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Preparation and microwave dielectric properties of new Co_2NdNbO_6 ceramic materials

Ming Wei[a,b], Zhanguo Ma[a], Wangsuo Xia[c], Tianliang Tang[c], Zhao Li[d], and Lin Tian[d]

[a]State Key Laboratory for Geomechanics & Deep Underground Engineering, China University of Mining & Technology, Xuzhou, China; [b]College of Mathematics and Physics, Xuzhou University of Technology, Xuzhou, China; [c]School of Physics, China University of Mining & Technology, Xuzhou, China; [d]College of Chemistry and Chemical Engineering, Xuzhou University of Technology, Xuzhou, China

ABSTRACT

By using a solid-state reaction method, new ceramics compounds Co2NdNbO6 were synthesized. These compounds consisted of two phases and had a stable structure. The relation of dielectric constant (ε_r), relative density, grain size, and polarizability were discussed. The quality factor (Q_f) values were strongly affected by the density, and a sudden decrease of the density can lead to the decline of the Q_f values. The ceramics possessed a dielectric constant ε_r value of 17.62, a Q_f value of 12,200 GHz and a temperature coefficient τ_f value of −65.98 ppm/°C at 1200 °C for 4 h.

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KEYWORDS
Ceramics; Co_2NdNbO_6; microwave dielectric properties

1. Introduction

The rapid progress of wireless communication demands new and different kinds of ceramics to meet its applications. In the past years, several new functional ceramic oxides had attracted much attention for the application in resonator, filter and so on [1–3]. Many ceramic oxygen compounds with the chemical formula A_2BCO_{6+x} were synthesized and characterized, where A = Mg, Ca, Mn, and so on; B = Nd, La or Ce; C = Ta, Nb and $-0.5 < x < 0.5$ [4–12]. For example, Kumar [4] had synthesized two new oxide ceramic materials Mg_2NdTaO_6 and Mg_2LaTaO_6 and discussed the relation of the sintering temperature, the lattice structure, dielectric constant and conductivity. Zhang [5] firstly and successfully prepared the metal oxide compounds Mg_2NdNbO_6 which possessed excellent microwave dielectric properties and would be a good candidate for future electronic application. They found that the trend of dielectric constant was strongly affected by the relative density and Q_f values were mainly dependent on the grain sizes respectively. In the same year, a new ceramic material Mg_2CeNbO_6 was synthesized [6], the results showed that the ε_r increased monotonously with the sintering temperature, the Q_f values were mainly affected by the intrinsic loss and the grain size. However, as an important bivalent ion the Co ion was frequently used in substitution, and the compounds of the Co_2NdNbO_6 seem to be ignored and their dielectric properties have not been investigated carefully. In this paper, a new compounds Co_2NdNbO_6 were successfully synthesized, the preparation and their properties including the

CONTACT Zhanguo Ma 1275753577@qq.com; Wangsuo Xia 306785396@qq.com

microstructure structure, dielectric constant, the temperature coefficient values and quality factor values were presented.

2. Experimental process

According to the formula of Co_2NdNbO_6 composition, CoO(99%), Nd_2O_3(99%), and Nb_2O_5(99%) powders were mixed thoroughly and ball-milled for 6 hours in ethanol using ZrO_2 balls. then the slurries were dried, and the mixed powders were pre-sintered at 1000 °C for 4 hours. Next the pre-sintered powders were re-milled for 24 hours. After then the slurries were dried again, the dried solid particles were crushed and sieved by an 80-mesh screen. At last, the mixed powders were pressed into cylinders with 10 mm diameter and 5 mm thickness, these cylindrical samples were sintered between 1100 and 1200 °C for 4 hours in air.

The crystallographic structures of samples were detected by the X-ray powder diffraction (XRD) with Cu Kα radiation (Rigaku Ultima IV, Japan). The surface characteristics of samples were analyzed by the means of the SEM (Sigma Zeiss, German). The bulk densities of samples were measured by a densitometer (Mettler Toledo XS64) with the Archimedes method. Using the Hakki-Coleman's dielectric resonator method [13–15], the dielectric properties of specimen were characterized by a network analyzer (N5234A, Agilent, USA). At last, the temperature coefficient of resonant frequency (τ_f) was achieved by calculating the next formula and measuring the resonant frequency in the temperature ranges from 25 to 85 °C.

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

Where f_1 and f_2 were $TE_{01\delta}$ resonant frequencies at T_1 and T_2, respectively.

3. Results and discussions

The XRD patterns of the Co_2NdNbO_6 ceramics were demonstrated in Fig. 1. Two phases were observed: the main phase was the $NdNbO_4$ with the monoclinic fergusonite structure (ICDD-PDF #32-0680) belonged to the space group I2/a (no.15) for all the samples. Another phase was the CoO, which possessed the cubic structure (ICDD-PDF #48-1719) and belonged to the space group Fm-3m (225). With the sintering temperature increased, the peak positions of the phases of $NdNbO_4$ and CoO remained unchanged, and the relative intensities exhibited no obvious difference. Above all, it suggested that the crystal structure of the Co_2NdNbO_6 ceramics was stable during the sintering temperature.

Figure 2(a–e) showed SEM photographs of the Co_2NdNbO_6 ceramics. According to the graphs, two different crystal grain shapes were detected, the crystal grains had homogeneous and straight grain boundaries. With the sintering temperature increased, the grain sizes had an increased trend, when the sintering temperature was higher than 1175 °C, the grain sizes increased slightly. All of the samples had well-packed grains and no obvious porous were detected. By the image processing technology, the particle sizes were estimated in the range from 0.57 to 1.5 μm and were showed in the Fig. 2(f). In order to confirm the chemical composition of the two kinds shapes,

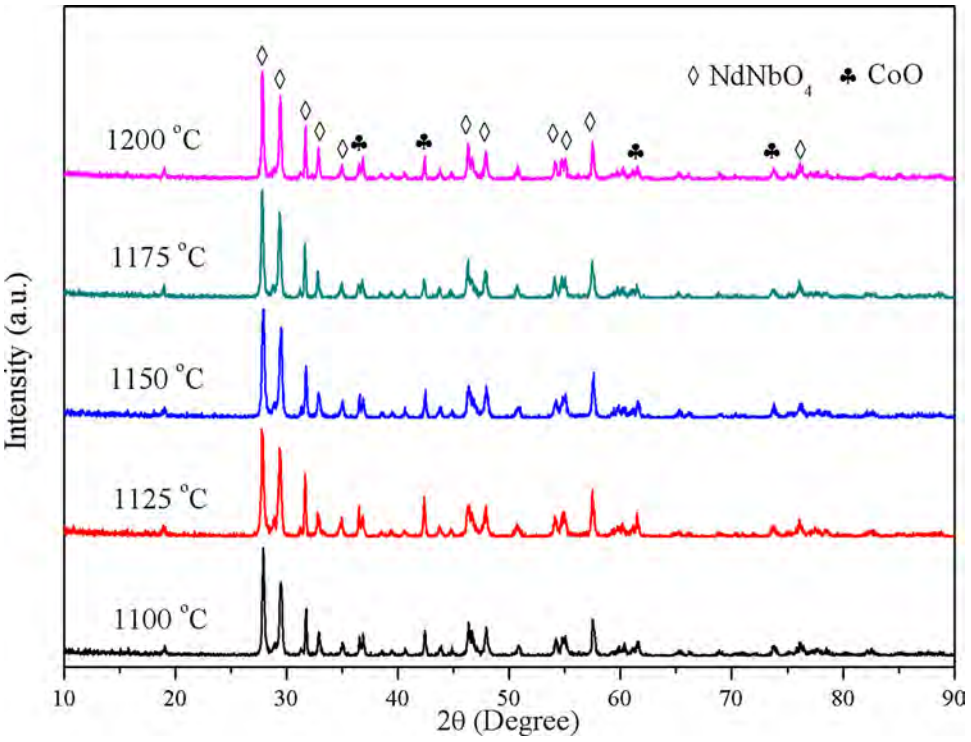

Figure 1. The XRD diffraction patterns of the Co_2NdNbO_6 ceramics sintered from 1100 °C to 1200 °C.

the Co_2NdNbO_6 sample sintered at 1175 °C was analyzed by the energy-disperse spectroscopy (EDS), see Fig. 3. Three elements were surrounded by each other, the Nd ions and Nb ions distributed uniformly and formed the $NdNbO_4$ phase, the Co ions formed the CoO phase alone. The distribution of Nd ions and Nb ions presented a complementary relationship with Co ions, and this was confirmed that the $NdNbO_4$ and CoO co-existed in Co_2NdNbO_6 ceramics, which was in accordance with the XRD patterns.

According to the Fig. 4(b), The dielectric constants ε_r of Co_2NdNbO_6 ceramics increased to the maximum value at 1150 °C and then decreased slowly with the sintering temperature changed from 1100 to 1200 °C. There were several factors affected the dielectric constant such as density, dielectric polarizabilities and structure characteristics [16], as to the XRD patterns of samples had no differences at different sintering temperature, the intrinsic factors weren't considered in the dielectric constant.

According to the Fig. 4(d), when the sintering temperature change from1100 to 1150 °C, the density has a same tendency to the dielectric constant, it indicates that during this temperature range, the density can have a major influence on the dielectric constant. When the sintering temperature was higher than 1150 °C, the ε_r tended to steady, but the density increased and then declined slightly, which indicated that density weren't the main factors affected the ε_r.

In the Fig. 4(c), stable τ_f values indicated Co_2NdNbO_6 ceramics had a stable structure during the sintering temperature range, and the τ_f values also showed the same

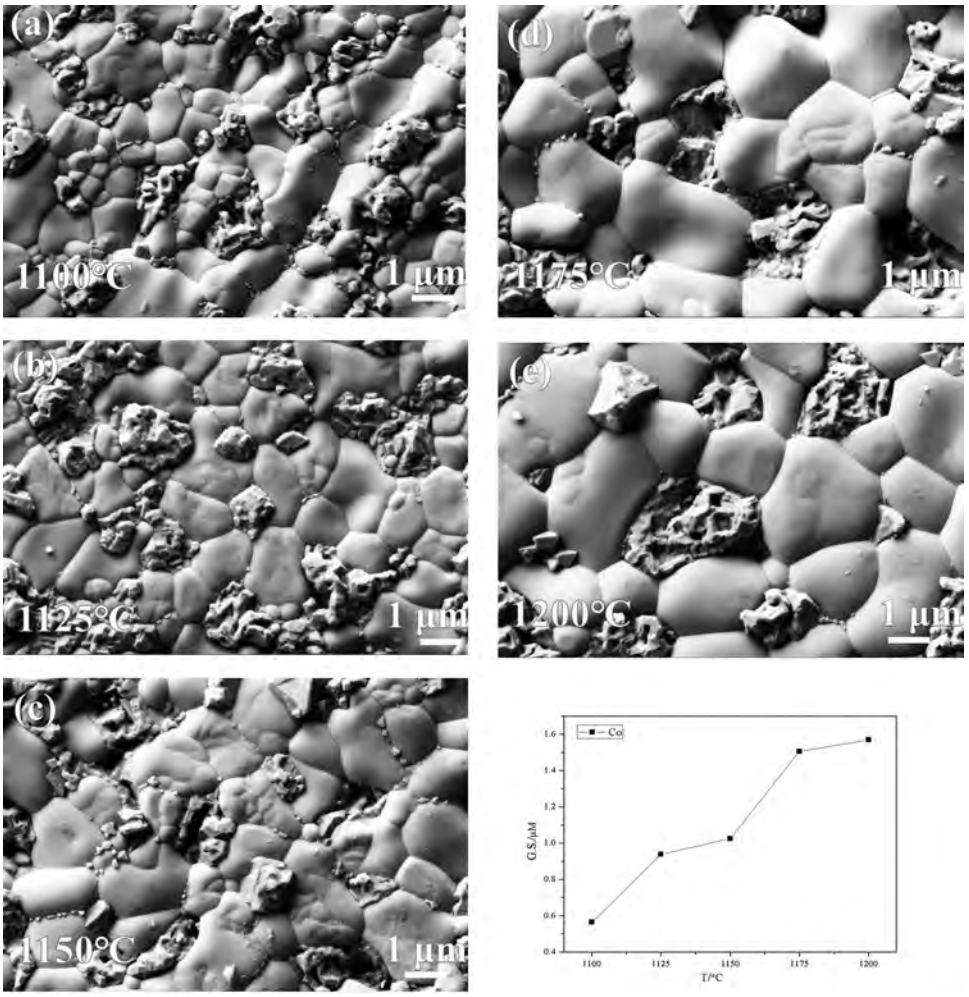

Figure 2. (a–e) The FEM images of Co_2NdNbO_6 ceramics sintered at 1100 °C, 1125 °C, 1150 °C, 1175 °C, 1200 °C and (f) the grain sizes change with temperature.

tendency with the dielectric temperature during the sintering temperature, which were consistent with the relation was founded as follows [17,18]:

$$\tau_f = \alpha_L \times \left(\frac{\varepsilon_r}{2} - 1\right) \propto \varepsilon_r \tag{2}$$

Here α_L was the linear thermal expansion coefficient, τ_f was temperature coefficient and ε_r was dielectric constant.

In the Fig. 4(a,d), the density values showed a same behavior with the Q_f values during the sintering temperature range. Both the values of Q_f and the density rose to the maximum values at 1175 °C, and then they showed a decreasing trend. The intrinsic loss and the extrinsic loss were the two major factors can affect the Q_f values. According to the SEM figures, the stable XRD patterns and the same chemical composition of samples, the intrinsic loss factors were not under consideration, and the extrinsic loss which was mainly affected by gran size, density and the second phase was

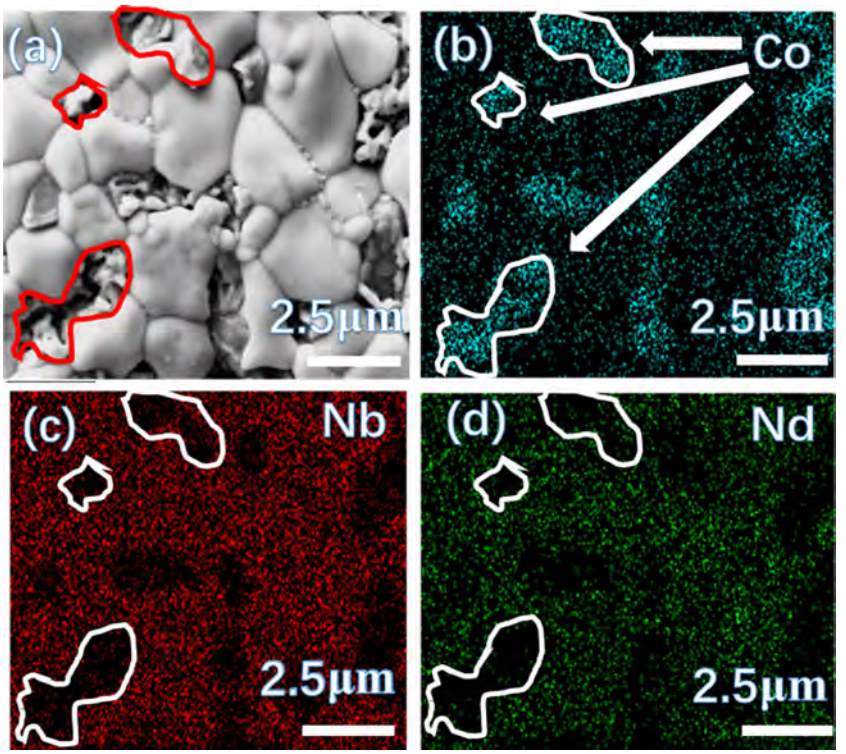

Figure 3. The EDS data of Co_2NdNbO_6 ceramics sintered at 1175 °C for 4 hours. (a) SEM image, (b–d) are Co, Nd, Nb distribution respectively.

discussed as follows. According to the Fig. 2, during the sintering temperature range of 1100–1200 °C, the monotonously increased grain size can lead to the decline of grain boundary, which can induce the increase of Q_f values. When the sintering temperature was higher than 1175 °C, the grain size slightly increased and the density declined suddenly indicated the existence of an abnormal grain growth, which can induce a significant decline of the Q_f values. In the meanwhile, the second phase of CoO might induce the decrease of Q_f values [19,20]. All of above, Q_f values were strongly influenced by the density and grain size, and the sudden decrease can lead to the significant decline of Q_f values, so we concluded that the density dominated Q_f values of the Co_2NdNbO_6 ceramics.

4. Conclusion

In this paper, a new Co_2NdNbO_6 ceramics were synthesized successfully and their microwave dielectric properties were reported for the first time. The new Co_2NdNbO_6 ceramics possessed two phases, the major phase is $NdNbO_4$ and another phase is CoO. There was no difference or additional phased were exhibited in the crystalline phase of all samples at different sintering temperatures. The EDS results showed that the distribution of Nd and Nb elements was the same, but Co element was not, and three

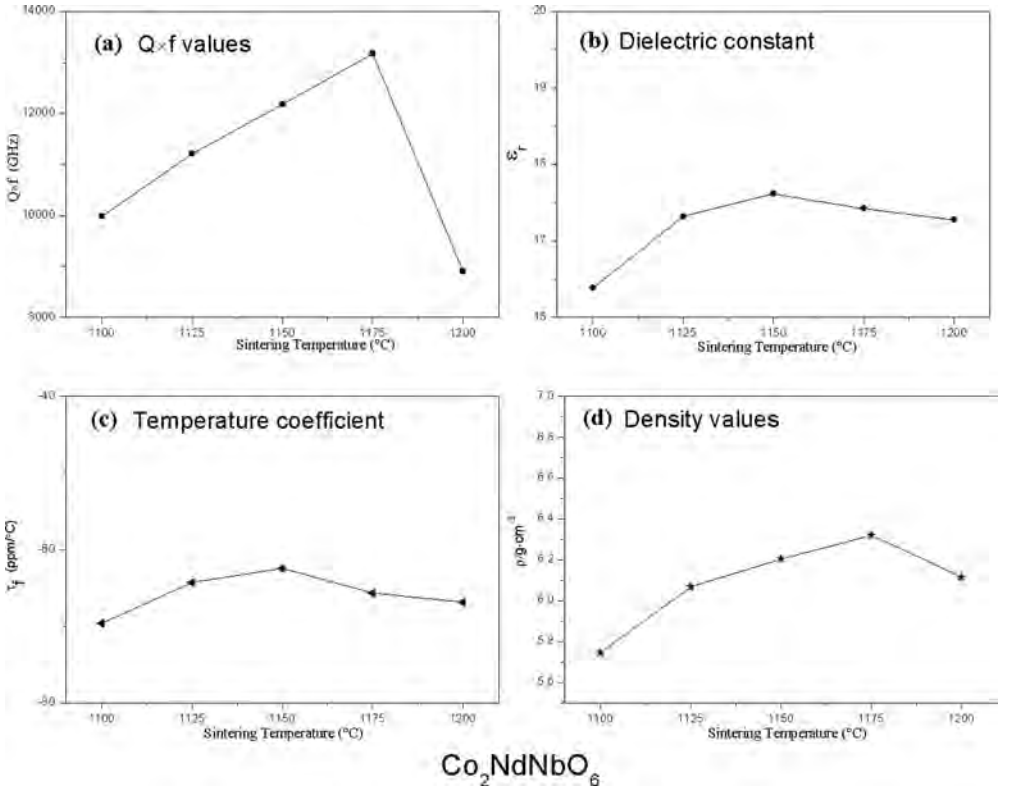

Figure 4. (a) The quality factor Q_f values; (b) Dielectric constant ε_r; (c) Temperature coefficient τ_f; (d) Density values change with the sintering temperature.

elements surrounded by each other which proved the $NdNbO_4$ and CoO co-existed in Co_2NdNbO_6 ceramics.

During the temperature range from1100 °C to 1150 °C, the dielectric constant ε_r increased with the density, when the temperature was higher than 1150 °C, the dielectric constant kept stable. The significant change of τ_f were not observed and the variation of τ_f was consistent with ε_r. The trend of Q_f values increased first and then decreased was proved to be dominated by the density. Sintered at 1175 °C for 4 hours, the Co_2NdNbO_6 ceramic possessed promising microwave properties (Q_f =12,200GHz, ε_r =17.62 and τ_f = −65.98 ppm/°C).

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