

Structure, morphology, and microwave dielectric properties of SmAlO₃ synthesized by stearic acid route

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Abstract: A rapid and facile approach was developed for the synthesis of ultrafine SmAlO₃ powders through the combustion of stearic acid precursors. The obtained products were characterized by typical techniques including X-ray diffraction (XRD), Fourier Transform Infrared (FT-IR), thermogravimetric and differential thermal analysis (TG-DTA), scanning electron microscopy (SEM), and transmission electron microscopy (TEM) to analyze the phase composition and microstructure. The dielectric characteristics of SmAlO₃ microwave ceramics, using the as-obtained products as original materials, were also studied. Compared with the conventional solid-state reaction method, the synthesis temperature was dramatically reduced to 750 °C. The large-size sheet structure was composed of a number of micro/nano-scale crystallites, which were mostly irregular in shape due to the mutual growth and overlapping shapes of adjacent grains. The SmAlO₃ ceramics with high density and uniform microstructure were obtained after sintering at 1500 °C for 4 h due to the favorable sintering activity of the as-synthesized powders. In addition, desired dielectric properties at microwave frequencies (dielectric constant $\epsilon_r = 20.22$, quality factor $Q \cdot f = 74110$ GHz, and a temperature coefficient of resonant frequency $\tau_f = -74.6$ ppm/°C) were achieved.

Keywords: stearic acid precursor process; nanoparticle; perovskite; microwave ceramics

1 Introduction

In the high-frequency region, microwave dielectric ceramics are regarded as suitable materials for application in modern communication systems because they exhibit good properties such as high ϵ_r , high $Q \cdot f$, and near-zero τ_f [1–4]. Rare-earth aluminate ceramics with perovskite

structure are an example of such materials, and therefore have attracted a great deal of interest in recent decades. Cho *et al.* [5] reported the ϵ_r and $Q \cdot f$ values of 20.4 and 65000 GHz for SmAlO₃ ceramics respectively, prepared by solid-state reaction method, indicating a potential application in wireless communication as dielectric resonators, filters as well as dielectric antenna. High dielectric constant and $Q \cdot f$ can miniaturize the size and ensure a favorable frequency selectivity in these microwave devices, respectively. SmAlO₃ can also find an extensive application in superconducting microwave devices due

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to its high $Q \cdot f$ value, favorable lattice matching, and excellent thermal expansion matching. Moreover, it exhibits much lower cost than NdAlO_3 whose microwave dielectric properties are similar to those of SmAlO_3 since the raw material Nd_2O_3 is rather expensive.

The large negative τ_f of SmAlO_3 ceramics, which is a shortcoming for practical applications, can be easily adjusted to a near-zero value by ion doping or constructing multiphase materials. A variety of novel SmAlO_3 -based ceramic systems have been explored in recent years. For example, Suvorov *et al.* [6] reported a new ceramics system with satisfactory microwave dielectric properties ($\epsilon_r = 41$, $Q \cdot f = 42000$ GHz, and $\tau_f = -18$ ppm/°C) by mixing 0.65CaTiO_3 and 0.35SmAlO_3 and sintering them at 1450 °C. Soon afterwards, a similar competitive composition of 0.66CaTiO_3 – 0.34SmAlO_3 in this system was developed by Xu *et al.* [7], displaying the best ϵ_r and $Q \cdot f$ of 42.49 and 46069.86 GHz respectively when sintered at 1500 °C for 4 h. Comprehensive dielectric properties with $\epsilon_r = 58.3$, $Q \cdot f = 18800$ GHz, and $\tau_f = 2.3$ ppm/°C, could be obtained in the ceramic $0.75(\text{Sr}_{0.3}\text{Ca}_{0.427}\text{Nd}_{0.182})\text{TiO}_3$ – 0.25SmAlO_3 after being sintered at 1500 °C for 4 h [8]. Solomon [9] revealed that the 0.1SmAlO_3 – $0.9\text{Ba}(\text{Zn}_{1/2}\text{Nb}_{2/3})\text{O}_3$ composite, sintered at 1400 °C for 4 h, had $\epsilon_r = 37$, $\tau_f = 25$ ppm/°C, and high $Q \cdot f$. Compared to other ceramics, these novel SmAlO_3 -based microwave dielectric ceramics are attractive candidates for the base stations of mobile communications, in which higher $Q \cdot f$ (≥ 40000 GHz) and ϵ_r ($25 \leq \epsilon_r \leq 50$) values are demanded. Moreover, the composite could find a potential application in optoelectronics communication. In addition, improved electrical properties were achieved by doping a small amount of SmAlO_3 into $(\text{K}_{0.44}\text{Na}_{0.52}\text{Li}_{0.04})(\text{Nb}_{0.91}\text{Ta}_{0.05}\text{Sb}_{0.04})\text{O}_3$ lead-free ceramics [10]. Based on these attempts, it is concluded that SmAlO_3 is a favorable competitive temperature compensation material, which is similar to most microwave dielectric ceramics with large negative τ_f .

Nevertheless, an outstanding drawback, namely ultrahigh sintering temperature, still prevents wide-scale industrial application in microwave components for SmAlO_3 -based ceramics due to the poor reactive nature of the powders obtained from the conventional solid-state reaction method. This method generally requires a prolonged preparation cycle, and thus results in compositional deviation and particle coarsening of final products, as well as excessive energy consumption.

Especially, the SmAlO_3 ceramics prepared by the solid-state reaction route require very high calcination and sintering temperatures of about 1400 and 1650 °C to densify, respectively [11]. Recently, tremendous efforts have been devoted to enhancing the sintering performance of ceramics, and a variety of synthesis approaches have been carried out, such as co-precipitation method, molten salt synthesis, hydrothermal synthesis, sol–gel route, and polymer complex method [12–17]. In the chelating sol–gel route, which is commonly used to prepare a variety of nanometer complex oxides, many metal ions are inclined to generate precipitation due to their strong hydrolysis ability in aqueous solution, and thus lead to their inhomogeneous dispersion. Stearic acid can be used as a solvent in the sol–gel route because of its low melt point of about 70 °C. Correspondingly, the hydrolysis-precipitation phenomenon can be avoided due to the absence of water solvent. In fact, stearic acid has a dual role by acting as not only a complexing agent but also a surface active agent. The carboxylic acid group in its structure can strongly complex with most metal ions. Various metal ions can be dispersed easily at the molecular/atomic level in molten stearic acid by a liquid-state mixing route. Each component can be homogeneously mixed even in the resultant products after combustion to remove organic substances and therefore remarkably reduce the synthesis temperature of complex oxides. As a non-hydrolytic method, the stearic acid method has attracted much attention in recent years for the synthesis of several oxide compounds. However, there are no reports on the synthesis of nanocrystalline SmAlO_3 with this method for now. Herein, we synthesized pure phase SmAlO_3 nanosized powders with high reaction yield and synthetic reproducibility by the stearic acid method. The phase composition, microstructure, and morphology of the obtained nanoparticles were analyzed and the microwave dielectric properties of the ceramics created by the nanoparticles were also discussed.

In addition, in our previous work, we investigated the synthesis of SmAlO_3 powders by citrate precursor and EDTA precursor methods [18,19], exhibiting specific advantages in the reduced sintering temperature and enhanced microwave dielectric properties. The present work can provide an important supplement to wet-chemistry methods for SmAlO_3 ceramics. Also, it presents a cost advantage due to the utilization of inexpensive metal salts and stearic acid as starting materials.

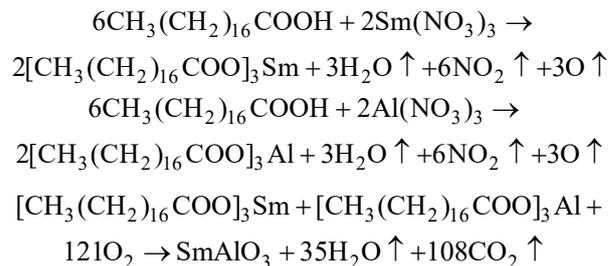
2 Experimental procedures

2.1 Powder synthesis

SmAlO₃ powders were synthesized according to a synthetic procedure, as shown in Fig. 1. In the experiments, analytical grade reagents samarium oxide (Sm₂O₃), aluminum nitrate hexahydrate [Al(NO₃)₃·9H₂O], and stearic acid (C₁₇H₃₅COOH) were chosen as raw materials. In light of its hygroscopic nature, Sm₂O₃ powders must be pre-fired at 900 °C for 4 h before weighing procedure. First of all, 0.005 mol Sm₂O₃ was dissolved in diluted nitric acid to form a transparent solution under magnetic stirring at 80 °C. In addition, 30 g stearic acid was weighed and then heated slowly to melt completely at 75 °C in a beaker, and then 0.01 mol Al(NO₃)₃·9H₂O was slowly added and dissolved in the molten stearic acid until a clear solution was obtained with continuous stirring. Next, the Sm(NO₃)₃ solution was added dropwise into the molten stearic acid in the same manner. By increasing the heating temperature to 100 °C for a sufficient period of time, the unnecessary water in the mixture was removed. After that, the mixture was continuously magnetic stirred and maintained at 130 °C until a uniform sol-like solution was produced, accompanied by the release of a large volume of reddish brown gases. By naturally cooling down to room temperature and drying for 12 h in an oven, a dried gel was formed. Subsequently, the dried gel was reheated to 230 °C for 2 h until it transformed into a black resinous precursor.

Finally, the SmAlO₃ powders were obtained after the precursor was calcined in air at various temperatures.

The suggested chemical reactions are as follows:



2.2 Samples preparation

The as-synthesized SmAlO₃ powders, along with a small amount of PVA binder, were granulated and then pressed into cylindrical green samples with a size of about Φ10 mm × 5 mm. Finally, these as-pressed cylinders were pressurelessly sintered for densification at 1350–1550 °C for 4 h.

2.3 Characterization and measurements

XRD patterns were collected using a powder X-ray diffraction analyzer (Bruker D8, Germany) with Cu Kα radiation at 2θ = 20°–80°. Thermal analysis of dried gel was employed on a Shimadzu DTG-60H instrument in a temperature range of 30–900 °C, in air with a heating rate of 10 °C/min. FT-IR spectra were obtained on a Nicolet 6700 FT-IR device by the KBr pellet method, ranging from 400 to 4000 cm⁻¹. FE-SEM was used to evaluate the morphology and microstructure by

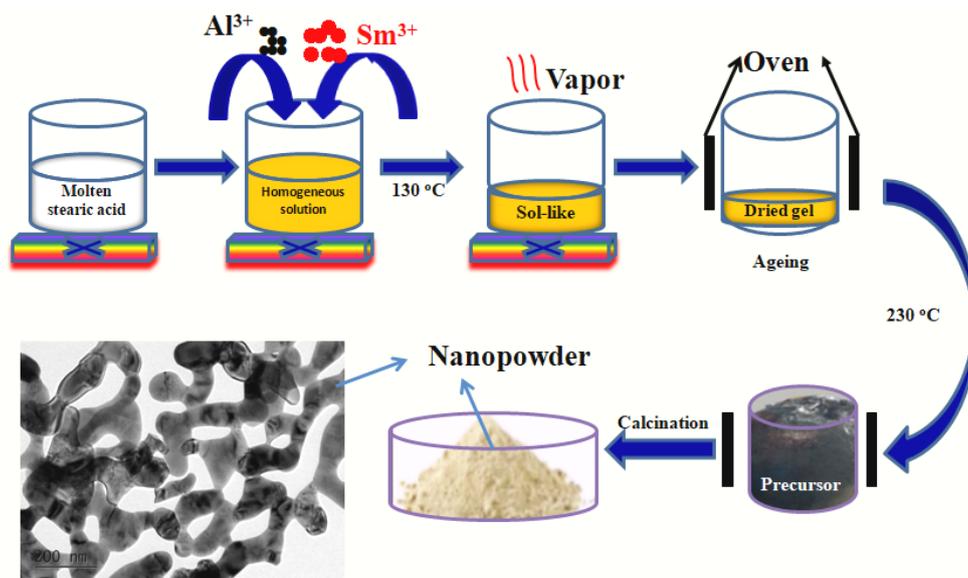


Fig. 1 Flow diagram of SmAlO₃ synthesized by the stearic acid method.

depositing a gold coating on the surface of samples. The particulate properties of synthesized powders were examined using transmission electron microscopy (JEOL-200). In accordance with the Archimedes method, the bulk densities of sintered ceramics were measured. Relative dielectric constant and dielectric loss of sintered microwave ceramics were evaluated using the Hakki–Coleman DR method by a vector network analyzer (Agilent 8722ET) [20,21]. τ_f was determined through Eq. (1):

$$\tau_f = \frac{f_{80} - f_{25}}{f_{25}(80 - 25)} \quad (1)$$

where f_{25} and f_{80} denote the resonant frequencies at 25 and 80 °C, respectively.

3 Results and discussion

3.1 Thermal decomposition process

In order to understand the synthesis process of SmAlO_3 , the thermal analysis including TG-DTA for the dried gel was examined, and the results are shown in Fig. 2. The endothermic peak in the DTA curve at 110 °C is due to the volatilization of residual water and the melting of gel, accompanied by a weight loss of about 13.3% between room temperature and 230 °C in the TG curve. When the temperature is over 230 °C, all other peaks are exothermic, which indicates that a series of oxidation and combustion reactions are involved in the thermal decomposition process. In the second step, a huge weight loss (43.9%) at the temperature range of 230–600 °C can be attributed to the combustion of organic substances in the gel, and the formation and decomposition of samarium and aluminum complexes. It can be observed from the DTA curve that there are four relatively strong exothermic peaks at 318, 388, 465, and 585 °C. Especially, the strongest peak at 585 °C may indicate the decomposition of samarium and aluminum complexes, and the formation of some amorphous phases. When the temperature exceeds 585 °C, no apparent weight loss is found. However, a weak exothermic peak at 683 °C appears in the DTA curve, indicating that a phase transition from an amorphous phase to a crystalline phase occurs. This means that the minimum crystalline temperature to synthesize SmAlO_3 powders by stearic acid method is approximately 683 °C.

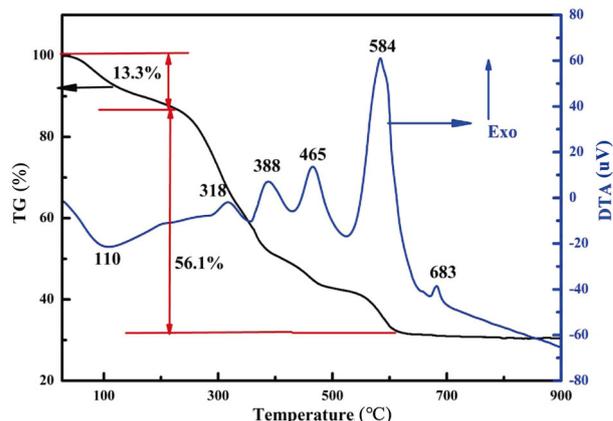


Fig. 2 TG and DTA curves of the dried gel obtained by the stearic acid method.

3.2 XRD patterns and FT-IR spectra analysis

Figure 3 displays the XRD patterns of SmAlO_3 powders after calcination from 750 to 900 °C for 2 h. After calcination at 750 °C for 2 h, the product exhibits favorable crystallinity, which can be characterized by some sharp diffraction peaks. These XRD peaks, corresponding to the reflections from the orthorhombic phase, are in strong agreement with the database in JCPDS # 71-1597 for SmAlO_3 with a perovskite structure. Further increasing the calcination temperature makes the XRD peaks of the powders stronger and stronger. Additionally, no impurity or other intermediate phase is observed in the investigated calcination temperatures. The XRD patterns of SmAlO_3 in our present work are similar to those of the nanopowders synthesized by the polymeric precursor method [19],

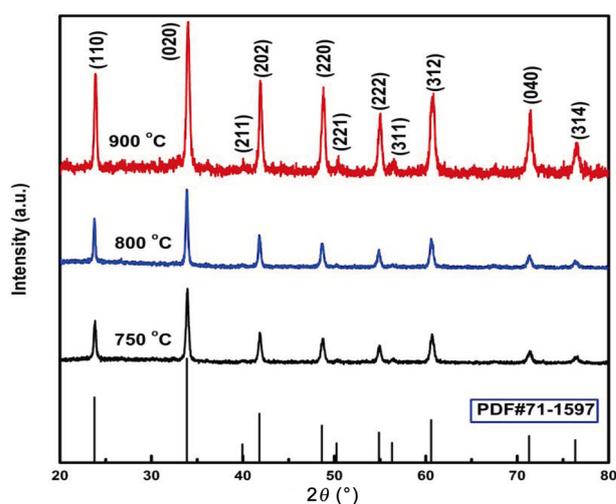


Fig. 3 XRD patterns of SmAlO_3 calcined at different temperatures for 2 h.

and the sol–gel route using malic acid as a complexing agent [22]. However, the synthesis temperature of the stearic acid method is about 750 °C, which is lower than those of other wet-chemistry routes (800 °C for the polymeric precursor method and 950 °C for the sol–gel technique route). This synthesis temperature for the solid-state reaction to obtain pure phase SmAlO₃ is as high as 1400 °C [5]. This evidence indicates that the stearic acid approach is a speedy and energy-saving route for the synthesis of ultrafine SmAlO₃ powders with high sintering activity.

In addition, the refined lattice parameters, cell volume, and crystallite size for the products derived from different synthesis approaches are calculated using the FULLPROF software package, and the Debye–Scherrer formula [23,24]. Table 1 summarizes these calculated results.

$$D = \frac{0.89\lambda}{\beta \cos\theta} \quad (2)$$

where D , λ , β , and θ are the average crystallite size, X-ray wavelength, full width at half maximum (FWHM) of the diffraction peak, and Bragg's angle, respectively. The calculated crystallite size is 29 nm for the SmAlO₃ powders calcined at 750 °C, while taking the diffraction angle $2\theta = 33.9^\circ$. It is seen that the cell volume of SmAlO₃ phase obtained by wet-chemistry methods, is relatively smaller than that of the conventional solid-state reaction method.

In order to confirm the structural changes between the precursor and the powders that underwent calcination processes, FT-IR analysis was done and the results are shown in Fig. 4. Small absorption bands at about 2920 cm⁻¹, 1630 cm⁻¹, and 1067 cm⁻¹ in the precursor after heat treatment at 230 °C reveal that a small amount of organic substance still exists. When increasing the temperature to 750 °C, no obvious absorption bands of organic substances are observed, as shown in Fig. 4(b). Meanwhile, the absorption bands in the range of 400–700 cm⁻¹ get stronger,

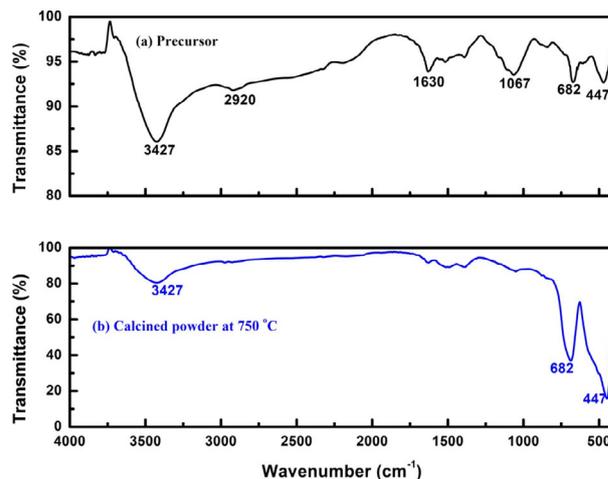


Fig. 4 FT-IR spectra of the dried gel and the powders calcined at 750 °C.

which is attributed to the formation of MO₆ octahedra (M = Sm/Al). In addition, a broad band centered at 3427 cm⁻¹ is assigned to the adsorbed water of these powders.

3.3 Morphology observation

Figure 5 shows the SEM image of SmAlO₃ powders calcined at 750 °C for 2 h without grinding. As displayed in Fig. 5, a large-size sheet structure is composed of a number of micro/nano-scale crystallites. To draw a reliable conclusion based on the above result, the transmission electron analysis was carried out and the corresponding TEM image is present in Fig. 6. The particle size estimated from the TEM image is about 70 nm, which is significantly larger than that calculated from the Debye–Scherrer formula. It is observed that nearly all crystallites are mostly irregular in shape due to the mutual growth and overlapped shapes of adjacent grains. This may be due to the variety of solution concentration and pH values. In a recent study, Zhang *et al.* [25] have found that the pH value has remarkable effects on the crystal structure, surface morphology, and microwave dielectric properties of

Table 1 Lattice parameters and average crystallite size of SmAlO₃ obtained using solid state (SmAlO₃-SS), polymeric precursor (SmAlO₃-PP), hydrothermal (SmAlO₃-H), and stearic acid (SmAlO₃-SA) techniques

Parameter	SmAlO ₃ -SS	SmAlO ₃ -H	SmAlO ₃ -PP	SmAlO ₃ -SA
Lattice parameter (Å)	$a = 5.2912$	$a = 5.2888$	5.2672	5.2834
	$b = 5.2904$	$b = 5.2784$	5.2779	5.2866
	$c = 7.4740$	$c = 7.4733$	7.4448	7.4707
Cell volume (Å ³)	209.22	208.63	206.96	208.66
Crystallite size (nm)	—	—	45	29 ($2\theta = 33.9^\circ$)

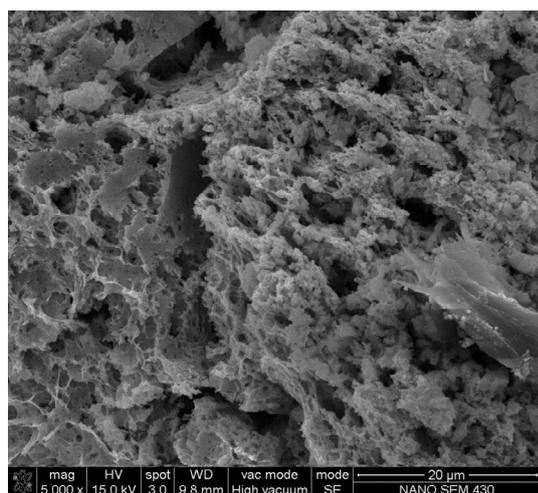


Fig. 5 SEM micrograph of SmAlO_3 powders calcined at $750\text{ }^\circ\text{C}$ for 2 h without grinding.

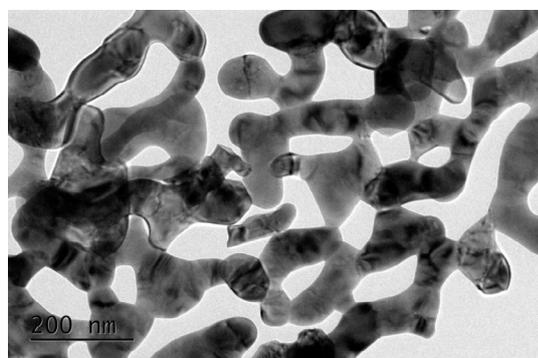


Fig. 6 TEM photos of SmAlO_3 powders calcined at $750\text{ }^\circ\text{C}$ for 2 h.

$\text{Bi}_{12}\text{TiO}_{20}$ ceramics fabricated by the sol–gel method. Therefore, it is of great necessity to synthesize fine particles with high reactive activity and favorable sintering activity in our future research.

3.4 Sintering of SmAlO_3

The powders synthesized by the stearic acid method are used as starting materials to prepare SmAlO_3 ceramics for the purpose of application as microwave dielectric materials. The synthesized powders were uniformly mixed with 5 wt% PVA solution, pressed into some bulk specimens, and sintered at $1350\text{--}1550\text{ }^\circ\text{C}$ for 4 h.

Figure 7 illustrates the bulk densities of SmAlO_3 ceramics sintered at different temperatures. At $1350\text{--}1425\text{ }^\circ\text{C}$, the bulk density drastically increases. But the low bulk densities indicate that the current SmAlO_3 ceramics are not dense. When the sintering temperature is over $1425\text{ }^\circ\text{C}$, the bulk density gradually increases until $1500\text{ }^\circ\text{C}$, and then it decreases. The maximum

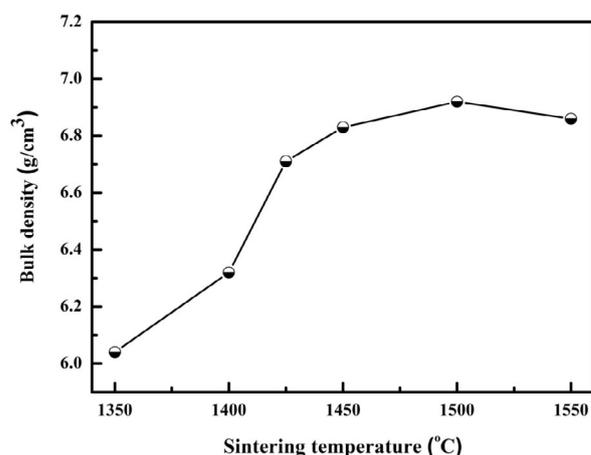


Fig. 7 Bulk densities of SmAlO_3 ceramics sintered at different temperatures.

bulk density of $6.92\text{ g}\cdot\text{cm}^{-3}$, which is about 96.5% of the theoretical density, was obtained at $1500\text{ }^\circ\text{C}$ for 4 h. Compared with the densification temperature of $1650\text{ }^\circ\text{C}$ required by the conventional solid-state reaction route, the best sintering temperature of SmAlO_3 ceramics prepared using the powders synthesized by the stearic acid method as raw materials can be reduced about $150\text{ }^\circ\text{C}$. In fact, the theoretical density values all exceed 95%, at which the influence of pores can be ignored for the microwave dielectric properties. A recent study suggested that a reasonable calcination temperature is critical for obtaining dense ceramics with favorable microwave dielectric properties [26]. Low or high calcination temperature is disadvantageous to achieving high-density ceramics in the sintering process. A systematic investigation is worthy to explore for greater optimization of density and microwave dielectric properties.

The SEM image of SmAlO_3 ceramics at $1500\text{ }^\circ\text{C}$ for 4 h is shown in Fig. 8. A dense, uniform microstructure with an average grain size of about $1.8\text{ }\mu\text{m}$ can be observed, which is advantageous to enhancing microwave dielectric properties. In addition, a layered growth step morphology can be found based on the SEM image. Similar results have been reported in the ceramic systems of $0.7\text{CaTiO}_3\text{--}0.3\text{SmAlO}_3$ and $0.95(\text{Ca}_{0.88}\text{Sr}_{0.12})\text{TiO}_3\text{--}0.05(\text{Bi}_{0.5}\text{Na}_{0.5})\text{TiO}_3$ [26,27]. These phenomena have a close relationship with the grain growth and mass transfer in the sintering process.

3.5 Microwave dielectric properties of as-prepared SmAlO_3 ceramics

The microwave dielectric properties of SmAlO_3 ceramics

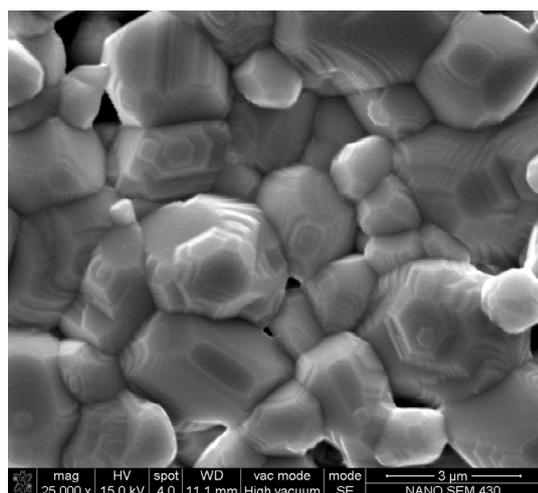


Fig. 8 SEM image of the sintered ceramics at 1500 °C for 4 h.

are demonstrated in Fig. 9. As can be seen, the relationship between the ϵ_r value and sintering temperature is in firm agreement with that between the density and sintering temperature. The ϵ_r firstly increases to the saturated value at 1500 °C and then decreases slightly in the investigated sintering temperature. Also, the $Q \cdot f$ value exhibits a similar behavior. The dielectric constant and $Q \cdot f$ are strongly affected by the pores in the structure before the ceramics are well sintered. Therefore, the ϵ_r and $Q \cdot f$ values are obviously reduced when the sintering temperature is below 1525 °C. However, when the sintering temperature is in the range of 1450–1550 °C, the variations in the ϵ_r and $Q \cdot f$ values are relatively smaller, which are attributed to high density and uniform microstructure with reduced porosity. The maximum values of 20.22 and 74110 GHz for the SmAlO_3 ceramics at 1500 °C are obtained, respectively. Compared to the conventional solid-state reaction method, the $Q \cdot f$ value can be improved by the stearic acid method. As is well known, the $Q \cdot f$ of ceramics is closely related to the two factors of intrinsic defects and extrinsic defects. Intrinsic defects are related with lattice vibrational modes, while extrinsic defects depend on density and secondary phase, as well as grain size. The fine microstructure with homogeneous grain morphology and grain size, obtained using fine nanopowders as starting materials, should be responsible for the increase of the $Q \cdot f$ value for the present ceramics, though the microstructure of the ceramics was not provided in the reference [5].

Figure 9(b) shows the τ_f of SmAlO_3 ceramics obtained

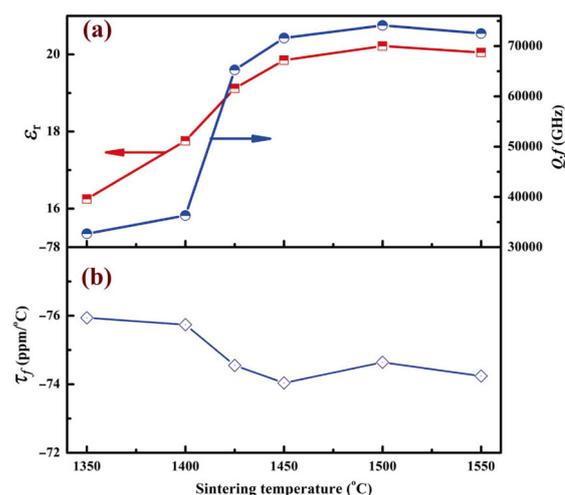


Fig. 9 Variation in microwave dielectric properties containing ϵ_r , $Q \cdot f$, and τ_f .

from the stearic acid method. At 1350–1550 °C, the τ_f varies between -74.04 and -75.94 ppm/°C. It is generally known that the τ_f is closely related to the ϵ_r of a given composition, which depends on the distortion of the oxygen octahedral in the crystal structure [28]. Therefore, no significant rule can be found for the relationship between τ_f and sintering temperature.

The sintering properties and microwave dielectric characteristics of SmAlO_3 ceramics, derived from various methods, are summarized in Table 2. Compared to the conventional solid-state reaction method, the stearic acid approach significantly reduces the calcination temperature and sintering temperature of SmAlO_3 ceramics. Moreover, the microwave dielectric properties, especially the $Q \cdot f$ value, are greatly improved. As a result, the stearic acid method is one of the most facile, energy-saving, and promising approaches to preparing SmAlO_3 ceramics with favorable microwave dielectric properties. However, the ϵ_r and $Q \cdot f$ values obtained by the stearic acid method are slightly lower than those achieved by polymeric precursor method, as shown in Table 2. It is essential to study the process optimization of the current synthesis method such as the adjustment of solution concentration, pH, and calcination temperature, for further improvement of the microwave dielectric properties.

4 Conclusions

In the current study, SmAlO_3 powders were synthesized successfully by the stearic acid method. The characterization

Table 2 Sintering parameters and microwave dielectric characteristics of SmAlO₃ ceramics

Method	CT (°C)	ST (°C)	RD (%)	Average particle size (nm)*	ε _r	Q·f (GHz)	τ _r (ppm/°C)	Ref.
SS	1400	1650	—	—	20.4	65000	−74	[5]
PP	900	1550	97.2	26	20.94	78600	−71.8	[19]
CP	900	1550	97	45	20.54	75380	−69.2	[18]
SA	750	1500	96.5	29	20.22	74110	−74.6	This work

CP: citrate precursor; CT: calcination temperature; ST: sintering temperature; RD: relative density.

*Calculated from the XRD patterns.

results strongly demonstrated that the stearic acid method is one of the most facile, low-cost, and energy-saving approaches to synthesize well-crystallized SmAlO₃ phase. The average crystalline size of SmAlO₃ powders synthesized at 750 °C was about 70 nm and in a mostly irregular shape based on the TEM results. The stearic acid approach significantly reduced the calcination temperature and sintering temperature of SmAlO₃ compared with the conventional solid-state reaction method. Also, the microwave dielectric properties, especially the Q·f value, were greatly improved. However, the ε_r and Q·f values obtained in our present work were slightly lower than those achieved by the polymeric precursor method. A systematic investigation needs to be done for better optimization of density and microwave dielectric properties of SmAlO₃ ceramics derived from the stearic acid method in the future work.

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