

High-temperature heat treatment tailored crystals and microstructures of multicomponent rare-earth tantalates RETaO₄ nano powders

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ABSTRACT: Tailoring crystals and microstructures of multicomponent rare-earth tantalate RETaO₄ nano powders can promote their application as thermal protective coating materials because their properties are dominated by the structures. In this work, multicomponent RETaO₄ nano powders are synthesized via the chemical coprecipitation method, and their structures are tailored by changing the annealing temperature. After annealed at 800–1500 °C, the multicomponent RETaO₄ nano powders can be crystallized into metastable tetragonal (t'), monoclinic-prime (m'), and monoclinic (m) phases, and their particle sizes (8–652 nm) gradually increase with increasing temperature. The optimal annealing temperature of RETaO₄ powders is determined to be 1000 °C based on the crystallinity degree and particle sizes, which are 11.3 nm and 89.5%, respectively. High-resolution transmission electron microscopy (HR-TEM) and corresponding energy dispersive X-ray spectroscopy (EDS) mapping have validated the compositional uniformity of each element at the nanoscale, and the interplanar spacing of different phases corresponds to the X-ray diffraction (XRD) Rietveld refinements. This work demonstrates that high-temperature heat treatment can act as an effective mean to tailor the particle sizes and crystal structures of RETaO₄ ceramic, which can be further applied to synthesize nano spherical powders and coatings in future studies.

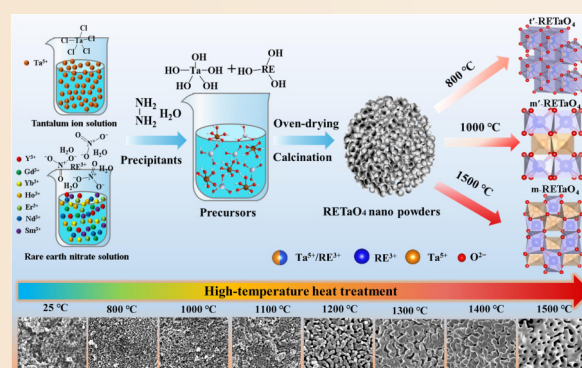
KEYWORDS: rare-earth tantalates; nanopowders; heat treatment; structural regulations; thermal barrier coatings (TBCs)

1 Introduction

Yttria-stabilized zirconia (YSZ), as a traditional thermal barrier coating (TBC), markedly enhances the service temperature of aerospace hot sections [1–3]. Nevertheless, owing to significant volume variations during the phase transition at 1200 °C and comparatively high thermal conductivity, its application is severely limited in high-temperature structural components [4–6]. To overcome the above challenges, novel thermal barrier coating materials, such as rare-earth zirconates [7–13], tantalates [14–20], silicates [21–24], phosphates [25–29], and so on [30–37], have garnered significant attention and are widely studied. The above materials possess distinct merits, and they fail to completely substitute YSZ as next-generation TBCs. Researchers have been able to enhance the shortcomings of the above materials by alloying [18,38–41], nanosizing [42–49], doping [32,49–52], and so on [11,19,20,31,53–55]. Among the various methods, nanoization is an effective method that does not change the

intrinsic characteristics. Zheng *et al.* [44] reported that nano-structured Yb₂SiO₅ coatings display a fracture toughness of 1.21 MPa·m^{1/2}, representing a 70% improvement over the conventional counterpart (0.71 MPa·m^{1/2}). The nanograins within the nano-structured Yb₂SiO₅ coatings reinforce the coating interface, thereby facilitating crack deflection. The aforementioned deflection mechanism not only impedes crack extension and extends the crack propagation route but also ultimately enhances the fracture toughness. Xue *et al.* [48] demonstrated that the micro-nanostructured ceramic coatings deform under tension or pressure, but they automatically return after the force disappears. The micropores and cracks in the nanozone can dissipate stress and reduce stress damage. Thus, nano-structure coatings can alleviate thermal stress accumulation during thermal cycling and suppress crack initiation and propagation, which consequently enhances thermal cycling durability.

The described studies focus on the fracture toughness and thermal properties of nano-structured coatings, and nanoization



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induces a remarkable enhancement in the properties. The preparation process of nano powders plays a key role in nanostructured materials and their coatings. The structure, microstructure, and crystallinity of nanopowders directly influence the thermal-physical properties of materials [42–49]. How to better control these powder characteristics is the primary focus of this research. Ferroelastic tantalates RETaO_4 have garnered substantial research interest owing to their outstanding fracture toughness [18,19,56], low thermal conductivities, and high coefficients of thermal expansion [57–59]. It is expected that RETaO_4 is the next-generation TBC material; however, the optimized heat treatments of multicomponent RETaO_4 nanopowders and the influences on the crystal structure and microstructure are not yet clear and will substantially affect their properties.

For RETaO_4 ceramic, the properties are significantly affected by the crystal structures and microstructures [17,57,58]. RETaO_4 ceramic synthesized via the solid-state reaction is crystallized into monocline (m) and monocline-prime (m') phases [19,58], and it is difficult to obtain the tetragonal (t) phase due to the t–m transition during cooling [59–62]. By changing the heat-treatment temperatures during the chemical coprecipitation method, it is possible to tailor the crystal structures and microstructures of RETaO_4 ceramic, which potentially enables the regulation of mechanical and thermal properties in future studies. Additionally, nanopowders of RETaO_4 can be obtained by controlling the annealing temperatures after chemical coprecipitation synthesis, which can be further used to synthesize nanospheres of RETaO_4 , and their crystals and microstructures can be tailored by changing the heat treatment temperature. This work proposes the relationship among heat-treatment temperatures, crystals, and microstructures of RETaO_4 nanopowders. Simultaneously, the method of solid-solving rare-earth atoms with varying ionic radii and masses at site A serves as an effective means to regulate material properties. To investigate their synthesis process, we designed and synthesized six multicomponent RETaO_4 nanopowders, i.e., $\text{Y}_{1/2}\text{Gd}_{1/2}\text{TaO}_4$ (S2), $\text{Y}_{1/3}\text{Gd}_{1/3}\text{Yb}_{1/3}\text{TaO}_4$ (S3), $\text{Y}_{1/4}\text{Gd}_{1/4}\text{Yb}_{1/4}\text{Ho}_{1/4}\text{TaO}_4$ (S4), $\text{Y}_{1/5}\text{Gd}_{1/5}\text{Yb}_{1/5}\text{Ho}_{1/5}\text{Er}_{1/5}\text{TaO}_4$ (S5), $\text{Y}_{1/6}\text{Gd}_{1/6}\text{Yb}_{1/6}\text{Ho}_{1/6}\text{Er}_{1/6}\text{Nd}_{1/6}\text{TaO}_4$ (S6), and $\text{Y}_{1/7}\text{Gd}_{1/7}\text{Yb}_{1/7}\text{Ho}_{1/7}\text{Er}_{1/7}\text{Nd}_{1/7}\text{Sm}_{1/7}\text{TaO}_4$ (S7), and investigated their crystal structures and microstructural evolution with different compositions.

Finally, we summarize the preparation processes and heat-treatment parameters of RETaO_4 nano powders, and it is successful to tailor their crystals and microstructures by changing the heat-treatment temperatures. The differences in ionic radius and mass influence the synthesis pathways of nanopowders. To better illustrate the reaction pathways, we selected a component (S7) exhibiting significant variations in ionic radius and mass to be the typical studied ceramic in this work, and other samples were investigated based on its results.

2 Experimental

2.1 Specimen synthesis

Six different multicomponent RETaO_4 nano powders were synthesized via the chemical co-precipitation method. Rare-earth nitrates ($\text{RE}(\text{NO}_3)_3 \cdot x\text{H}_2\text{O}$ (RE = Y, Gd, Yb, Ho, Er, Nd, and Sm), purity $\geq 99.9\%$, Macklin, China) and TaCl_5 (purity $\geq 99.9\%$, Macklin, China) were used as preceramic polymers for S2 to S7. Hydrazine hydrate ($\text{N}_2\text{H}_4 \cdot \text{H}_2\text{O}$, purity $\geq 99.9\%$, Macklin, China) was added as the alkaline precipitant. Figure 1 outlines the detailed synthesis process of RETaO_4 nano powders. Initially, $\text{RE}(\text{NO}_3)_3 \cdot x\text{H}_2\text{O}$ and TaCl_5 were dissolved into $\text{C}_2\text{H}_5\text{OH}$ (purity $\geq 99.99\%$, Keyi, China) with a $\text{RE}^{3+}/\text{Ta}^{5+}$ molar ratio of 1 : 1. Subsequently, 5 wt.% of $\text{N}_2\text{H}_4 \cdot \text{H}_2\text{O}$ was added to regulate the pH. When the pH value exceeded 9, the precursor coagulated to form a precipitate that settled out. Eventually, the precursors were dried at 60–70 °C for 12 h and calcinated at 800, 1000, 1500, and 1600 °C for 3 h. The crystals and microstructures were tailored by changing the calcination temperatures, as discussed in detail in Section 3.

2.2 Thermal stability measurements

A simultaneous thermal analyzer (STA; 449F3, NETZSCH, Germany) was used to characterize the phase transitions and microstructural evolutions at different temperatures. Thermogravimetry (TG), differential scanning calorimetry (DSC), and differential thermogravimetry (DTG) analyses were performed. Specifically, the TG and DSC curves were recorded from room temperature (RT = 25 °C) to 1500 °C at a heating rate of 5 °C/min.

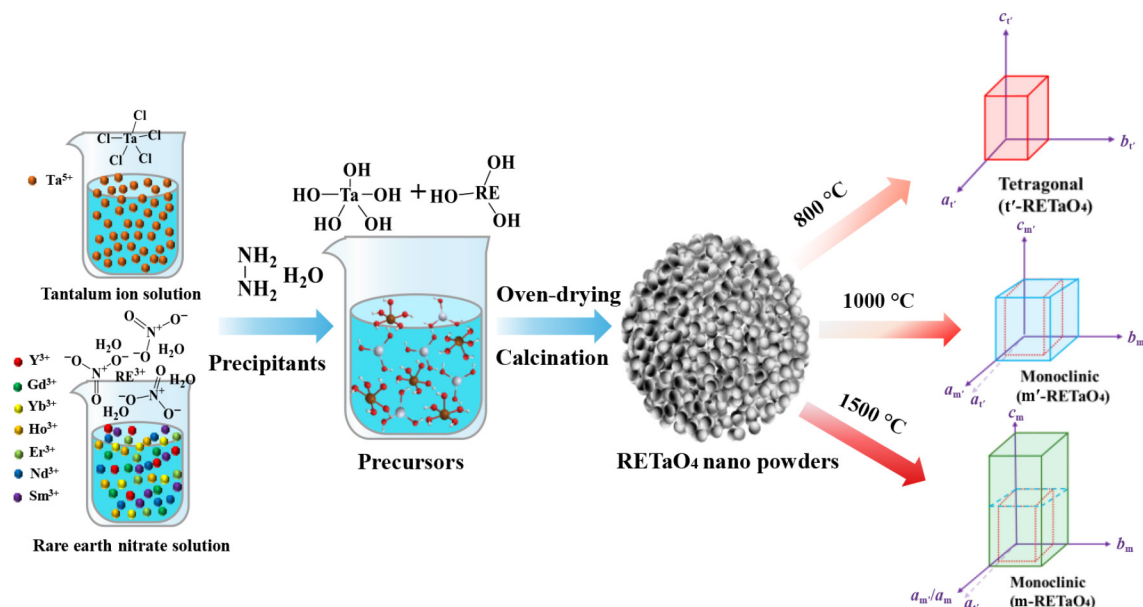


Fig. 1 Synthesis process of multicomponent RETaO_4 nano powders.

2.3 Structural characterizations

The crystal structures were determined by an X-ray diffractometer (XRD; MiniFlex 600, Rigaku, Japan). The XRD Rietveld refinements were performed by GSAS II. The wavelength of Cu K α radiation ($\lambda = 0.15406$ nm), constant parameter equal ($k = 0.94$), Bragg angle (θ), and peak full-width at half-maximum intensity (β_D) were used to calculate the particle sizes (D) of the as-synthesized nanopowders. The detailed procedures of the Scherrer equation were elaborated as Eq. (1) [63]:

$$D = \frac{k\lambda}{B_D \cos \theta} \quad (1)$$

To contrast the crystallinity degree of the as-synthesized ceramic powders at different annealing temperatures. The crystallinity degree (X_c) were elaborated by the area of the crystalline peaks (I_c), and the area of the amorphous peaks (I_a) as shown in Eq. (2) [63]:

$$X_c = \frac{I_c}{I_c + I_a} \times 100\% \quad (2)$$

2.4 Microstructural characterizations

The microstructures were investigated by a scanning electron microscope (SEM; EVO 300, Zeiss, Germany) and a high-resolution transmission electron microscope (HR-TEM; Tecnai F20G2, FEI, USA) equipped with an energy dispersive X-ray spectroscopy (EDS). In addition, the particle size distribution was obtained via SEM images. The general calculation formula for Span was defined as Eq. (3) [64]:

$$\text{Span} = \frac{D_{90} - D_{10}}{D_{50}} \quad (3)$$

where D_{10} is the 10th percentile particle size, D_{50} is the median particle size, and D_{90} is the 90th percentile particle size.

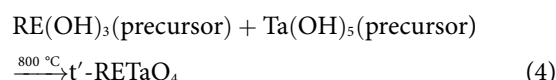
3 Results and discussion

3.1 Formation of RETaO₄ phases

Figure 2 depicts the reaction process of S7 precursors from RT to 1500 °C based on TG-DSC-DTG analysis. To mitigate the influence of bound water, the sample is first annealed at 600 °C for 5 h before measurement. There are three stages during the thermal decomposition process, named A, B, and C. At temperatures ranging from RT to 750 °C, no distinct endothermic or exothermic peaks are observed in the DSC curve, which

indicates that the sample does not undergo a phase transition. It is defined as the first stage (A). In the second stage (B), the TG curve exhibits a steep decline, which signifies that the phase transition has occurred. Other studies have reported that within temperatures ranging from 750 to 1400 °C, the RETaO₄ ceramic undergoes a phase transition from the t' to m' phase. Above 1400 °C, the RETaO₄ ceramic has a phase transition from the m' phase to the t phase, which is hence defined as the third stage (C).

At stage A below 750 °C, a small amount of weight loss is found, and the DSC curve fails to exhibit a sharp endothermic drop, which indicates the evaporation of bound water and the thermal decomposition of residual solvents [15,63,65]. During stage B at 750–1400 °C, an obvious sharp exothermic peak is detected at 807.7 °C in Fig. 2(a), while the weight loss of the precursor is 6.08%. Figure 2(b) shows a pronounced and sharp exothermic peak at 813.5 °C. The following XRD characterizations of the samples annealed at 700–1000 °C reveal two distinct stages of phase transitions, and the reaction equations are expressed by Eqs. (4) and (5):



In stage A, the precursors of RE(OH)₃ and Ta(OH)₅ react and crystallize with increasing temperature. RETaO₄ exhibits four distinct phases: the low-temperature tetragonal phase (t'), monoclinic-prime phase (m'), high-temperature tetragonal phase (t), and monoclinic phase (m) at room temperature after cooling. The m' and m phases belong to the monoclinic crystal system, whereas the t' and t phases belong to the tetragonal crystal system. The unit cell volume of the t' phase is only half of the m' phase and a mere one-quarter of the m phase. The high-temperature t phase is unstable and difficult to retain at room temperature. The m phase forms via the reversible high-temperature ferroelastic transition (t $\leftrightarrow$ m). The t' phase and m' phase are analogous to intermediate phases. After high-temperature heat treatment, the t' phase and m' phase disappear, and the phase transformation is irreversible. To distinguish them from the high-temperature t phase and the room temperature m phase, they are named the t' phase and m' phase. The t phase with an I41/a space group was determined in previous works [60–62]. The t'-RETaO₄ with a P42/nmc space group and m' phase with a P21/a space group are determined based on the XRD patterns in Fig. 3(a). As shown in Fig. 2(b), no additional exothermic or endothermic peaks are

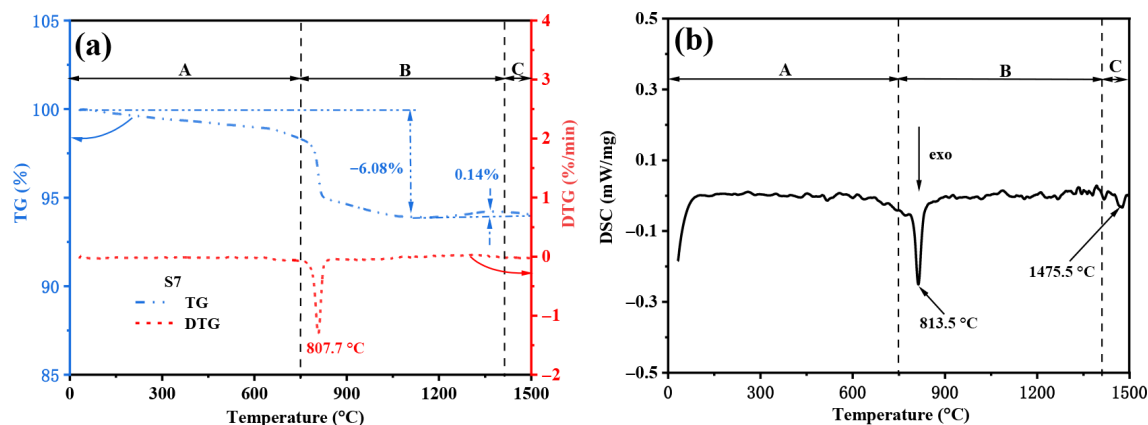
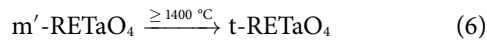


Fig. 2 TG-DSC-DTG curves of S7 precursor at RT–1500 °C: (a) TG and DTG curves and (b) DSC curve.

observed except at 813.5 and 1475.5 °C. These observations indicate that the inorganic salts have been fully decomposed, with the precipitation of multicomponent RETaO_4 nanopowders (S2–S7). Mather and Davies [60] have also reported that t' - RETaO_4 is retained at 25–850 °C, and a single-phase m' - RETaO_4 is stable at 1000–1425 °C. In agreement with our result in Fig. 7, S7 annealed at 1000–1400 °C is crystallized into an m' structure.

During stage C above 1400 °C, an additional exothermic peak is observed at 1475.5 °C, but no obvious other exothermic or endothermic peaks are observed in the TG or DTG curves. The crystal structure of S7 has converted from the m' to the stable high-temperature t phase. Stubičan [62] reported that an endothermic peak detected at approximately 1500 °C correlates with the m – t transition, which in turn converts to the m -fergusonite phase upon cooling, and it also serves as the key factor driving the formation of the m phase. At the same time, Qu *et al.* [59] also have proven that $(\text{Gd}_{1-x}\text{Y}_x)\text{TaO}_4$ will be transformed from the m to t phase at 1400–1500 °C. Furthermore, Feng *et al.* [61] demonstrated that the m – t phase transition temperature of YTao_4 is approximately 1430 °C, which is influenced by the RE^{3+} ion radius. Subsequently, the m' – t and t – m phase transitions of RETaO_4 nanopowders take place via Eqs. (6) and (7), respectively:



3.2 Crystal structures

To obtain a single m phase, we select a higher annealing temperature of 1600 °C for these samples. Based on Fig. 2, S2–S7

were annealed at 800, 1000, and 1600 °C to obtain RETaO_4 powders with tailored crystal structures. The XRD patterns are shown in Figs. 3(a)–3(c). Single-phase RETaO_4 powders are successfully synthesized after annealing at different temperatures. Three crystal structures, including the t' , m' , and m phases, of RETaO_4 are obtained, which indicates that the crystal structures can be tailored by changing the annealing temperature. After annealing at 800 °C for 3 h, S2–S7 are crystallized into the t' phase with a $P42/nmc$ space group, which corresponds to PDF#50-0846, as shown in Fig. 3(a). Figure 3(b) shows the XRD patterns of the m' - RETaO_4 nanopowders. After the annealing temperature increases from 800 to 1000 °C, the t' phase is transformed to a m' phase with a $P21/a$ space group. To obtain powders with an m phase, the annealing temperature of S2–S7 is increased to 1600 °C. Figure 3(c) shows the XRD patterns of S2–S7 annealed at 1600 °C for 3 h, and the m' - RETaO_4 is transformed to an m phase with an $I2/a$ space group. According to XRD, Fig. 3(d) shows the lattice parameters of RETaO_4 , and their unit cell volume exhibits a pronounced trend toward expansion with increasing annealing temperature.

Figures 4–6 display the XRD refinement results of S2–S7 after annealing at 800, 1000, and 1600 °C, respectively, and the lattice parameters are listed in Tables 1–3. The XRD Rietveld refinement results are reliable, as the goodness-of-fit indicators (χ^2), and R -weighted pattern (R_{wp}) match well with standard reference values. The crystal structure plays a key role in thermal/mechanical properties. m - RETaO_4 exhibits higher thermal conductivity and Young's modulus and lower thermal expansion coefficients (TECs) [5,56]. In contrast, m' - RETaO_4 shows lower thermal conductivity and Young's modulus, as well as relatively high TECs

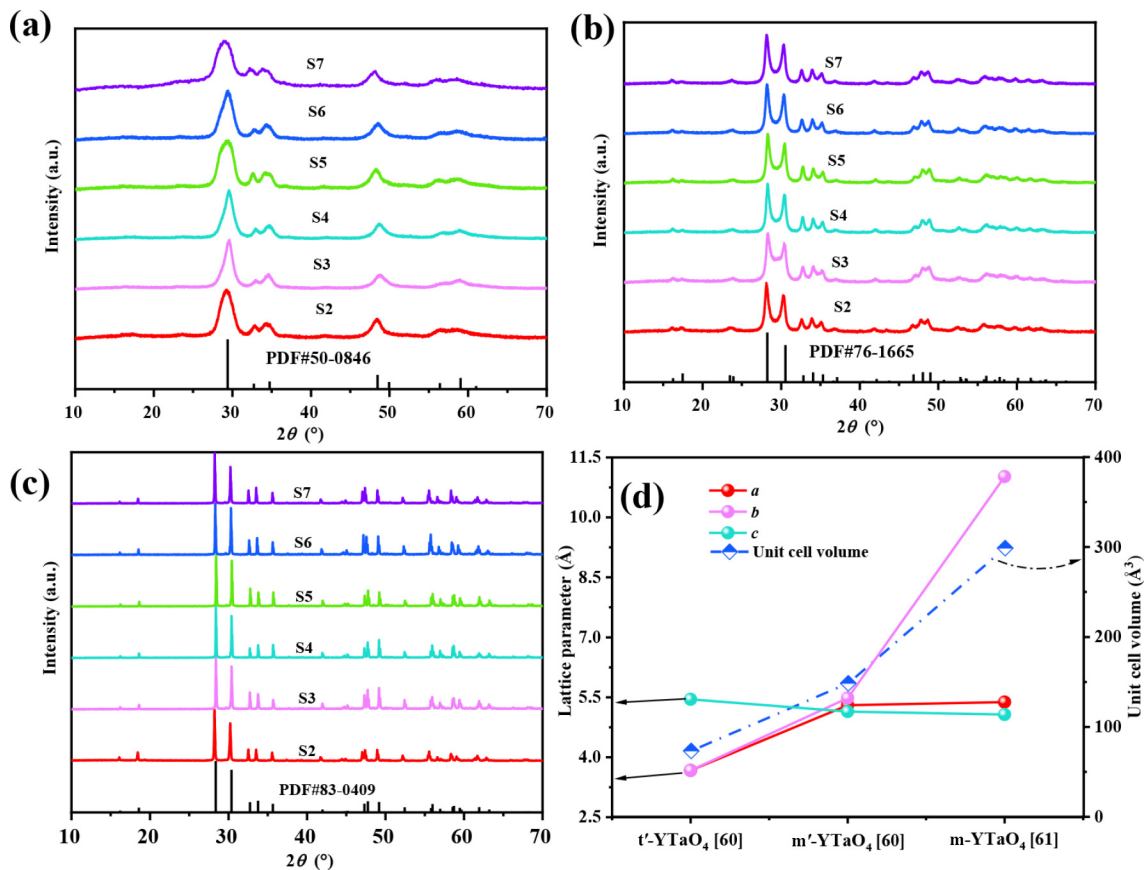


Fig. 3 Crystal structure identification and lattice parameters of S2–S7 after annealing at 800, 1000, and 1600 °C for 3 h: (a) 800 °C, (b) 1000 °C, and (c) 1600 °C. (d) Lattice parameters of YTao_4 with different structures [60, 61].

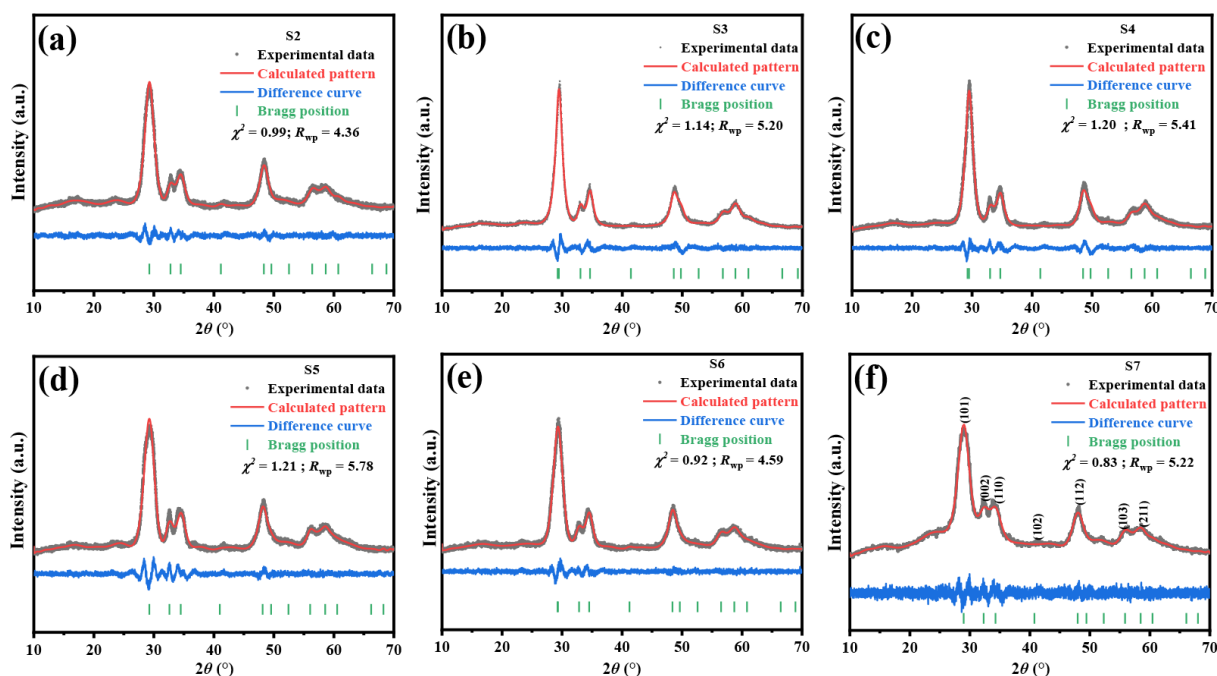


Fig. 4 XRD refinement results of S2–S7 after annealing at 800 °C: (a) S2, (b) S3, (c) S4, (d) S5, (e) S6, and (f) S7.

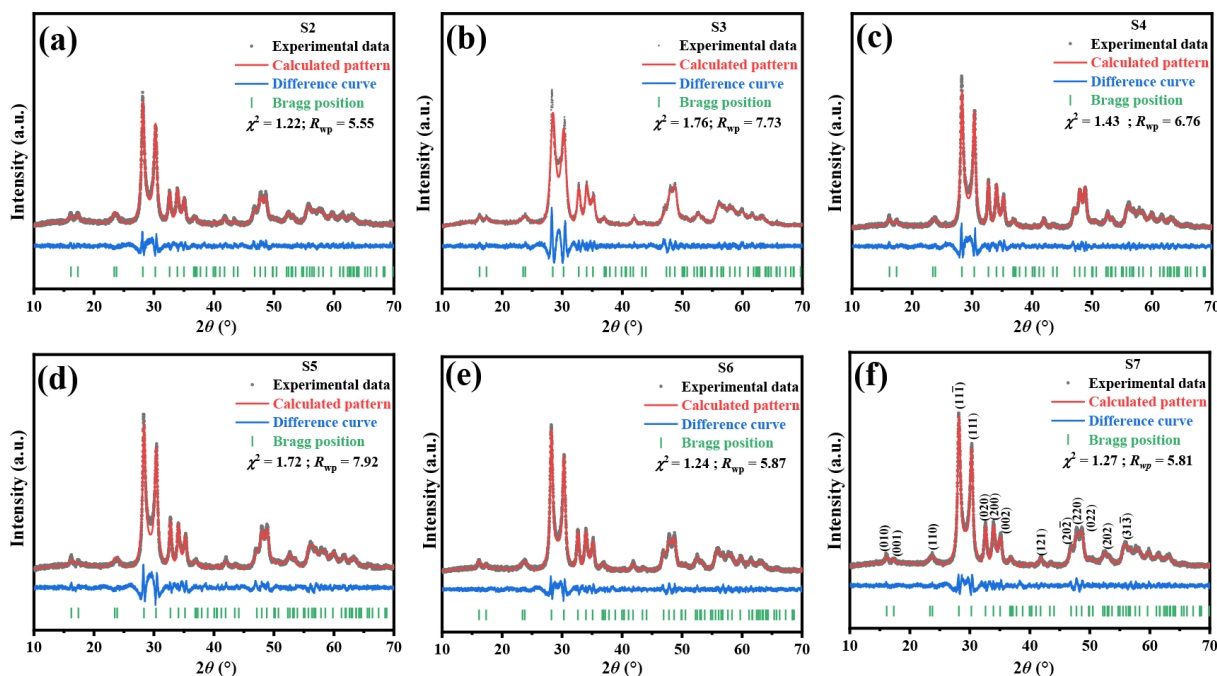


Fig. 5 XRD refinement results of S2–S7 after annealing at 1000 °C: (a) S2, (b) S3, (c) S4, (d) S5, (e) S6, and (f) S7.

[5,56–59]. Although research on t' - RETaO_4 is relatively limited, the significant differences in its crystal structure imply that its properties will be different. Therefore, the crystal structures of RETaO_4 are tailored by adjusting the heat treatment temperature. Furthermore, the lattice parameters of S2 are in excellent agreement with previous research [59], which further confirms the reliability of this work.

3.3 Microstructures and grain sizes

Based on the TG-DSC-DTG analysis, the precipitates of S7 were annealed at 800–1500 °C for 3 h to obtain nano powders with different crystal structures, as shown in Fig. 7. The as-synthesized S7 exhibits the t' -phase after annealed at 800 °C for 3 h. The

derivation of XRD is consistent with the results of TG-DSC-DTG curves and Eqs. (1) and (2) [15,61]. As the annealing temperature increases from 1000 to 1400 °C, the t' phase transitions into an m' phase [60].

The grain size distributions of S7 annealed at different temperatures are determined, and the microstructures are shown in Figs. 8(a)–8(i). After annealed at 800 °C for 3 h, Fig. 8(b) shows that numerous nanoparticles are formed, and the amorphous phase is dissociated, as shown Fig. 8(c). According to the results of the TG-DSC-DTG curves, the t' – m' phase transform temperature of S7 is 800–900 °C. To obtain a single-phase structure, the samples are annealed at a higher temperature of 1000 °C, and the SEM images are shown in Fig. 8(d). From Figs. 8(c)–8(h), the

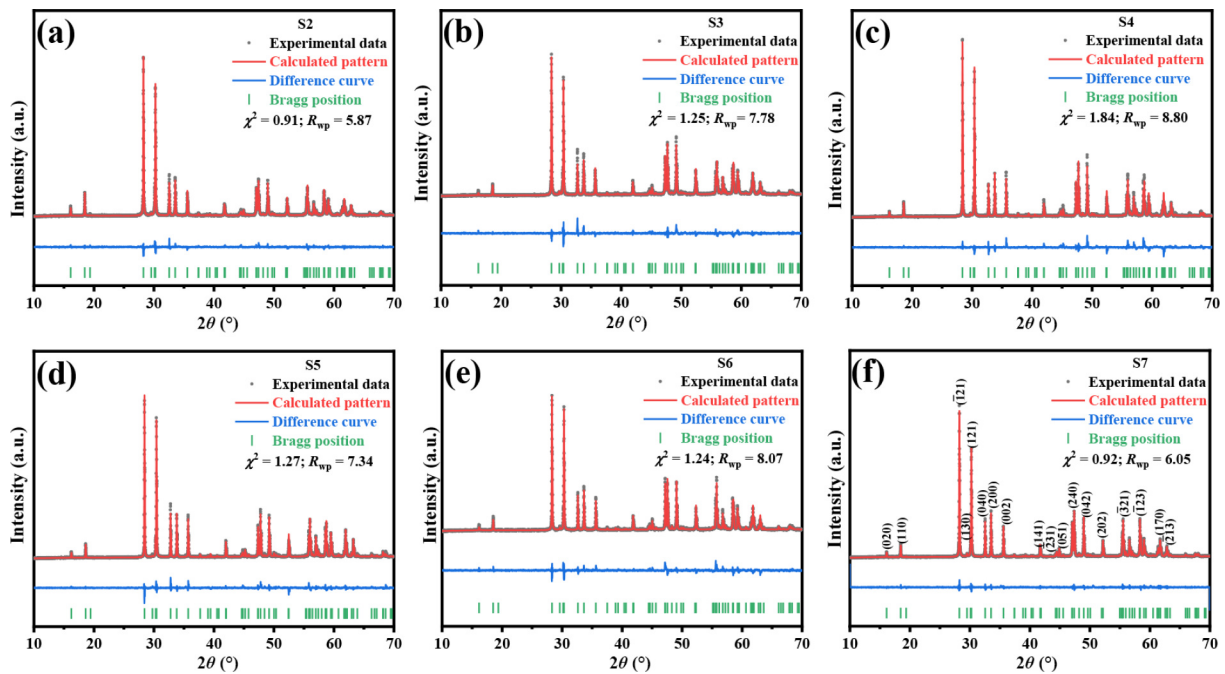


Fig. 6 XRD refinement results of S2–S7 after annealing at 1600 °C: (a) S2, (b) S3, (c) S4, (d) S5, (e) S6, and (f) S7.

Table 1 Lattice parameters (a , b , c), unit cell volume (V), theoretical density (ρ_0), and effective atomic ionic radius (R) of S2–S7 after annealing at 800 °C for 3 h

Sample	R (nm)	a (Å)	b (Å)	c (Å)	V (Å ³)	ρ_0 (g/cm ³)
S2	0.0919	3.677	3.677	5.491	74.240	8.234
S3	0.0900	3.667	3.667	5.428	72.986	8.754
S4	0.0901	3.646	3.646	5.459	72.573	8.948
S5	0.0898	3.644	3.644	5.460	72.486	9.056
S6	0.0908	3.668	3.668	5.444	72.706	9.006
S7	0.0919	3.667	3.667	5.454	73.326	8.933

Table 2 Lattice parameters (a , b , c), unit cell volume (V), and theoretical density (ρ_0) of S2–S7 after annealing at 1000 °C for 3 h

Sample	a (Å)	b (Å)	c (Å)	V (Å ³)	ρ_0 (g/cm ³)
S2	5.316	5.492	5.153	149.659	8.169
S3	5.288	5.473	5.133	148.005	8.634
S4	5.286	5.470	5.124	147.429	8.809
S5	5.285	5.469	5.124	147.418	8.906
S6	5.303	5.484	5.137	148.626	8.834
S7	5.306	5.487	5.141	148.923	8.797

Table 3 Lattice parameters (a , b , and c), unit cell volume (V), and theoretical density (ρ_0) of S2–S7 after annealing at 1600 °C for 3 h

Sample	a (Å)	b (Å)	c (Å)	V (Å ³)	ρ_0 (g/cm ³)
S2	5.367	11.002	5.070	297.935	8.207
S3	5.336	10.952	5.058	294.219	8.686
S4	5.333	10.946	5.057	293.848	8.840
S5	5.330	10.939	5.056	293.401	8.950
S6	5.305	10.973	5.062	295.805	8.877
S7	5.367	11.000	5.064	297.592	8.804

particle sizes of S7 gradually increase, and no ferroelastic domains are found in these samples. After annealed at 1500 °C for 3 h, a significant sintering is observed. The ferroelastic domains are extensively distributed inside the grains of S7, as shown in Fig. 8(i) [4,15,18,57–59].

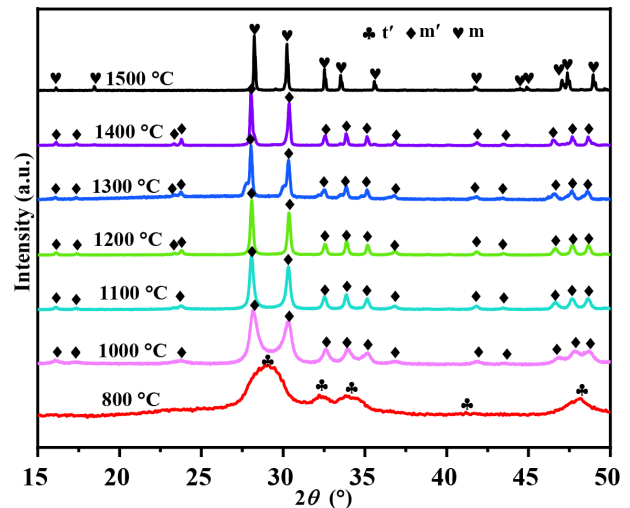


Fig. 7 XRD patterns of S7 annealed at 800–1500 °C.

In Figs. 9(a)–9(c), the linear intercept method is employed to statistically analyze the grain size distribution of S7 after annealed at 1200–1400 °C. After annealed at 1500 °C, significant sintering is triggered in S7, which is unsuitable for statistical analysis. Below 1200 °C, the excessively small grain sizes and powder agglomeration prevent statistical analysis from yielding accurate values. Above 1200 °C, the powders exhibit sintering behavior, and they gradually transform from powder particles into a structure with continuous grain boundaries. Figures 9(a)–9(c)

show the grain sizes of S7 after annealed at 1200–1400 °C. Based on the measured particle size parameters, the results of the

distribution width coefficient are listed in Table 4. The Span of the powders is determined to be approximately 0.79–0.86, indicating a

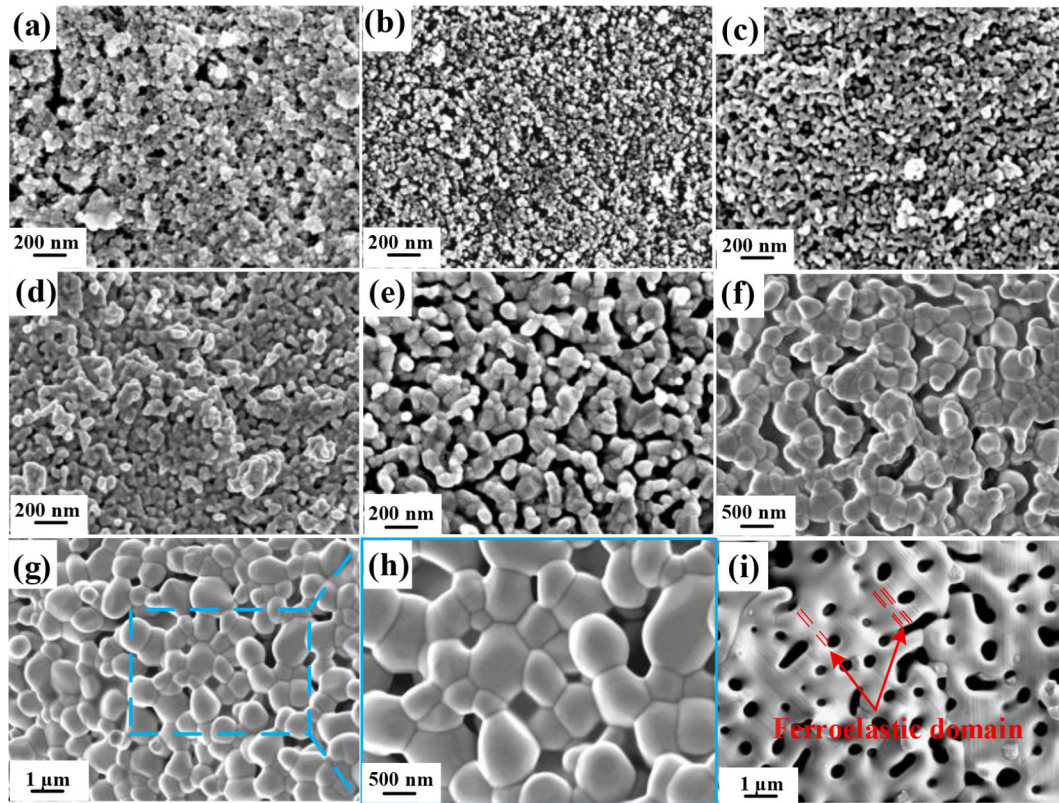


Fig. 8 Microstructures of S7 precipitates after annealing at different temperatures: (a) RT, (b) 800 °C, (c) 1000 °C, (d) 1100 °C, (e) 1200 °C, (f) 1300 °C, (g, h) 1400 °C, and (i) 1500 °C.

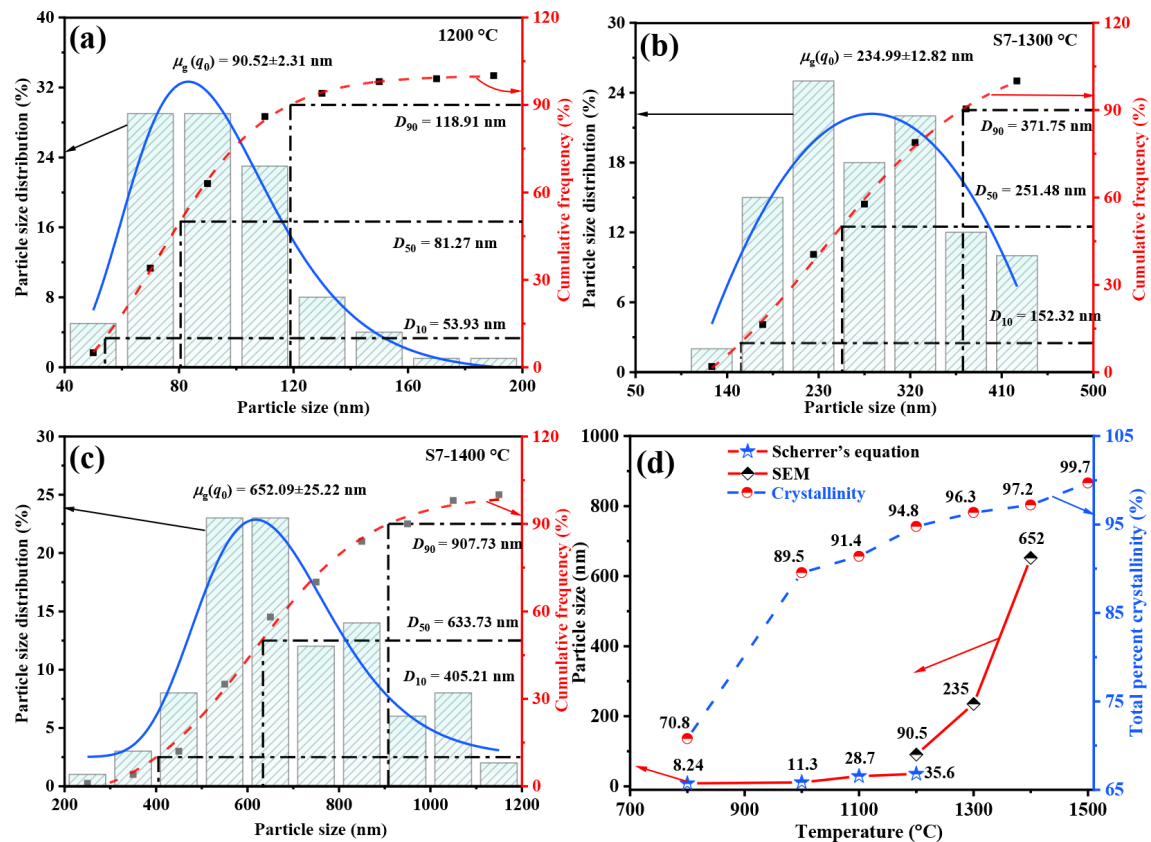


Fig. 9 Grain sizes of S7 after annealing at different temperatures: (a) 1200 °C, (b) 1300 °C, (c) 1400 °C, and (d) particle sizes of S7 calculated by Scherrer's equation and SEM analysis and the total percent crystallinity.

narrow particle size distribution [64]. Specifically, after annealed at 1200 °C, D_{50} is measured to be 81.27 nm, which is below the 100 nm threshold. The aforementioned data provide empirical support for the later-stage synthesis of nanomaterials. As shown in Fig. 9(d), the particle sizes of the S7 powders calculated by Scherrer's equation are 8.24–35.6 nm after annealed at 800–1200 °C. The particle size data are roughly in line with the particle size range

Table 4 Particle sizes of S7 after annealed at 1200–1400 °C

Temperature (°C)	D_{10} (nm)	D_{50} (nm)	D_{90} (nm)	Span
1200	53.93	81.27	118.91	0.80
1300	153.48	253.05	371.75	0.86
1400	405.21	633.73	907.73	0.79

Note: D_{10} , D_{50} , and D_{90} are the 10th, 50th, and 90th percentiles of the cumulative grain size, respectively, and Span is the distribution width coefficient.

documented in Mather's paper [60]. Agglomeration of particles and lattice strain are the primary causes for the significant discrepancies between the results of electron microscopy analysis and Scherrer calculations [66–68]. A further calculation of crystallinity is performed to more effectively identify the optimal annealing process. Figure 9(d) shows the total percent crystallinity after annealed at 800–1500 °C. Upon annealing from 800 to 1000 °C, the crystallinity exhibits a significant surge, whereas the particle size shows a negligible variation. The crystallinity degree reaches 89.5% after annealed at 1000 °C, which meets the crystallinity requirement for spray powders [69,70]. Coupled with the nanoscale particle size, 1000 °C is the ideal annealing temperature.

Figures 10 and 11 show the TEM results of S7 after annealed at 800, 1000, and 1500 °C. The TEM-EDS mapping shows that each rare-earth element is homogeneously distributed in ceramic powders, as shown in Figs. 10(a)–10(c). Meanwhile, compared

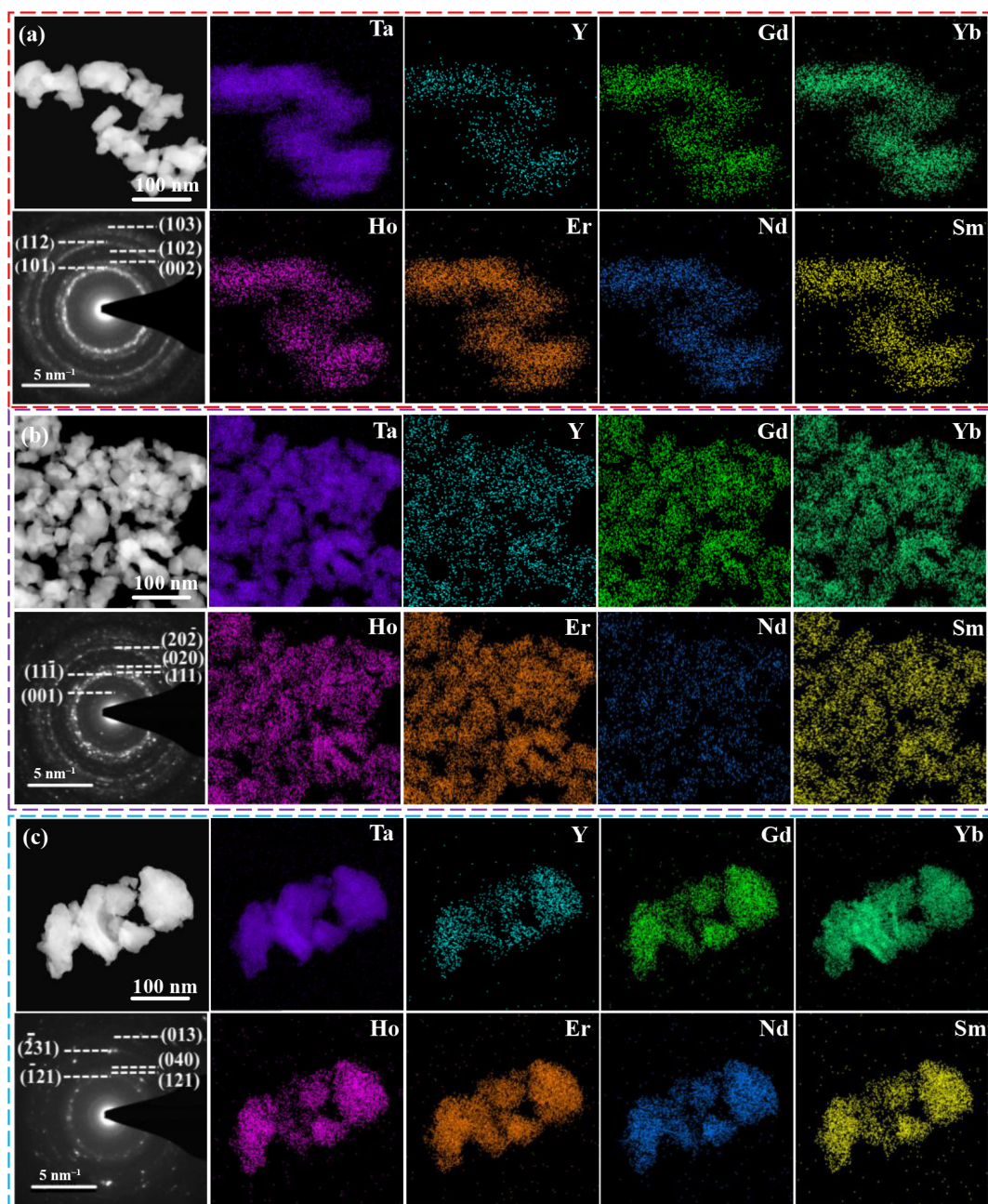


Fig. 10 Typical nanostructures, corresponding EDS mapping, and SAED patterns of t' , m' , and m phases of S7: (a) t' phase, (b) m' phase, and (c) m phase.

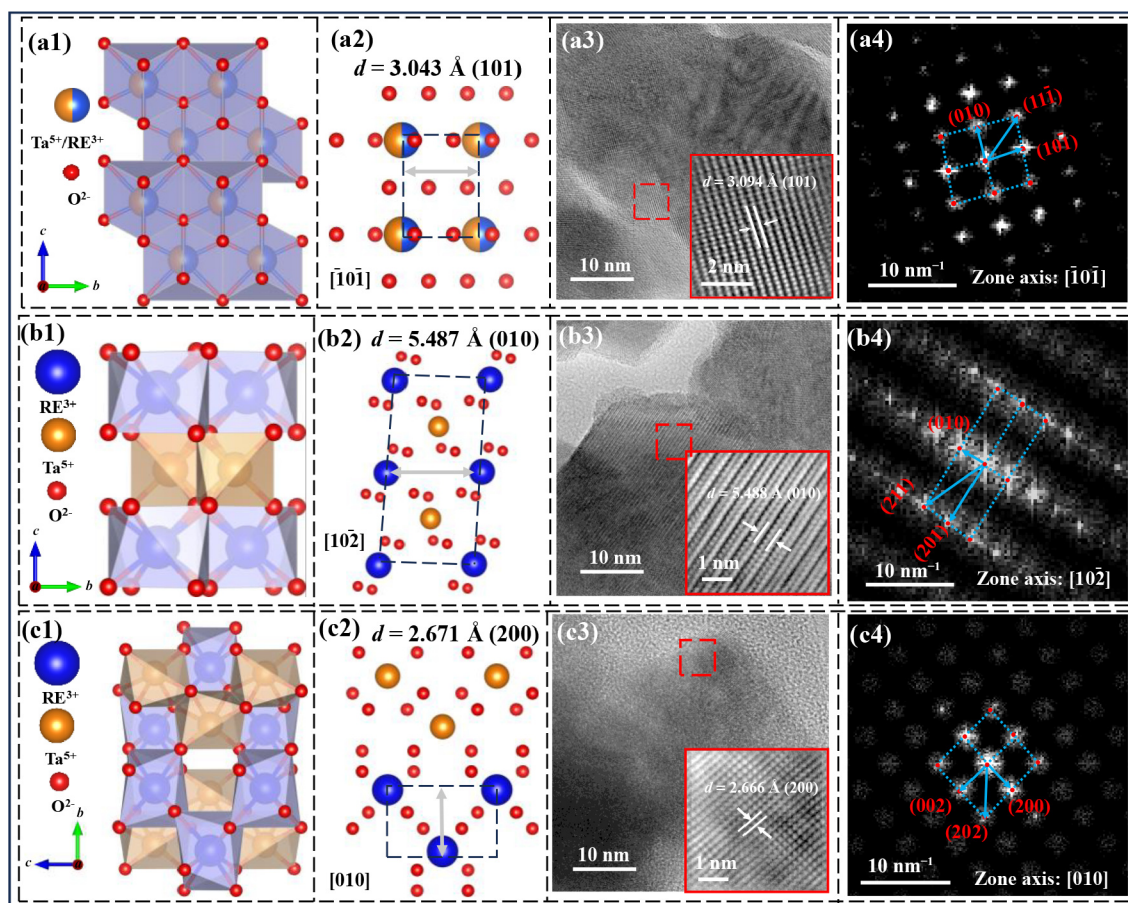


Fig. 11 Crystal structures, HR-TEM images, and SAED patterns of S7 with the t', m', and m phases: (a1)–(a4) t' phase, (b1)–(b4) m' phase, and (c1)–(c4) m phase.

with the particle size shown in Figs. 10(a) and 10(b), it can be observed that the powder size of S7 exhibits little variation, and they have an average size less than 100 nm, which is also consistent with the results shown in Figs. 7–9. The selected area electron diffraction (SAED) images prove the crystal structures of three samples. In Fig. 10(a), the interplanar spacing of the (103), (112), (102), (101), and (002) diffraction patterns is consistent with the results of t'-S7, and similar results are found in the m' (Fig. 10(b)) and m (Fig. 10(c)) phases of S7.

Figures 11(a1)–11(a4) present the interplanar spacing of the XRD refinement results and HR-TEM images, which correspond to the (101) plane of t'-S7. Notably, the lattice fringes of t'-phase S7, acquired via fast Fourier transformation (FFT), exhibit an ordered and uniform atomic arrangement, as observed in Fig. 11(a3). The above observation further confirms good consistency with the crystal structure derived from VESTA. Consistently, the SAED patterns show excellent agreement with the atomic projection of the corresponding crystal planes in Fig. 11(a2). The coherence between macroscopic phase analysis and atomic-level TEM characterization effectively verifies the accuracy of both the phase and structure of t'-phase S7. The HR-TEM image reveals a lattice fringe spacing of approximately 5.488 Å in Fig. 11(b3), which corresponds to the *d*-spaces of the (010) crystal plane of the m' phase. This result is near the refined interplanar spacing value ($d = 5.487$ Å), as shown in Fig. 11(b2), confirming the reliability of the data. Additionally, the SAED patterns further confirm its consistency with the refined atomic projection in Fig. 11(b4). Furthermore, the corresponding HR-TEM images, crystal structure information, and SAED patterns in Figs. 11(c1)–(c4) collectively demonstrate consistent interplanar spacings ($d = 2.671$ and 2.666 Å) of m-phase S7. It is obvious that

the atomic characterizations of TEM are consistent with our XRD results, and the crystal structures and microstructures are successfully tailored by changing the high-temperature heat treatment conditions in the current study.

Numerous studies have been reported on m' phase and m phase ceramics [4,17–19,56–61]. The m' phase exhibits a high hardness, low TECs, and low thermal conductivity, making it a promising candidate for protective coatings applied to substrates such as carbon composites and silicon carbide [4,56]. In contrast, m-phase ceramic possesses high TECs and a low thermal conductivity, making them suitable for the thermal protection of metal-based materials [4,17–19,56–61]. However, the performance of the t' phase and t phase remains scarce. As a high-temperature phase, t-phase ceramic is difficult to retain during cooling, which complicates the precise characterization of their properties. Reports on the t' phase are even sparser, and the literature provides only limited information regarding its application scope and performance characteristics. Further experimental evidence and theoretical calculations are therefore required to clarify its feasible application domains.

4 Conclusions

The multicomponent RETaO₄ nano powders are synthesized via the chemical co-precipitation method, and their crystals and microstructures are tailored by changing the heat-treatment temperatures. By adjusting the heat-treatment temperatures of S2–S7, the crystallinity degree, crystal structures, and particle sizes are regulated, and they maintain the nanoscale. Based on the comprehensive analysis presented above, the key conclusions are elaborated as follows:

(1) Based on the TG-DTG curve data, three distinct crystalline phases were successfully tailored by changing the heat-treatment temperatures, and the detailed temperatures were determined to be 800–1000 °C for the t' phase, 1000–1400 °C for the m' phase, and above 1400 °C for the m phase.

(2) The powder particles and grain morphology were successfully tailored by adjusting the annealing temperatures. Specifically, the grain sizes vary from the nanoscale (8.24 nm) to the micrometer scale (> 1 μm), and each rare-earth element is homogeneously distributed on the atomic scale in each ceramic.

(3) The optimal annealing parameter is determined to be 1000 °C based on the high crystallinity degree (89.5%) and nanoscale particle sizes (11.30 nm) of multicomponent RETaO₄ ceramics, and the synthesized nano powders can be further used to synthesize nano ceramics and coatings of RETaO₄ for various applications in future studies.

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Author contributions

Jiang Tian: Conceptualization, Methodology, Investigation, Formal Analysis, Writing—original draft; Lin Chen: Conceptualization, Methodology, Formal Analysis, Revised Draft; Xunlei Chen: Investigation; Jiankun Wang: Resources; Zipeng Gao: Investigation; Hao Xu: Investigation; Jing Feng: Conceptualization, Methodology, Investigation, Formal Analysis, Revised Draft.

Availability of data and materials

The data support that the findings of this study can be downloaded from the corresponding author upon reasonable request.

Competing interests

The authors have no competing interests to declare that are relevant to the content of this article. The author Jing Feng is the Editorial Committee member of this journal.

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