Sintering and dielectric properties of perovskite SrTaO$_2$N ceramics

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Single-phase perovskite oxynitride SrTaO$_2$N ceramics were prepared by pressureless sintering at 1400 °C under a nitrogen atmosphere, using 5 wt% of SrCO$_3$ or La$_2$O$_3$ as a sintering additive. In contrast, SrTaO$_2$N bulks without additives contained TaC and Ta$_3$N$_5$ impurities. Sintering in nitrogen led to oxygen/nitrogen deficiencies in the oxynitride, while post annealing in flowing ammonia was effective to eliminate the anion vacancies. The introduction of additives significantly improved the sinterability of SrTaO$_2$N ceramics. A relative density of >90% in the bulk was achieved with a highly dense microstructure with smaller grain sizes. The SrTaO$_2$N bulk with SrCO$_3$ additive exhibited superior dielectric performance with a high relative permittivity ($\varepsilon_r = 1.0 \times 10^4$) and dissipation factor ($\tan \delta = 0.039$) at a frequency of 1 MHz.

Keywords: Perovskite oxynitrides; Sintering; Dielectric property; Sintering additives

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Introduction

High-permittivity dielectric materials play a significant role in microelectronics, due to their numerous technological applications such as capacitors and memory devices. To date, the most widely used dielectric ceramics are lead-based perovskite ferroelectrics or relaxor oxides, e.g., Pb(Zr,Ti)O₃ (PZT) and Pb(Mg₁/₃Nb₂/₃)O₃ (PMN) [1,2]. However, the toxicity of lead and its high vapor pressure during sintering have led to a demand for alternative environmentally friendly lead-free dielectric materials.

Recently, perovskite transition metal oxynitrides with the general formula of \(AB(O,N)_3\) have drawn much attention due to their superior dielectric properties [3-7]. Within the oxynitride family, samples of the SrTaO₂N ceramic were reported to possess large dielectric permittivity of ca. 2900 with moderate temperature dependence over the range of 180-300 K [3]. To date, the origin for this dielectric behavior has been studied on the basis of structural investigations [3,8,9]. It is suggested that the \textit{cis} arrangement of nitrogen in TaO₄N₂ octahedra may induce tilting of each octahedron [9]. In SrTaO₂N with statistically-averaged centrosymmetric \(I4/mcm\) space group, the coupling of anion ordering with octahedral tilting could lead to structural arrangements that possess a local dipole, which displays a similarity to the polar nanoregions (PNRs) in relaxor ferroelectrics [10]. Accordingly, the nonzero polar momentum units in SrTaO₂N result in high dielectric permittivity under an applied electric field. SrTaO₂N ceramics with large permittivity are stable in air, water and even in
One of the problems pertaining to dielectric oxynitride ceramics is the difficulty in fabricating high-density ceramics. There are only a few reports on dielectric oxynitrides [3-7], especially in the ceramic form [3]. The reported SrTaO$_2$N bulks have considerable porosity (~45%) when sintered by means of a conventional method, which is far from ideal for assessment of the inherent dielectric properties, because these measurements are always affected by the grain interfaces. Therefore, the densification of SrTaO$_2$N ceramics is necessary for fundamental characterization and also for practical applications, and should therefore be subject to further careful investigation. Hot pressing, hot isostatic pressing and spark plasma sintering are often used to achieve the densification of ceramics. However, these techniques are expensive when applied to mass production in commercial applications. Therefore, we have considered pressureless sintering with certain additives as a far more practical solution. The sintering of dielectric perovskite oxides can be promoted by the addition of A-site donor dopants because the densification kinetics is enhanced by the subsequently-induced A-site defects [11-13]. Meanwhile, during the sintering process of dielectric materials, incorporation of suitable additives could compensate the loss of easy-to-volatilize components. Yang et al. reported that the evaporation of PbO in PMN could be compensated by doping with SrO, resulting in the decrease of sintering temperature from 1200 to 800 °C as well as smaller grain sizes [14]. In addition, the dielectric properties can also be improved using specific dopants.
For instance, Kim et al. reported that the relative dielectric permittivity of La-modified PbTiO₃ with A-site vacancies was an order of magnitude larger than those of relaxor ferroelectrics [15]. Meanwhile, for PZT ceramics the isovalent substitution of Sr²⁺ for Pb²⁺ at the A-site led to a high dielectric constant of 1.0-1.3×10³ due to the increased tetragonality [16]. Accordingly, SrCO₃ and La₂O₃ can be considered as potential sintering additives for the development of dense SrTaO₂N bulks with high permittivity.

In the present work, the pressureless sintering, microstructure, and dielectric performance of SrTaO₂N ceramics with and without additives was investigated. To the best of our knowledge, this is the first report on the sintering behavior of SrTaO₂N with resulting high relative density.

**Experimental Procedure**

The perovskite oxynitride SrTaO₂N was synthesized by ammonolysis of precursors obtained from a citrate route [17]. Stoichiometric amounts of SrCO₃ (Wako Pure Chemicals, 99.9%) and TaCl₅ (Sigma-Aldrich, 99.99%) powders were dissolved in 60 mL of anhydrous ethanol, in which citric acid (Wako Pure Chemicals, 98.0%) was added as a complexing agent. The amount of citric acid was equimolar to that of the Ta ions. The solution was heated and stirred at 150 °C to promote polymerization. The resultant viscous gel was then pre-fired at 350 °C for 1 h, which resulted in brownish amorphous oxide precursors. The ammonolysis
reaction was carried out in flowing ammonia (Sumitomo Seika Chemicals, 99.9%, 50 mL/min) at 1000 °C with a heating rate of 5 °C/min, then held for 12 h before cooling to room temperature.

The as-nitrided SrTaO2N powders were mixed with 5 wt% SrCO3 or La2O3 (Sigma-Aldrich, 99.9%; fired at 970°C for 4 h prior to use) as an additive. Our preliminary experiments indicated that the sintered density and dielectric permittivity of SrTaO2N ceramics increased significantly when the additive amount was 5 wt% but further excessive addition had negative effects. These powders were die-pressed into 7 mm diameter and 2 mm thick disks, followed by cold isostatic pressing (CIP) at 150 MPa. SrTaO2N and SrTaO2N with SrCO3 or La2O3 additive (hereafter denoted as SrTaO2N-Sr and SrTaO2N-La, respectively) were placed in a BN crucible and sintered at 1400 °C for 3 h under a nitrogen pressure of 0.2 MPa using a graphite furnace (High Multi 500; Fuji Dempa Kogyo). The sintered bulks were then annealed at 1000 °C for 12 h in flowing ammonia. This post-annealing procedure was necessary to obtain highly resistive bulks for reliable dielectric property measurements.

The SrTaO2N products were characterized using X-ray diffraction (XRD) with Cu Ka radiation (Rigaku, Ultima IV). The data were collected over the angular range of 10-120° with a step size of 0.02°. A counting time of 2.0 s per step was used for the phase identification, while a longer counting time of 9.0 s per step was required for the structural refinement. The
lattice parameters of the SrTaO$_2$N products, including as-nitried powder, as-sintered and post-annealed bulks (in powder form), were refined by the Rietveld method using RIETAN2000 software [18]. The bulk density was measured by both the geometric and Archimedes methods. Scanning electron microscopy (SEM; Jeol JSM-6390LA) was used to observe the microstructure of the ceramics. The post-annealed samples were polished with a 4000-grit sandpaper (particle size: 3 µm) to obtain parallel surfaces and then coated with silver paste to act as electrodes. The dielectric properties were measured with impedance analyzers (Yokogawa-Hewlett-Packard, 4274A; Hewlett-Packard, 4285A) in the frequency range from $10^2$ to $10^6$ Hz at room temperature. The impedance plots were fitted and extrapolated using the Cole-Cole empirical relation [19].

Results and discussion

Figure 1 shows XRD patterns for the SrTaO$_2$N powder, as-sintered SrTaO$_2$N and post-annealed SrTaO$_2$N bulks with and without sintering additives. Compared with the as-nitried powder, a small amount of TaC impurity was detected in the as-sintered SrTaO$_2$N bulk without any additive. The TaC impurity is likely to form when excess Ta is combined with organic residue due to the loss of SrO from SrTaO$_2$N during sintering. To minimize the amount of organic residue in the precursor powder, the combustion at 600 °C was examined, but this procedure did not lead to a reduced amount of TaC in the as-sintered SrTaO$_2$N bulk.
In spite of the high melting temperature, SrO loss may occur in high-temperature processing, as reported in SrTiO₃, SrZrO₃ and Sr₂RuO₄ [20-22]. The diffraction peaks of TaC impurity disappeared in the as-sintered SrTaO₂N-Sr and SrTaO₂N-La bulks because Sr²⁺ or La³⁺ from the additives compensates the Sr loss, which inhibits the appearance of this secondary phase.

It was found that all the sintered bulks changed from brown to dark gray, which indicates reduction of the tantalum associated with anion deficiencies in the crystal lattice. Post-annealing in an ammonia atmosphere was necessary because heat treatment in air/oxygen leads to oxidation and decomposition of the oxynitrides [23]. After the annealing process, TaC and Ta₃N₅ impurities were observed in SrTaO₂N without additives, while both SrTaO₂N-Sr/La samples were phase-pure perovskites. At high temperatures, the TaC impurity was oxidized and then nitrided in ammonia to form Ta₃N₅ in the post-annealed SrTaO₂N bulk [24], as indicated in Fig. 1(e). We tentatively believe that a trace amount of oxygen from the not-fully airtight furnace is the oxidizing agent. It can be assumed that a part of TaC was not oxidized and still remained as it was, due to the limited amount of oxygen.

The XRD patterns were refined to obtain the lattice parameters of the SrTaO₂N products, including the as-nitrided powder, as-sintered and post-annealed bulks, based on the tetragonal I₄/mcm space group [9]. A summary of the crystallographic features is given in Table 1. Rietveld refinement plots for the SrTaO₂N products are given in the Supporting Information. A trace amount (0.43 wt%) of SrCl₂·2H₂O impurity was observed in the as-nitrided powder.
The $c$ parameter of the tetragonal lattice was slightly reduced in the as-sintered bulk. Recovery of the $c$ value was then achieved by annealing in ammonia, which was accompanied by the color change from dark gray to brown. The lattice parameter variations upon sintering/post-annealing were reproducible in different sample batches. It is thus reasonable to interpret the lattice parameter variations as a hint for the changing anion vacancy contents upon sintering/post-annealing. Anion vacancies are assumed to be introduced during the high temperature sintering, because the thermal reduction environment in the graphite dies led to oxygen/nitrogen deficiency. However, the anion vacancies should be mainly ascribed to a loss of nitrogen because the reported standard free energy of formation is much lower for metal nitrides than for the corresponding oxides [25]. Compared with transition metal oxides, the relatively low stability of the associated nitrides, related to weaker bonding, has already been understood through theoretical calculations [26]. Therefore, the loss of nitrogen is more likely to occur by heating SrTaO$_2$N at 1400 °C, which results in shrinking of the crystal lattice due to its covalent nature. The formation of nitrogen vacancies may involve the release of N$_2$ gas, as suggested by the previous thermogravimetric study [23]. Similar results have been documented in nonstoichiometric metal nitrides, such as TiN$_x$ and $\delta$-NbN$_x$ [27].

The relative densities of the post-annealed SrTaO$_2$N, SrTaO$_2$N-Sr and SrTaO$_2$N-La bulks were 70, 93, and 90%, respectively. The SrTaO$_2$N bulk without additives could not be well sintered, while a high density was achieved at 1400 °C by the addition of 5 wt% SrCO$_3$ or
La$_2$O$_3$. The fracture surface morphology did not change with the post-annealing, but was changed by the addition of sintering additives, as shown by the SEM micrographs in Fig. 2. After the annealing process, densification and smaller grain size was achieved with the sintering additives. The SrTaO$_2$N bulk without additives was porous with large voids between the grains, indicating insufficient densification. In contrast, the SrTaO$_2$N ceramics sintered with additives were considerably dense and consisted of a well-developed microstructure. The reason lies in that the SrO (from SrCO$_3$) or La$_2$O$_3$ could react with Sr-deficient SrTaO$_2$N (Sr$_{1-\delta}$TaO$_2$N) to promote the efficient reaction sintering and further densification, while no obviously segregated compounds such as strontium tantalum oxides and lanthanum tantalum oxides were observed in the microstructures. Take the SrTaO$_2$N-La as an example, La$^{3+}$ could partly substitute the A-site Sr$^{2+}$, due to their similar ionic radii [28]. The entropy gain by (Sr$_{1-x}$La$_x$)Ta(O,N)$_{3-\delta}$ solid solution formation are beneficial to mass transport and enhancement of the densification process. The loss of SrO offered an opportunity for the reaction sintering after the incorporation of additives, which was found to be necessary to obtain relatively dense SrTaO$_2$N bulks. It is noteworthy that smaller grains were obtained in the SrTaO$_2$N-Sr/La ceramics. As the densification occurs, Sr$^{2+}$/La$^{3+}$ appears not only in the bulk regions but also near grain boundaries due to the relatively high additive level, resulting in the inhibition of grain growth as well as the smaller grain sizes [29-31].

All the as-sintered SrTaO$_2$N ceramics exhibited semiconductive behavior, typically with
several thousand ohms at room temperature. The successive heat treatment in ammonia was found to be essential to recover the dielectric properties which accompany resistance values several order of magnitudes larger than those of the as-sintered bulks. Figure 3 shows the dielectric constant ($\varepsilon_r$) and dielectric loss (tan $\delta$) as a function of frequency for the post-annealed SrTaO$_2$N samples. Both the $\varepsilon_r$ and tan $\delta$ values decrease with increasing frequency. The SrTaO$_2$N bulk samples without additives have $\varepsilon_r$ values in the order of $1.8 \times 10^3$ at $10^2$ Hz, which is lower than that reported previously [3], despite its relatively higher density. It seems that the presence of secondary phases such as TaC and Ta$_3$N$_5$ is responsible for the lower dielectric constant. In contrast, the addition of SrCO$_3$ or La$_2$O$_3$ resulted in a significant increase in the dielectric constant, which was enhanced to $1.6 \times 10^4$. This value is several times higher than the permittivity (~2900) reported by Kim et al. [3]. The permittivity of SrTaO$_2$N-Sr/La ceramics is comparable to those reported for ferroelectric and relaxor oxides at their Curie temperatures [32]. Another intriguing feature of SrTaO$_2$N with additives is the weak dependence of the dielectric constant on frequency, e.g., SrTaO$_2$N-Sr had high relative permittivities of $1.7 \times 10^4$, $1.3 \times 10^4$, and $1.0 \times 10^4$ at frequencies of $10^2$, $10^4$, and $10^6$ Hz, respectively. The dielectric constants of the SrTaO$_2$N ceramics are sensitive to density. Enhanced densification is beneficial to obtain desirable dielectric properties. Accordingly, the highly densified SrTaO$_2$N-Sr/La bulks with uniform grain structure also exhibit much higher room-temperature dielectric constants than SrTaO$_2$N without additives.
It is worthy to note that the dielectric loss for the present SrTaO$_2$N bulk without additives is comparable to that obtained for a LaTiO$_{x}$N$_y$ thin film [5], which is an order of magnitude lower than that reported for the SrTaO$_2$N bulk in the previous work [3]. It has been indicated that the dielectric loss decreases significantly as the sample sintering improves, while the electrical conductivity remains low [3]. The present SrTaO$_2$N-Sr/La bulks has a significantly improved grain microstructure, but the dielectric loss is relatively high. The tan $\delta$ values are all approximately 0.04 at 1 MHz, which is still higher than that required for practical applications. The number of grain boundaries was increased in the SrTaO$_2$N-Sr/La bulks due to the smaller grain size, leading to an accumulation of the space charge [33]. This may be one reason for the high dissipation factor. In addition, the formation of lattice defects within the crystal also affects the dielectric loss. The SrTaO$_2$N ceramics with additives are well densified; therefore, it is likely that the anion deficiencies were not completely removed during the post annealing process. A small amount of anion vacancies (or tetravalent tantalum) is sufficient to cause a large increase in the dielectric loss [34]. Such Ta$^{4+}$ species with paramagnetic moments should be detectable by means of electron paramagnetic resonance and be subject to further exploration. A similar result was also reported for BaTi$_4$O$_9$/Ba$_2$Ti$_9$O$_{20}$-based ceramics, where even a low defect density caused by reductive heat treatment significantly increased the dielectric loss [35].

Complex impedance plots of the SrTaO$_2$N and SrTaO$_2$N-Sr/La bulks are shown in Fig. 4.
To interpret these impedance spectra, an equivalent circuit model consisting of two parallel RC elements connected in series was assumed. These two serial RC units represent the bulk ($R_b$) and grain boundary ($R_{gb}$) contributions [3]. For the SrTaO$_2$N-Sr/La bulks, the first semicircle corresponding to the bulk contribution was clearly observed, whereas the low frequency ($<10^2$ Hz) semicircle related to the grain boundaries was not evident due to the instrumental limitations of the experimental range. It has been recognized that the choice of the electrical contact type sometimes influences the overall electrical behaviors [36]. Nevertheless, effects of the electrodes are assumed to be insignificant, since different contact types (here Ag-pasted and Pt-sputtered electrodes) resulted in essentially similar results, as presented in Supporting Information. The $R_b$ value, assigned as the intercept of the $Z'$-axis in the Cole-Cole plot, is quite different among the SrTaO$_2$N and SrTaO$_2$N-Sr/La bulks. The impedance spectrum of the SrTaO$_2$N bulk without additives did not form a semicircle, which is indicative of a much higher value for $R_b$. The lower bulk resistance of the SrTaO$_2$N-Sr/La bulks can be attributed to the presence of residual oxygen/nitrogen deficiencies. However, for the SrTaO$_2$N bulk without additives, the anion vacancies are easily removed from the porous ceramic body to reach cation-anion stoichiometry. Accordingly, a more resistive bulk gave rise to a large semicircle in the impedance spectrum. For dielectric materials, low values of electrical resistance often lead to high dielectric loss; the increase in tan δ with the SrCO$_3$ or La$_2$O$_3$ additives could be related to the smaller bulk resistance.
In the present work, the dielectric permittivity of the dense SrTaO$_2$N ceramics is extremely high and comparable with the giant $\varepsilon_r$ reported for the rocksalt-type Li, Ti co-doped NiO [37]. In addition, the thermal stability of SrTaO$_2$N can be ensured up to 480 °C in air [23], which is much higher than the Curie temperature of PZT [1], where a huge variation of permittivity occurs. Although the method of densification has been developed for the dielectric SrTaO$_2$N ceramics, the dielectric loss should be further reduced by eliminating anion vacancies in a considerably improved microstructure.

**Conclusion**

Pressureless sintering was applied to fabricate dielectric oxynitride SrTaO$_2$N ceramics using sintering additives. Both SrCO$_3$ and La$_2$O$_3$ were effective to achieve single-phase SrTaO$_2$N bulks with relative densities higher than 90%. The incorporation of sintering additives in the SrTaO$_2$N ceramic led to a well-developed microstructure with smaller grain sizes. The residual porosity of SrTaO$_2$N ceramics was necessary to remove anion vacancies during the post-annealing treatment in ammonia to realize smaller dielectric losses. The room temperature $\varepsilon_r$ values of SrTaO$_2$N-Sr and SrTaO$_2$N-La were $1.7\times10^4$ and $1.6\times10^4$ at 100 Hz, respectively; however, in contrast, the SrTaO$_2$N without additives exhibited lower dielectric permittivity of approximately $1.8\times10^3$ due to the much higher porosity, the presence of secondary phases as well as the possible grain boundary effects.
Acknowledgement.

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Figure Captions:

Fig. 1. XRD patterns of (a) as-nitrided SrTaO$_2$N powder, (b) as-sintered SrTaO$_2$N bulk, (c) as-sintered SrTaO$_2$N-Sr bulk, (d) as sintered SrTaO$_2$N-La bulk, (e) post-annealed SrTaO$_2$N bulk, (f) post-annealed SrTaO$_2$N-Sr bulk, and (g) post-annealed SrTaO$_2$N-La bulk.

Fig. 2. Scanning electron micrographs of the fracture surfaces of post-annealed (a) SrTaO$_2$N without additives, (b) SrTaO$_2$N-Sr, and (c) SrTaO$_2$N-La bulks.

Fig. 3. Relative dielectric constants ($\varepsilon_r$) and dielectric losses ($\tan \delta$) of post-annealed (a) SrTaO$_2$N without additives, (b) SrTaO$_2$N-Sr, and (c) SrTaO$_2$N-La bulks as a function of frequency.

Fig. 4. Impedance spectra for the post-annealed SrTaO$_2$N ceramics measured at $10^2$-$10^6$ Hz.
Table 1.
Summary of structural refinement details for the as-nitried SrTaO$_2$N powder, as-sintered and post-annealed SrTaO$_2$N products.

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