the same crystal structure with $AZrNb_2O_8$ (A = Mn, Mg, Zn and Co) ceramics. The $NiZrNb_2O_8$ possessed promising microwave dielectric properties ($\varepsilon_r = 23.77$, $Q \times f = 40280$ GHz, $\tau_f = -27.5$ ppm/°C) and low sintering temperature ($T_s = 1200$ °C) in $AZrNb_2O_8$ system.

2. Experimental procedure

$NiZrNb_2O_8$ ceramics were synthesized using mixed oxide route from the raw materials including NiO (98%), ZrO_2 (99%) and Nb_2O_5 (99.5%). These oxides were mixed through de-ionized water and zirconia balls for 24 h. The mixture was dried, crushed and sieved with a 40 mesh screen. Then the powders were calcined at 900 °C for 3 h in air. The calcined powders were afterwards milled for 24 h.

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http://dx.doi.org/10.1016/j.jallcom.2015.10.008
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After drying, the powders were pressed into pellets with 10 mm in diameter and 5 mm in thickness. These pellets were then sintered at the temperature range of 1125–1250 °C for 6 h with a heating rate of 5 °C/min.

The crystal structures of the sintered samples were confirmed by analyzing powder's X-ray diffraction (XRD) patterns using a Rigaku diffractometer (Model D/Max-B, Rigaku Co., Japan) with Ni filtered Cu Kα radiation (λ = 0.1542 nm) at settings of 40 kV and 40 mA. The microstructure observation on ceramic surfaces was performed and analyzed by a scanning electron microscopy (SEM, FEI Quanta 250, USA), and the compositions of the ceramics were characterized by Energy Dispersive Spectrometer (EDS, Bruker Quantax400-10, Germany). Microwave dielectric properties of the sintered pellets were measured by a network analyzer (N5234A, Agilent Co., USA) in the frequency range of 7–8 GHz using Hakki–Coleman's dielectric resonator method [16,17]. The temperature coefficient of resonant frequency was obtained by measuring the $TE_{01\delta}$ resonant frequency from 25 °C to 85 °C. The τ_f (ppm/°C) could be calculated by noting the change in resonant frequency (Δf)

$$\tau_f = \frac{f_2 - f_1}{f_1(T_2 - T_1)} \tag{1}$$

where f_1 was resonant frequency at T_1 and f_2 was the resonant frequency at T_2.

The theoretical density and relative density were calculated by Eqs. (2) and (3):

$$\rho_{theory} = \frac{nA}{V_c N_A} \tag{2}$$

$$\rho_{relative} = \frac{\rho_{bulk}}{\rho_{theory}} \tag{3}$$

where n was the number of atoms in unit cell, A was atomic weight, V_c was unit-cell volume, and N_A was Avogadro number.

The packing fraction, defined as the summation of the volume of packed ions over the volume of a primitive unit cell, was calculated from Eq. (4) [18]:

$$\text{Packing fraction (\%)} = \frac{\text{volume of the atoms in the cell}}{\text{volume of primitive unit cell}} = \frac{\text{volume of the atoms in the cell}}{\text{volume of unit cell}} \times Z \tag{4}$$

where Z was the number of formula units per unit-cell.

3. Result and discussion

The XRD patterns of $NiZrNb_2O_8$ samples sintered at 1125–1250 °C were shown in Fig. 1. All the diffraction patterns were indexed with a wolframite structure belonged to $P2/c$ (C_{2h}^4) space group (monoclinic: ICDD-PDF #48-0327). The crystal phase of the samples with various sintering temperature exhibited no phase difference, with no second phase being detected. The Rietveld refinement analysis of $NiZrNb_2O_8$ ceramics sintered at 1200 °C was shown in Fig. 2. Moreover, as the results showed in Fig 3, the unit-cell volume of the samples decreased with an increase of the sintering temperature, and then increased when the sintering temperature was greater than 1200 °C. Furthermore, the details of peak shift with different sintering temperature were also showed in Fig. 3. The peak positions shifted to the higher angle when the sintering temperature increased from 1125 °C to 1200 °C and then shifted to the lower angle, which owed to the variation of unit-cell

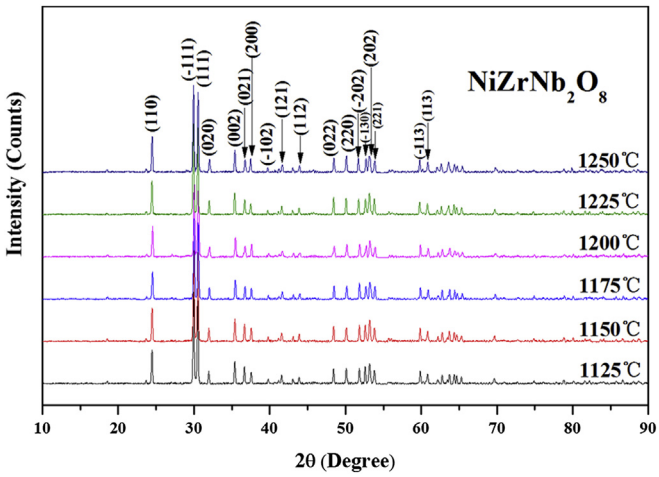

Fig. 1. The X-ray diffraction patterns of $NiZrNb_2O_8$ ceramics sintered at different sintering temperatures.

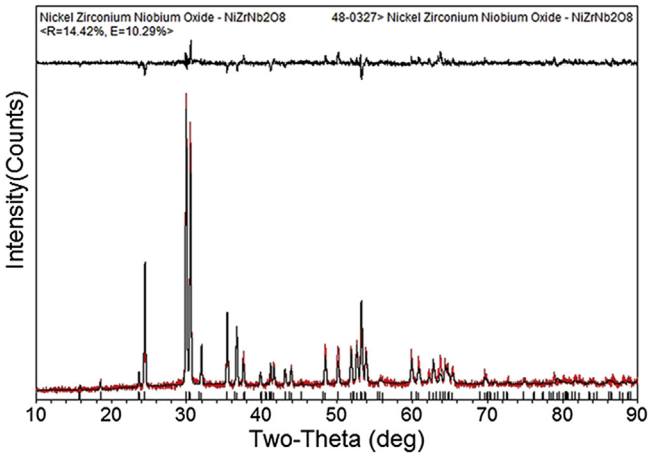

Fig. 2. The observed and calculated X-ray diffraction patterns by Rietveld analysis for $NiZrNb_2O_8$ ceramics sintered at 1200 °C.

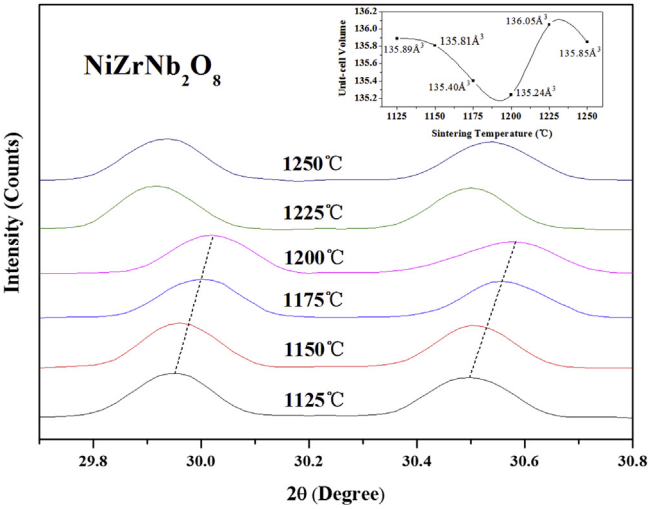

Fig. 3. Shift of X-ray diffraction peaks and unit-cell volumes of $NiZrNb_2O_8$ ceramics sintered at different sintering temperatures.

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