Effects of CuO additives and sol-gel technique on $NiNb_2O_6$ dielectric ceramics for LTCC application

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Abstract

The effects of CuO additives and sol-gel method synthesis on the sintering behavior, microstructure and the microwave dielectric properties of $NiNb_2O_6$ ceramics were investigated systematically. The $NiNb_2O_6$ ceramics were synthesized with traditional solid state method and sol-gel method, and the CuO additives were used in the solid state method for comparison. The sintering temperature of $NiNb_2O_6$ ceramics with the highest densification can be effectively reduced from about 1275˚C to 1050˚C and 1100˚C respectively by using CuO additions and sol-gel technique. To study their applicability in low temperature cofired ceramic (LTCC) technology, dielectric properties have been characterized. The dielectric properties exhibited a significant dependence on the sintering condition, composition and crystal structure of the ceramics. In particular, the 2.5 wt.% CuO-doped $NiNb_2O_6$ ceramics sintered at 1050˚C have excellent microwave dielectric properties: ε_r=21.45, $Q\times f$=23531GHz, τ_f=-27.9ppm/˚C.

While the $NiNb_2O_6$ ceramics prepared by sol-gel method obtain microwave dielectric properties as: ε_r=19.16, Q×*f*=11149GHz, τ_f=-27.3ppm/˚C after sintered at 1100˚C for 2h.

1. Introduction

Recently, much attention has been concentrated on multilayer microwave devices due to the rapid progress on the satellite and mobile communications such as cellular phone and GPS [1,2]. In multilayer structures manufacturers, the microwave dielectrics with high dielectric constant (ε_r), high quality factor (Q×*f*) and close-zero temperature coefficient of resonant frequency (τ_f) were needed to co-fire with low loss conductors and low melting-point electrode such as silver and copper at low temperature [3,4], as known as Low-temperature Co-fired Ceramic (LTCC) technology. The columbite compounds MNb_2O_6 (where M=Mg, Zn, Ni, Ca, Cu, Mn and Co) have received much attentions to be promising candidates due to their good dielectric properties and low cost. These compounds with an orthorhombic columbite crystal structure have very low loss and high dielectric constant. Especially $ZnNb_2O_6$ ceramics were reported to exhibit excellent dielectric properties: ε_r=25, Q×*f*=83700GHz, τ_f=-56.1ppm/˚C [5]. The $NiNb_2O_6$ ceramics were also selected as one of the candidates with good dielectric properties. A report of ε_r=22.6, Q×*f*=40100GHz, τ_f=-38ppm/˚C was presented by Lee et al [6]. Liou et al reported that $NiNb_2O_6$ with higher shrinkage and density were observed after sintered in 1300˚C for 2h [7]. But the results showed that the sintering temperature was too high for low temperature co-fire manufactures.

It is common known that the addition of liquid phase and using chemical method to prepare ceramics can reduce the sintering temperature. CuO, Bi_2O_3 and V_2O_5, with melt point as 1326˚C, 825˚C and 678˚C respectively, are common LTCC additives. The most LTCC works have been carried out on the best-known columbite $ZnNb_2O_6$ by various researchers [8-15]. However, the efforts on the microstructure and microwave dielectric properties of $NiNb_2O_6$ ceramics for LTCC applications have not been thoroughly studied yet.

Sol-gel technique develops rapidly in recent years. Compared to traditional solid state method, sol-gel technique can obtain smaller uniform particle size which help to decrease the sintering temperature without introducing second phase. It is a promising synthesis method for the LTCC application.

In this present work, The $NiNb_2O_6$ ceramics were prepared by using both traditional solid state method and sol-gel method. The additives of CuO were added for lowering the sintering temperature in the solid state method prepared $NiNb_2O_6$ ceramics. The influences of CuO dopants and sol-gel techniques on the density, microstructure and microwave dielectric properties of $NiNb_2O_6$ ceramics were investigated.

2. Experimental

Analytical grade oxide powders of NiO and Nb_2O_5 were used as starting materials for preparing $NiNb_2O_6$ with solid state method. They were mixed according to the stoichiometric composition $NiNb_2O_6$ and ball-milled in a polyethylene bottle with agate balls using ethanol as medium. The mixtures were then dried and calcined at 1000˚C for 2 h. Partial calcined powders were then re-milled for 6 h with CuO powder additions in the mass fraction of 0.0%, 0.5%, 1.0%, 1.5%, 2.0%, 2.5% and 3.0% respectively. After drying and sieving, the powders were uniaxial pressed into pellets under the pressure of 100Mpa. The samples were sintered at 1000~1300˚C for 2 h with a heating rate of 5˚C/min and then quenched to room temperature.

$NiNb_2O_6$ nanocrystals were synthesized via a similar sol-gel method reported by Zhou et al [16]. Nb_2O_5 was dissolved in HF acid (40%) after heating in an 80˚C hot water bath. Ammonia water was added to the solution to obtain $Nb_2O_5 \cdot nH_2O$ as a precipitate, which was then filtered, washed and dissolved in a citric acid aqueous solution. Stoichiometric $Ni(NO_3)_2 \cdot 6H_2O$ was added, and then excessive ammonia water was added and mixed homogeneously to adjust the pH value. After dried in 150˚C for hours, the solution turned to a yellow fluffy product which was then calcined to form the single phase columbite at 700˚C. The powders were ball-milled in ethanol with zirconia balls for 3 hours and uniaxial pre-pressed into pellets under pressure of 100Mpa. And then used cold isostatic pressing method under the pressure of 250Mpa for 10 minutes. The samples were sintered at 1000~1200˚C for 2 h with a heating rate of 5˚C/min and then quenched to room temperature.

The crystal structures and purity of calcined powder samples were analyzed by X-ray powder diffraction using Cu Kα radiation (λ=1.540598 Å). The particle size and microstructures of samples were observed by transmission electron microscope (TEM) and scanning electron microscopy (SEM). Energy dispersive spectra (EDS) were used to analyze the elements composition in a selected micro zone. The bulk densities of the sintered ceramics were measured by Archimedes method using deionized water as the liquid. The microwave dielectric properties of sintered samples at microwave frequency (6–8 GHz) were measured with a HP8720ES network analyzer using Hakki–Coleman's dielectric resonator method, as improved by Courtney and Kobayashi et al [17-19].

3. Results and Discussions

3.1 Solid State Synthesis

$NiNb_2O_6$ ceramics were sintered at 1250-1350˚C by solid state route and the average densities after sintered were measured. The $NiNb_2O_6$ ceramics reached the highest density 5.603g/cm^3 (>99.5% theoretic density) at 1275˚C and the grain size was around 3-5 μm as shown in the SEM image Fig.1.

The dielectric properties were measured at 6-8 GHz and listed with other reported results in the Table 1. Among these reports, Pullar et al. [20] obtained the worst dielectric properties which can be due to the relatively low density. However, Lee et al. [6] got the highest quality factor $Q\times f$ value 40100 GHz and the lowest sintering temperature 1150 ˚C with only 95% theoretic density which was lack of explanation. In this research, the quality factor $Q\times f$ value for $NiNb_2O_6$ reached 37500GHz which was nearly double more than those reported by Pullar et al. [20], Liou et al. [7] and Butee et al [21], and get closed to the result of Lee et al. [6]. The dielectric constant was 22.1 and the temperature coefficient of resonant frequency was small negative as -30.4 ppm/˚C. The sintering temperature and density were 1275 ˚C and 5.603g/cm^3 (>99.5% theoretic density) respectively which are more or less the same with those reported by Liou et al. [7] and Butee et al [21] and higher density than Lee et al. [6].

3.2 Effects of CuO Additives

The densities of $NiNb_2O_6$ ceramic bulks with various amounts of CuO additions as sintering temperature ranged from 1000˚C to 1200˚C are shown in Fig.2. It can be seen that the sintering temperatures of $NiNb_2O_6$ ceramics with the highest densities point decreased as the increasing amounts of CuO additions. The sintering temperature with the highest density reached 1050˚C when 2.5% mass fraction CuO was added in and the density was 5.499 g/cm3 (97.5% of the theory density of $NiNb_2O_6$). No significant effect was found when CuO content reached 3.0wt%.

Fig.3 shows the XRD patterns of $NiNb_2O_6$ ceramics doped with different amounts of CuO sintered at their own optimum temperature. $NiNb_2O_6$ ceramics exhibited single phase with orthorhombic columbite crystal structure (matched with JSPDS files NO.32-0694) at all temperatures and no secondary phase were detected in the samples.

The SEM micrographs of $NiNb_2O_6$ ceramics with (x) wt.% CuO additions were presented in Fig.4. The homogeneous microstructures without obvious pores and abnormal grain growth were observed for small x values. The grain size was about 3-5μm. The trend of densification degree could not be observed clearly from the SEM figure. However as the increase of x value, some liquid phases were observed at grain boundary which was contributed to the density loss. To clarify the element distribution of the glass phases, a zone in 3.0 wt.% CuO doped $NiNb_2O_6$ ceramics sintered at 1050˚C was selected and measured by using EDS analysis equipped on SEM. The results which were shown in Fig.5 reveal that Cu and O were the main element compositions in the glass phases. This suggested that the liquid phases appeared at grain boundary were CuO additions which help to decrease the sintering temperature. No solid solution phase was observed in this case.

The microwave dielectric properties of $NiNb_2O_6$ ceramics doped with CuO additions were measured at 6-8 GHz and were shown in Table 2. No significant change of the dielectric constant values ε_r and the temperature coefficient of resonant frequency τ_f values was observed with the increase of CuO content. Generally, the addition of flux to lower sintering temperature is accompanied by degradation in dielectric properties. The stability of the dielectric constant values ε_r and the temperature coefficient of resonant frequency τ_f values was attributed to the slightly increase of bulk densities. However, the Q×f values decreased obviously with the increasing amount of CuO additions which was due to the formation and accumulation of the liquid phases at the grain boundary. The presence of the CuO liquid-phase help to lower the sintering temperature and promote the densification while introduce in dielectric loss. In this research, the Q×*f* values of $NiNb_2O_6$ decreased from 37500GHz to 19282GHz as the CuO addition content and sintering temperature varied from 0-3.0 wt.% and 1275-1050 ˚C respectively. The $NiNb_2O_6$ ceramics with proper amount of CuO additions such as 2.5 wt.% obtained good dielectric properties as ε_r=21.45, Q×*f*=23531GHz, τ_f=-27.9ppm/˚C with low sintering temperature 1050˚C.

3.3 Sol-gel Synthesis

The $NiNb_2O_6$ nano-particles were synthesized by using sol-gel method. The X-ray diffraction spectra of $NiNb_2O_6$ after calcined at 700˚C for 2h was presented in Fig.6. As we can see, no unexpected phase appeared. The spectra peaks were perfectly matched with JSPDS files NO.32-0694 giving orthorhombic columbite crystal structure. The $NiNb_2O_6$ nano-particles were dispersed and observed by transmission electron microscope (TEM) as shown in Fig.7. For all samples, the particles are irregular in shape, and the average size prepared at 700˚C was about 30-50 nm.

The densities of $NiNb_2O_6$ ceramics after sintered from 1000˚C to 1200˚C for 2 hours were measured respectively. As shown in Fig.8, the highest density was obtained at the 1100˚C point. The density was 5.090g/cm^3 (90.4% of the theory density of $NiNb_2O_6$. Compared with the reports of solid state reaction method, this value is a little small. It can be observed more clearly from the scanning electron microscopy (SEM) as shown in Fig.9. As the increasing of temperature, the grain sizes increased and formed continuous grained matrix. The grain sizes maintained homogeneous at 1100˚C while abnormal grain growth was observed as the temperature increased to 1150˚C. Plenty of pores which stayed at the grain boundary contributed the density loss. The grain size after sintered at 1100˚C for 2 hours is about 1-2 μm.

The dielectric properties of $NiNb_2O_6$ ceramics prepared by sol-gel method were also investigated in this research. As shown in Table 3, the dielectric constant ε_r and quality factor $Q \times f$ both decreased compared with those by using the solid state method. Especially the quality factor value, the change was tremendous. It was owe to the density loss introduced by the tiny grain particle and sintering process. Actually, Satapathy et al. [22] have first investigated the dielectric properties of $NiNb_2O_6$ ceramics which prepared by sol-gel method. In their report, the dielectric constant was found blow to 6 and the quality factor was 9000 GHz with lower sintering temperature (900˚C) and longer sintering time (6h). Obviously the samples in this present work have better dielectric properties, but the sintering temperature is much higher. It was indicated that the sintering process can affect the density and the dielectric properties. Longer sintering time can make up the density loss in some degree with lower sintering temperature. While the sintering time increased from 2 to 8 hours, the density can promoted from 5.090 g/cm^3 to 5.197 g/cm^3 and the surface image can be seen in Fig.10. As known that the dielectric properties especially quality factor $Q \times f$ value can be affected by the density, it can be deduced that better dielectric properties may be obtained with longer sintering time. But more systematic researches about the sintering process control are needed in future works. In this research, $NiNb_2O_6$ ceramics with dielectric properties as ε_r=19.69, $Q \times f$=11149GHz, τ_f=-27.3ppm/˚C were prepared by sol-gel method while sintered at 1100˚C for 2 hours.

4. Conclusions

The sintering behavior, microstructure and microwave dielectric properties of $NiNb_2O_6$ ceramics doped with CuO additions and prepared by sol-gel technique were investigated systematically in this research.

The $NiNb_2O_6$ ceramics were synthesized by solid state method after sintered at 1275˚C for 2h. Good dielectric properties were achieved ε_r=22.1, Q×*f*=37500GHz, τ_f=-30.4ppm /˚C with higher densification (>99.5% theory density).

The dopants CuO can significantly lower the sintering temperature of $NiNb_2O_6$ ceramics from 1275 to 1050˚C with high densification. The X-ray diffraction analysis was carried out and no secondary phases were observed. SEM and EDS results show that the CuO additions formed the liquid phases which were rich in Cu and O and gathered at the grain boundary. The CuO glass phases attributed to lower the sintering temperature and higher the densification while produce in more dielectric loss. Typically, the $NiNb_2O_6$ ceramics doped with 2.5 wt.% CuO additions obtain the lowest sintering temperature with high densification (97.5% of the theory density of $NiNb_2O_6$) while exhibiting good microwave dielectric properties: ε_r=21.45, Q×*f*=23531GHz, τ_f=-27.9ppm /˚C.

$NiNb_2O_6$ nano-particles about 30-50 nm were synthesized by using sol-gel method at 700˚C and no secondary phase was observed from the X-ray diffraction analysis. And after sintered at 1100˚C for 2 hours, a dielectric properties as ε_r=19.69, Q×*f*=11149GHz, τ_f=-27.3ppm/˚C was obtained. It should be noticed that the sintering process can affect the density and the dielectric properties efficiently. The densification promoted from 5.090 g/cm^3 to 5.197 g/cm^3 when the sintering time increased from 2 to 8 hours, and better dielectric properties and lower sintering temperature could be expected by controlling the sintering process. Sol-gel method is a promising technique to help to decrease sintering temperature for LTCC application.

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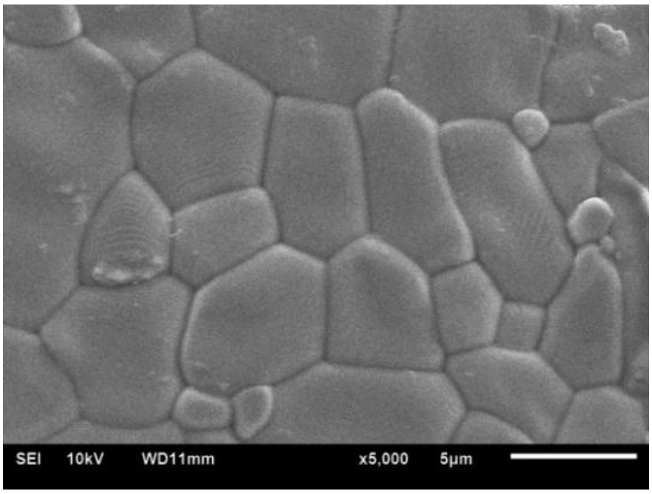


Fig.1 SEM surface image of $NiNb_2O_6$ ceramics prepared by solid state method sintered at 1275˚C

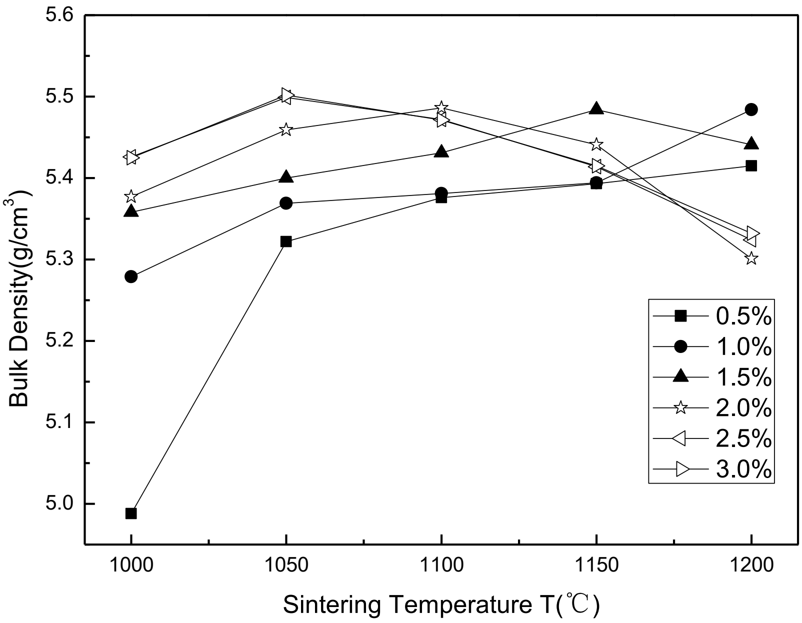


Fig.2 Density curves of $NiNb_2O_6$ with (x) wt.% (x=0.5-3.0) CuO additives sintered at 1000-1200˚C

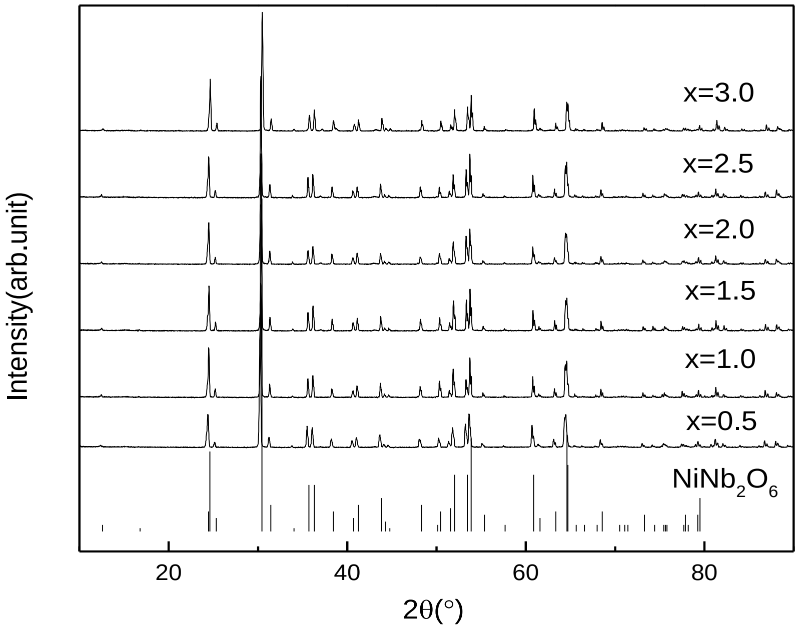
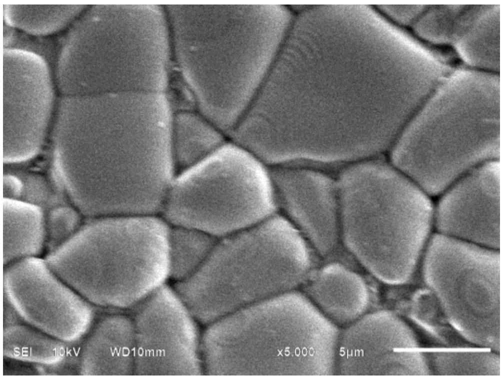
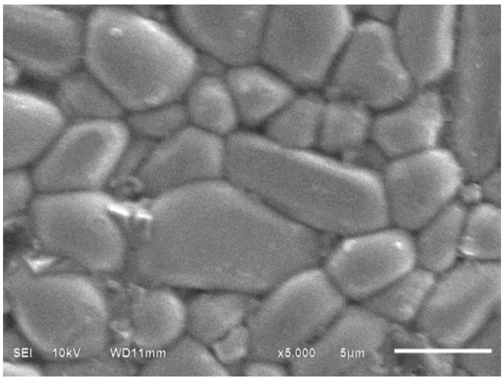


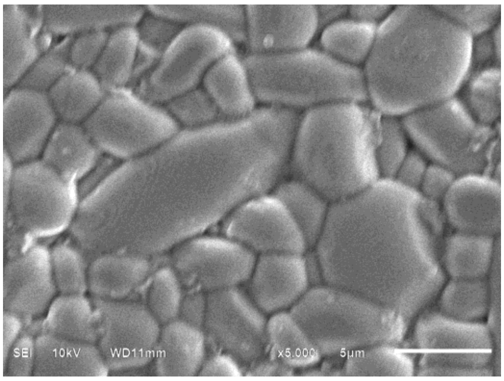
Fig.3 XRD patterns of $NiNb_2O_6$ doped with (x) wt.% CuO additives (x=0.5-3.0)



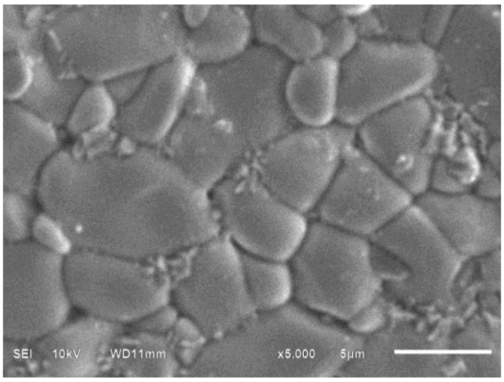
(a) 1.0wt.%，1200˚C



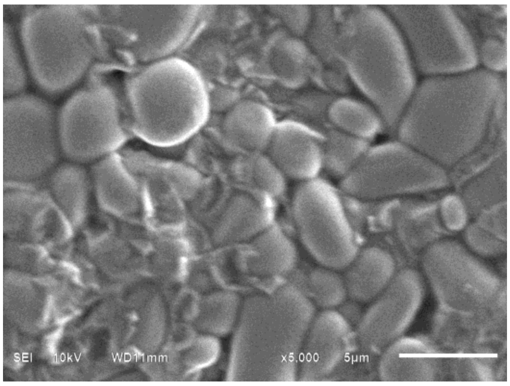
(b) 1.5wt.%，1150˚C



(c) 2.0wt.%，1100˚C

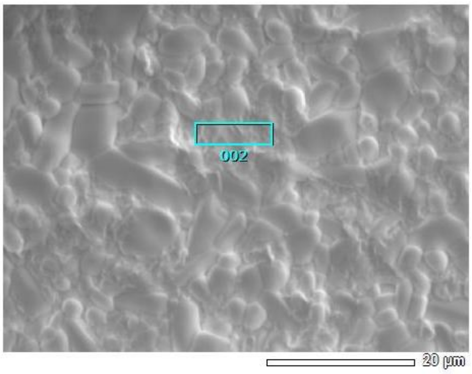


(d) 2.5wt.%，1050˚C



(e) 3.0wt.%，1050˚C

Fig.4 SEM surface images of $NiNb_2O_6$ ceramics with different amounts of CuO additives



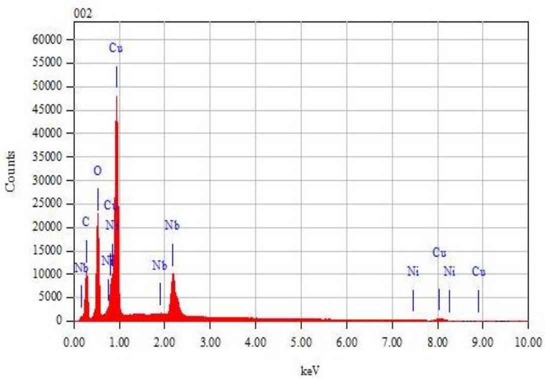


Fig.5 Energy dispersive spectrometer analysis of the liquid phase

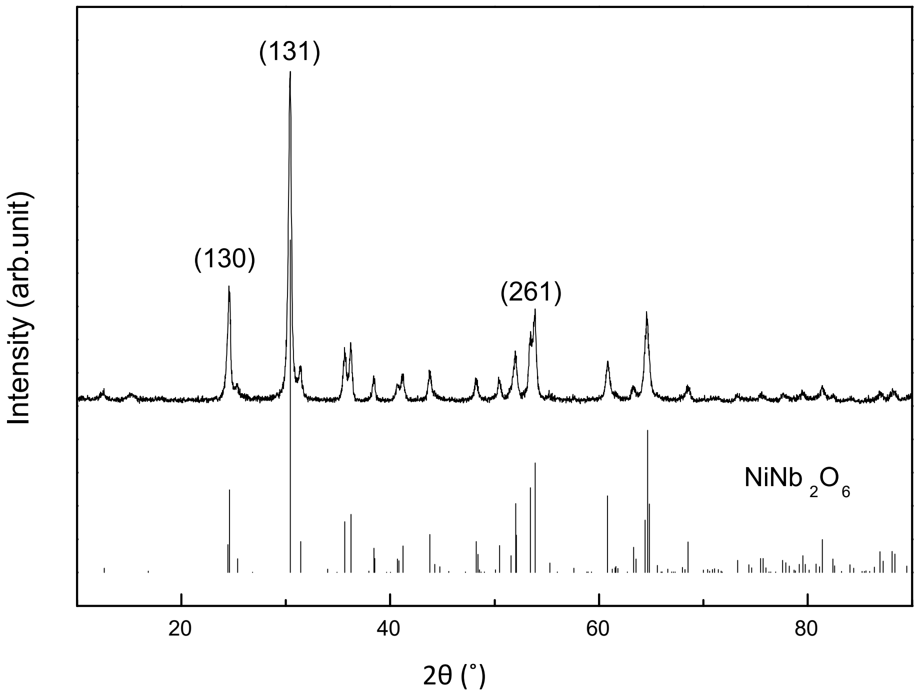


Fig.6 The XRD pattern of $NiNb_2O_6$ powders prepared by sol-gel method at 700 ˚C for 2h

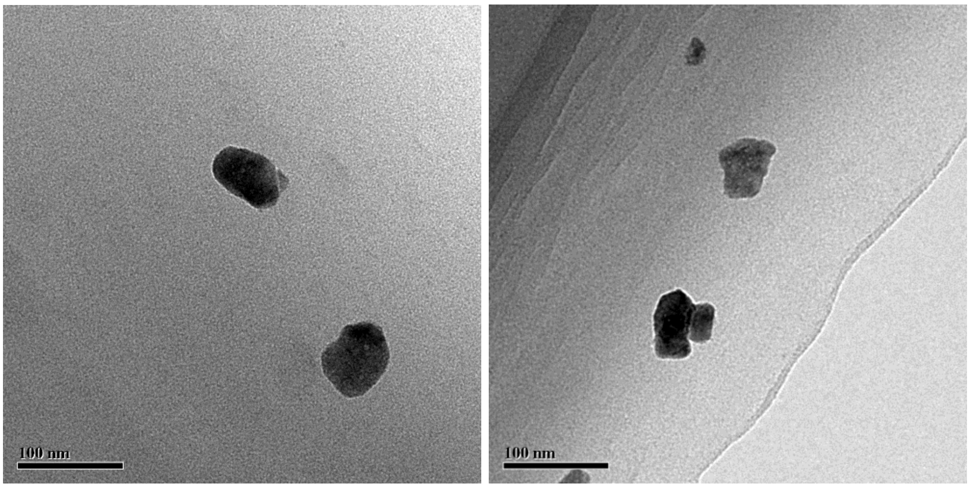


Fig.7 TEM view of $NiNb_2O_6$ powder particles prepared by sol-gel method at 700 ˚C for 2h

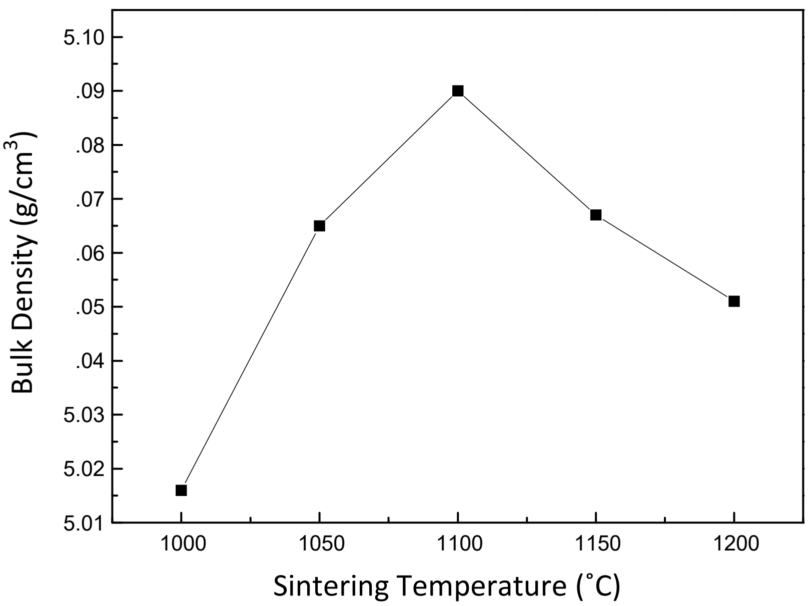
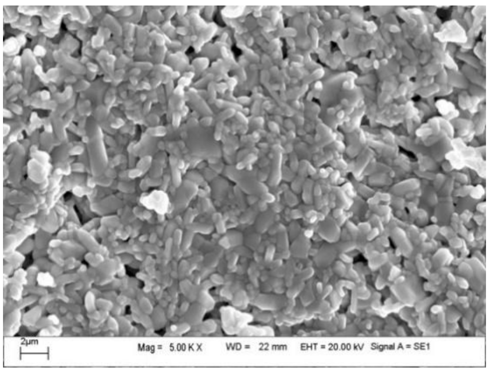
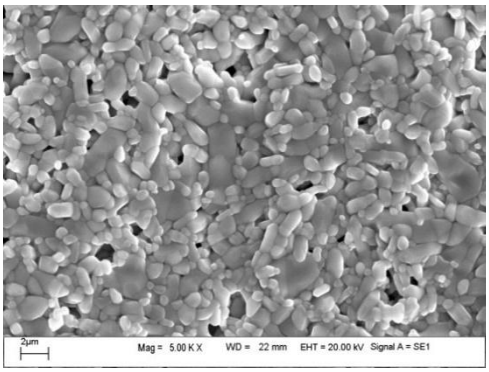


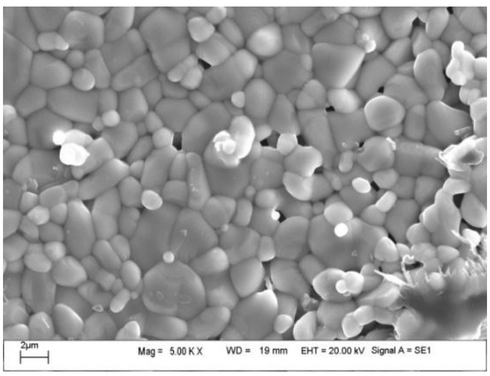
Fig.8 Densities of $NiNb_2O_6$ ceramics bulks prepared by sol-gel method sintered at 1000-1200℃



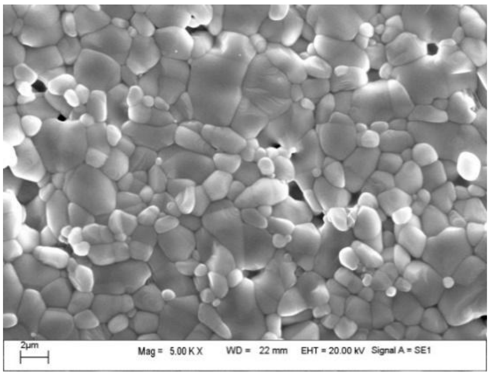
（a）1000 ℃



（b）1050℃



（c）1100℃



（d）1150℃

Fig.9 SEM surface patterns of $NiNb_2O_6$ ceramics prepared by sol-gel method sintered at 1000-1200 ˚C for 2 h

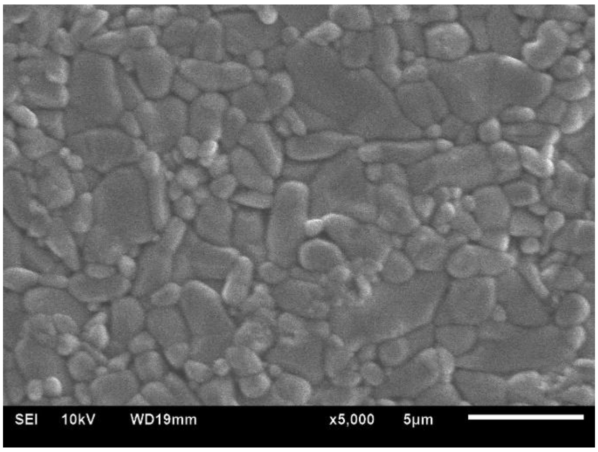


Fig.10 SEM surface patterns of $NiNb_2O_6$ ceramics prepared by sol-gel method sintered at 1100℃ for 8 h

Table 1 Comparison of Available Microwave Results for $NiNb_2O_6$

Compound	ε_r	$Q\times f$(GHz)	τ_f (ppm/˚C)	f_r (GHz)	Sintering	Density	Reference
$NiNb_2O_6$	22.6	40100	-38.0	10	1150 ˚C/2h	>95%	Lee et al. [6]
$NiNb_2O_6$	20.7	19800	-33	9	1300 ˚C/2h	99.8%	Liou et al. [7]
$NiNb_2O_6$	21.0	19300	-71.3	6.55	1200 ˚C/2h	94.5%	Pullar et al. [20]
$NiNb_2O_6$	23.6	18900	-62	4.36	1300 ˚C/4h	98%	Butee et al. [21]
$NiNb_2O_6$	22.1	37500	-30.4	6-8	1275 ˚C/2h	>99.5%	

Table 2 Microwave properties of $NiNb_2O_6$ ceramics with different amount of CuO additions

CuO Content	Sintering	Density(g/cm^3)	ε_r	$Q\times f$(GHz)	τ_f(ppm/˚C)
1.0 wt.%	1200 ˚C/2h	5.484	21.54	28896	-28.1
1.5 wt.%	1150 ˚C/2h	5.484	21.59	27506	-28.5

2.0 wt.%	1100 ˚C/2h	5.486	21.31	25704	-27.7
2.5 wt.%	1050 ˚C/2h	5.499	21.45	23531	-27.9
3.0 wt.%	1050 ˚C/2h	5.502	21.47	19282	-29.4

Table 3 Microwave properties of $NiNb_2O_6$ ceramics prepared by sol-gel method

Compound	ε_r	Q×f(GHz)	τ_f(ppm/˚C)	f_r (GHz)	Sintering	Density	Reference
$NiNb_2O_6$	19.69	11149	-27.3	6-8	1100 ˚C/2h	90.4%	
$NiNb_2O_6$	4.8-6	9000	NA	14-18	900 ˚C/6h	66.7%	Satapathy et al. [22]