

Article

A Promising High-Entropy Thermal Barrier Material with the Formula $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$

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Abstract: YSZ has been widely used as a TBC material, but its phase change at high temperatures limits its development, thus the need for developing new thermal barrier materials resistant to high temperatures. Rare-earth aluminate ceramics with a garnet structure ($Yb_3Al_5O_{12}$) have been considered as a potential thermal barrier material. The melting point of $Yb_3Al_5O_{12}$ is 2000 °C, which has a potential high temperature application prospect. However, $Yb_3Al_5O_{12}$ has lower thermal expansion and higher thermal conductivity than YSZ, which is a widely employed thermal barrier coating (TBC) material. To overcome these obstacles, $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$, a high-entropy ceramic, was prepared by a solid-state reaction and pressureless sintering. The thermal conductivity of the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ was 3.48 W/(m·K) at 300 K, approximately 25.48% lower than that of the $Yb_3Al_5O_{12}$ (4.67 W/(m·K)). The thermal expansion coefficient of the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ was $9.28 \times 10^{-6} K^{-1}$ at 673–1273 K, approximately 18.52% higher than that of the $Yb_3Al_5O_{12}$ ($7.83 \times 10^{-6} K^{-1}$, 673–1273 K). When the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ was annealed at 1550 °C for 7 days, its average grain size only increased from 0.7 μm to 1.3 μm. Moreover, the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ exhibited better chemical stability and a lower grain growth rate than the $Yb_3Al_5O_{12}$. This study reveals that $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ is a promising candidate for the future generation of thermal barrier materials.

Keywords: rare-earth aluminates; high-entropy ceramics; thermal barrier material; chemical stability



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1. Introduction

Thermal barrier coatings (TBCs) are widely employed in gas turbine engines for aerospace, power generation, and marine applications and can protect the superalloy by reducing the alloy surface temperature and increasing the engine efficiency of gas turbines [1–6]. TBCs must be able to withstand extreme temperature cycling and thermal shock [7–9]. Therefore, the selection of TBCs is restricted by some basic requirements: (1) excellent high-temperature stability, (2) a thermal expansion coefficient (TEC) consistent with the metallic substrate, (3) a low grain growth rate, (4) low thermal conductivity, and (5) chemical inertness. To date, YSZ has been widely employed as a TBC material, but the limitations of phase transformation at a high temperature and high oxygen diffusivity make it unsuitable for the high operating temperature of gas turbine engines [10,11]. At

high temperatures, the non-equilibrium tetragonal phase in YSZ is prone to decomposition to generate t-phase and c-phase, and the cumulative effect of the volume expansion due to the phase change will cause cracks in the coating. Moreover, oxygen ions from YSZ above 800 °C tend to oxidize the metal substrate through the coating [12–14]. Therefore, YSZ cannot serve for a long time at temperatures above 1200 °C. Thus, novel TBC materials with better high-temperature stability are urgently needed.

The physicochemical properties of rare-earth elements have made them a research priority in the field of thermal barrier coatings [15–17]. Wang et al. reported that the thermal barrier coating of $\text{La}_{1.4}\text{Nd}_{0.6}\text{Zr}_2\text{O}_7$ (LNZ) on a Mo substrate was prepared by air plasma spraying with a self-developed LNZ thermal spray powder [18]. Xu et al. reported that $(\text{Sm}_{0.2}\text{La}_{0.8})_2(\text{Zr}_{0.7}\text{Ce}_{0.3})_2\text{O}_7$ (SmLZC), as a candidate material for novel thermal barrier coatings, was prepared by electron beam-physical vapor deposition (EB-PVD) [19]. Thus, rare-earth oxides exhibit good feasibility in the field of thermal barrier coatings. Therein, $\text{Yb}_3\text{Al}_5\text{O}_{12}$ exhibits excellent high-temperature stability, isotropic elastic properties, and low intrinsic thermal conductivity and has recently been recognized as a potential thermal barrier material [20–23]. The melting point of $\text{Yb}_3\text{Al}_5\text{O}_{12}$ is 2000 °C, which has a potential high-temperature application prospect. Moreover, theoretical results obtained by Klemm confirmed that $\text{Yb}_3\text{Al}_5\text{O}_{12}$ has better chemical stability than several other promising TBCs, such as $\text{Y}_2\text{Si}_2\text{O}_7$ and $\text{Yb}_2\text{Si}_2\text{O}_7$ [24]. However, $\text{Yb}_3\text{Al}_5\text{O}_{12}$ has lower expansion and higher thermal conductivity than YSZ, which is a widely used TBC material [22]. The linear thermal expansion of $\text{Yb}_3\text{Al}_5\text{O}_{12}$ was in the range from 298 to 1273 K, $7.83 \times 10^{-6} \text{ K}^{-1}$, which was lower than that of YSZ ($(10\text{--}11) \times 10^{-6} \text{ K}^{-1}$). The measured thermal conductivities of $\text{Yb}_3\text{Al}_5\text{O}_{12}$ at 300 and 1400 K are 4.67 W/(m·K) and 2.05 W/(m·K), which were greater than those of YSZ. Thus, decreasing the thermal conductivity of $\text{Yb}_3\text{Al}_5\text{O}_{12}$ and increasing the coefficient of thermal expansion of $\text{Yb}_3\text{Al}_5\text{O}_{12}$ are critical to expanding its applications.

Currently, the emergence of high-entropy ceramics (HECs) has attracted attention [25–30]. It has been reported that HECs efficiently enhance the thermal performance of thermal barrier materials because they have higher temperature stability, more consistent TECs, and a slower grain growth rate than single-component compounds. Lattice distortion and component disorder can increase phonon scattering and thus reduce thermal conductivity. In addition, better chemical stability can be achieved due to retarded diffusion. Some researchers have successfully synthesized different kinds of high-entropy oxides (HEOs) for TBC applications. An increase of 6 h in annealing results in only a 1 µm increase in grain size for $(5\text{RE}_{0.2})\text{Ta}_3\text{O}_9$, while this increases to approximately 3 µm for EuTa_3O_9 [31]. Sun et al. reported that the thermal conductivities of $(\text{Eu}_{0.2}\text{Er}_{0.2}\text{Lu}_{0.2}\text{Y}_{0.2}\text{Yb}_{0.2})_2\text{O}_3$ and $(\text{Sm}_{0.2}\text{Er}_{0.2}\text{Lu}_{0.2}\text{Y}_{0.2}\text{Yb}_{0.2})_2\text{O}_3$ are 5.10 W/(m·K) and 4.60 W/(m·K), which are 23.8% and 21.5% of that of Y_2O_3 (21.40 W/(m·K)), respectively [32]. Ren et al. prepared $(\text{Y}_{1/4}\text{Ho}_{1/4}\text{Er}_{1/4}\text{Yb}_{1/4})_2\text{SiO}_5$ silicate as a multifunctional TBC material, and it showed good resistance to high-temperature water vapor [33]. In addition, due to the excellent properties of HECs, Chen et al. reported a high-entropy rare-earth aluminate $(\text{Y}_{0.2}\text{Yb}_{0.2}\text{Lu}_{0.2}\text{Eu}_{0.2}\text{Er}_{0.2})_3\text{Al}_5\text{O}_{12}$ [34]. Thus, HECs can effectively enhance the combined properties of thermal barrier materials.

Herein, we design and synthesize a promising high-entropy rare-earth aluminate ceramic with garnet, $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$, by the solid-state reaction method. The standards for the component design are as follows: (1) to guarantee the formation of solid solutions, differences in the size of the rare-earth ions must not be greater than 15.0%; (2) each component must have the structure of garnet; and (3) no phase change may occur at the working temperature. In addition, in terms of the selected elements, we try to use low-cost rare-earth elements to further reduce the economic problem. The thermal expansion coefficient, thermal conductivity, high-temperature stability, chemical inertness and hardness of $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ are studied in the present work. These properties are valuable and beneficial to the potential application of $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ as the future generation of thermal barrier materials.

2. Experimental Section

2.1. Synthesis of $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$

Y_2O_3 (99.99%), Dy_2O_3 (99.99%), Ho_2O_3 (99.99%), Er_2O_3 (99.99%), Yb_2O_3 (99.99%), and Al_2O_3 (99.999%) were purchased from Aladdin Reagent Ltd., Co. (Shanghai, China). Figure 1 shows the synthesis of $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ via a solid-state reaction. The total molar ratio of rare-earth elements to Al was 3:5. First, five rare-earth oxides with equal molar ratios were mixed with Al_2O_3 and alcohol, and ground at 360 r/min for 10 h using a planetary ball mill. Afterward, the obtained solution was dried in an oven at 80 °C for 10 h. The obtained powder mixture was pressed and molded at 5 MPa. Afterwards, the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ powders were synthesized by sintering at 1600 °C for 2 h. The as-prepared powder was ball-milled for 10 h and then dried and sieved through a 400 mesh sieve to obtain fine particles. The samples were densified by the pressureless sintering method. The process of cold isostatic pressing is that the powder is pressed through a mold, then pressed again in a cold isostatic press at a pressure of 200 MPa to make itself more dense, and then calcined in a muffle furnace at a temperature of 1650 °C for 5 h. Finally, the densified bulk samples can be obtained.

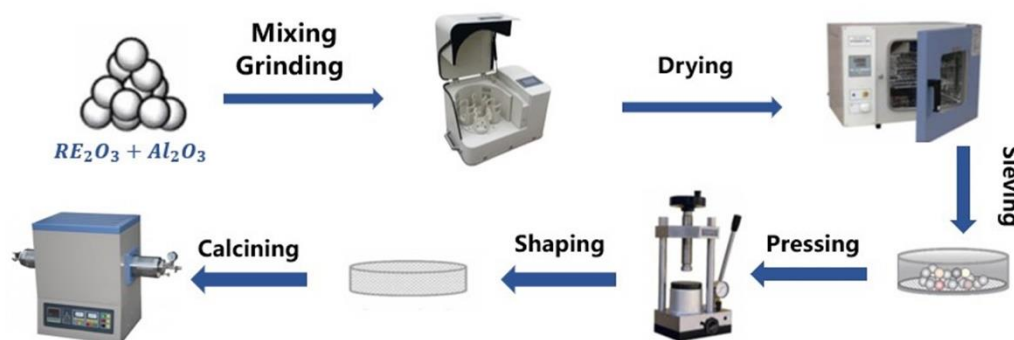


Figure 1. Schematic illustration of the preparation process of $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ by the solid-state reaction method and the pressureless sintering method.

2.2. Characterization of the Materials

A scanning speed of 2°/min was used for X-ray diffraction (XRD, Miniflex 600 instrument, Tokyo, Japan) with Cu K α radiation to determine the phase structure of the powder samples. Scanning electron microscopy (SEM, Apreo S LoVac Thermo Fisher Scientific, Waltham, MA, USA) was combined with field-emission transmission electron microscopy (TEM, FEI/Talos F200X G2, Lincoln, NE, USA) and energy-dispersive spectrometry (EDS) to examine the morphology and chemical composition of the samples. The TEC of the bulk samples was determined by a thermal expansion meter (Cryoall C15V, Beijing, China) with a size of Φ 30.0 mm $\times$ 2.0 mm. The thermal conductivity of the bulk samples was examined by a thermal constant analyzer (TPS 2500, Hot Disk, Göteborg, Sweden) with a size of Φ 6.0 mm $\times$ 10.0 mm. The thermal conductivities of the dense materials were corrected by the following Formula (1) [35].

$$\frac{K_S}{K} = 1 - 1.5P \quad (1)$$

where K_S is the thermal conductivity obtained by the test, K is the thermal conductivity of the fully dense materials, and P is the porosity.

A Vickers hardness test was conducted using a micro hardness tester (FT FM-700, Tokyo, Japan). The samples for Vickers hardness testing were cut by a diamond saw from the as-prepared bulk material and then polished down to 1.0- μ m diamond grits. The Vickers hardness number was determined by a microhardness tester with loads of 1, 3, 5,

and 10 N, respectively. Loads of 1, 3, 5, and 10 N were applied with a dwell time of 15 s. The Vickers hardness tests were calculated by using Formula (2).

$$H_v = P/S = 2P \sin\left(\frac{\theta}{2}\right) / d^2 = 1854.4P/d^2 \quad (2)$$

where P is the load, and d is the diagonal length of indentation. Five measurements for each load were taken to obtain the average value of hardness.

The cell dimension was calculated by the Rietveld refinement using GSAS software (EXPGUI-GSAS), and the reliability factors of R_p and R_{wp} were calculated by using Formulas (3) and (4) [36].

$$R_p = \sum |Y_{oi} - Y_{ci}| / \sum Y_{oi} \quad (3)$$

$$R_{wp} = \left[\sum W_i (Y_{oi} - Y_{ci})^2 / \sum W_i Y_{oi}^2 \right]^{1/2} \quad (4)$$

The Archimedes method was used to measure the bulk density (ρ), while Formula (5) was used to calculate the theoretical density.

$$\rho_{th} = m/v \quad (5)$$

where m is the mass of a unit cell of $\text{HE-RE}_3\text{Al}_5\text{O}_{12}$ and V is the cell volume. The relative density was calculated from the ratio of the actual density to the theoretical density (ρ/ρ_{th}).

The concentrations of dissolved ions in the leachates were determined by inductivity-coupled plasma mass spectrometry (ULTIMA 2, Horiba Jobin Yvon S.A.S, Edison Township, NJ, USA). A leaching test of the bulk samples by atmospheric pressure sintering at 1640 °C for 5 h was performed in a nitric acid solution in PTFE for 1–5 d. The $\text{HE-RE}_3\text{Al}_5\text{O}_{12}$ ceramics, which had a diameter of 15 mm and a height of 2 mm, were polished, cleaned, and dried. The ratio of the geometric surface area of the sample to the volume of the leachate was maintained at 1:10 cm^{-1} . The normalized element mass leaching rates (LR_i) of the (Al) were obtained based on Formula (6) [37].

$$LR_i = C_i \cdot V / f_i \cdot S \cdot t \quad (6)$$

where C_i is the concentration of elements (Al) in the leaching solution, V is the volume of the leaching solution, f_i is the proportion of element I in the ceramic, S is the surface area of the ceramic, and t is the date of leaching.

3. Results and Discussion

Figure 2a shows the XRD patterns of $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ at different temperatures (1500 °C, 1550 °C, 1600 °C). As can be seen from Figure 2a, when the synthesis temperature is 1500 °C or 1550 °C, impurity phases are $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})\text{AlO}_3$ and Al_2O_3 . Figure 2b shows the XRD patterns of $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ at 1600 °C for 2 h, with those of the single-component $\text{RE}_3\text{Al}_5\text{O}_{12}$ ($\text{RE} = \text{Y, Dy, Ho, Er, Yb}$) obtained from JCPDF cards. As can be seen from Figure 2b, when the green bodies were heated to 1600 °C in air with a dwell time of 2 h, no other impurity peak phase is observed. The XRD patterns of the $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ powder matched those of a single component, indicating that the $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ synthesized in this study had the same structure as a single component. No other contaminants were indicated by the diffraction peaks, confirming that a solid solution was prepared. Thus, a pure-phase $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ solid solution was successfully prepared. Figure 3a shows the XRD patterns of the calcined powder and the sintered solid. The XRD patterns were consistent before and after calcination, indicating that no impurities were introduced. The absence of holes and cracking can be seen in the SEM image in Figure 3b, indicating that the sintered $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ exhibited a high relative density.

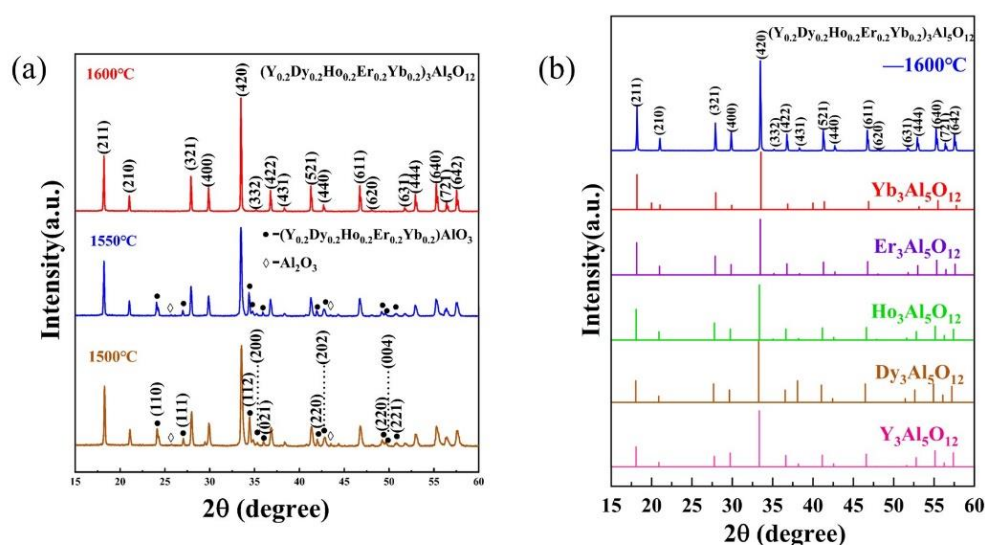


Figure 2. (a) XRD patterns of $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ at different temperatures (1500 °C, 1550 °C, 1600 °C); (b) XRD patterns of $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ at 1600 °C for 2 h with those of single-component $RE_3Al_5O_{12}$ (RE = Y, Dy, Ho, Er, Yb) obtained from JCPDF cards.

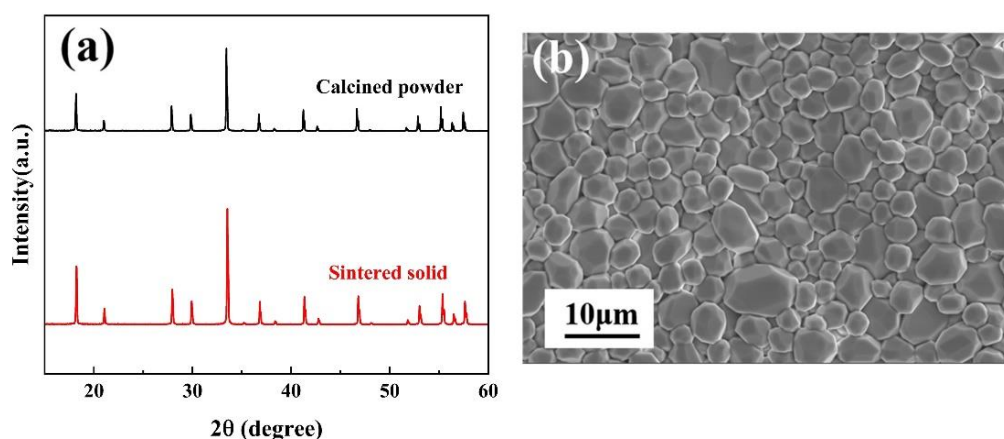


Figure 3. (a) XRD patterns of calcined powder and the sintered solid; (b) surface SEM image of $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$.

As shown in Figure 4, the cell dimension of the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ was calculated by the Rietveld refinement using GSAS software, and the reliable factors calculated via the equations are $R_p = 3.38\%$ and $R_{wp} = 4.41\%$. The refinement results are generally considered reliable for values of R_p and R_{wp} below 10.00%. The $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ ceramic lattice parameters were found to be $a = b = c = 11.992 \text{ \AA}$, which is approximately equal to the average cell size of the five single components ($11.986 \text{ \AA}$). Table 1 shows the refined cell dimensions and theoretical density of $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ and the five single-component garnets obtained from jade. The theoretical density of $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ was 6.02 g/cm^3 , based on the refined cell parameters. The sintered density, calculated by Archimedes' approach, is 5.88 g/cm^3 , 97.67% of the theoretical value (6.02 g/cm^3).

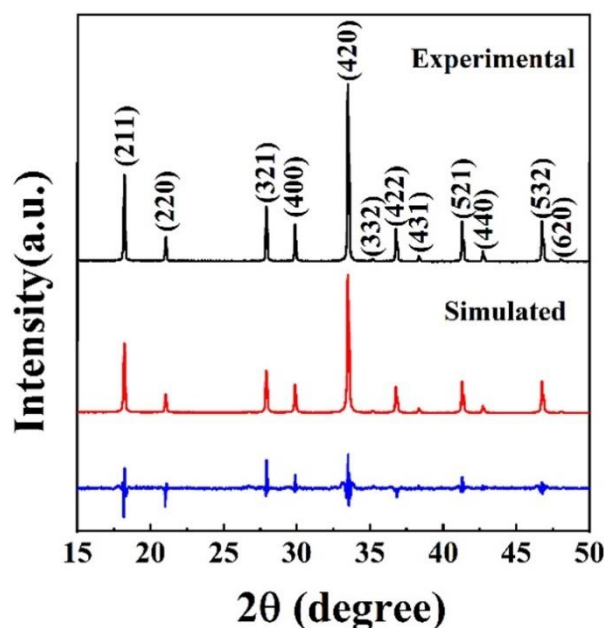


Figure 4. XRD patterns of the as-synthesized $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ powders, together with the Rietveld refinement data.

Table 1. Refined cell size and theoretical density of as-synthesized $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ and the five single component garnets.

Compounds	Theoretical Density (g/cm^3)	Relative Density	a (Å)
$(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$	6.02	97.70%	11.992
$Y_3Al_5O_{12}$	4.56	-	12.000
$Dy_3Al_5O_{12}$	6.20	-	12.038
$Ho_3Al_5O_{12}$	6.32	-	12.000
$Er_3Al_5O_{12}$	6.43	-	11.962
$Yb_3Al_5O_{12}$	6.62	-	11.930

Figure 5 shows the high-angle annular dark-field (HAADF) image and element mapping of the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ powders. The distribution of elements is clearly uniform, demonstrating the uniformity of the solid solution. The XRD and TEM–EDS results suggest that single-phase aluminates with excellent chemical uniformity were synthesized by the solid-state reaction approach. Figure 6a shows that the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ ceramics were further characterized by HR-TEM. Two positions were selected to calculate the lattice fringes, which exhibited typical (211) and (420) lattice planes with D-spacings of 0.488 nm and 0.267 nm, respectively. These results are consistent with the broad XRD peaks of the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ at 18.2° (0.48665 nm, (211)) and 33.5° (0.26662 nm, (420)). The selected area electron diffraction (SAED) pattern of the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ powders (Figure 6b) shows that $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ has a single-crystal ordered garnet structure.

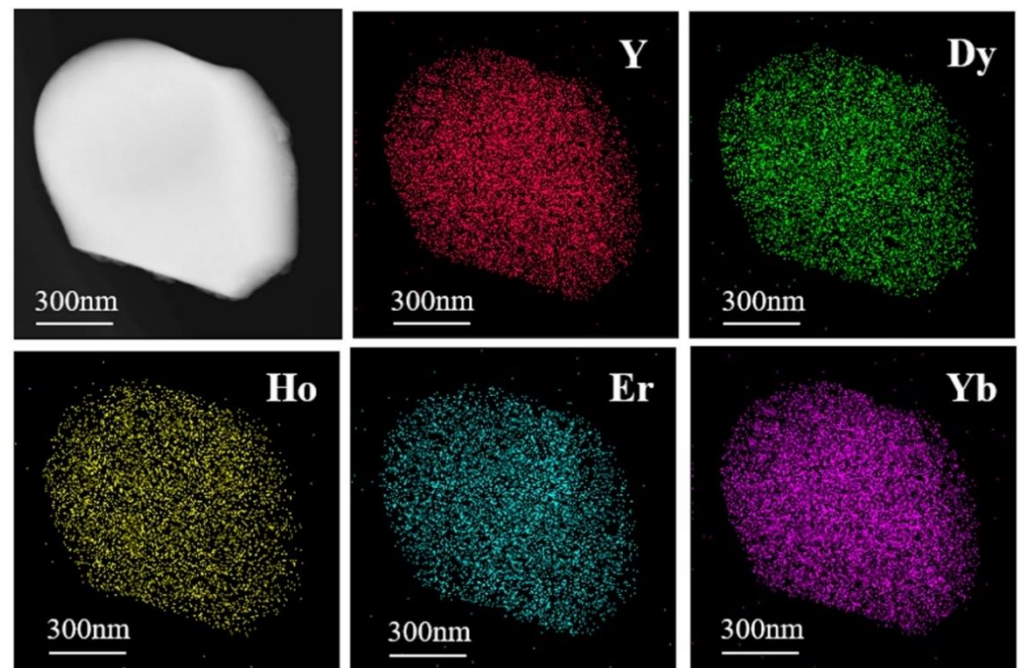


Figure 5. Elemental compositions of Y, Dy, Ho, Er, Yb in HAADF images.

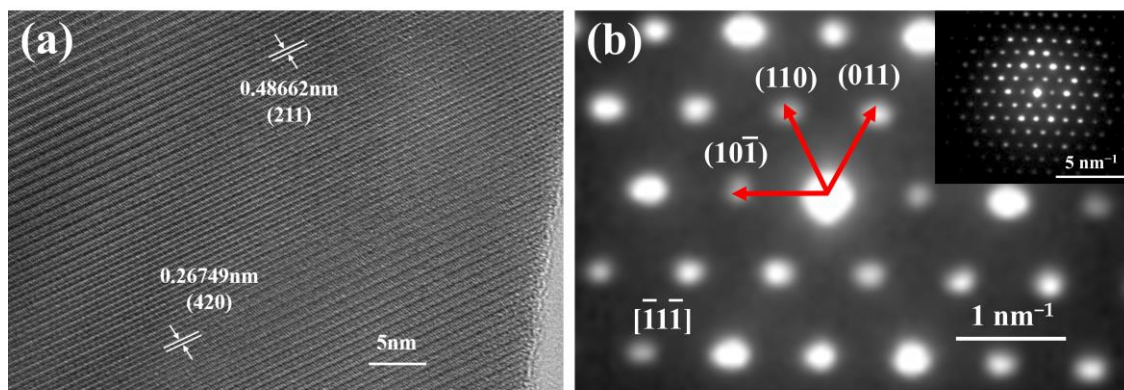


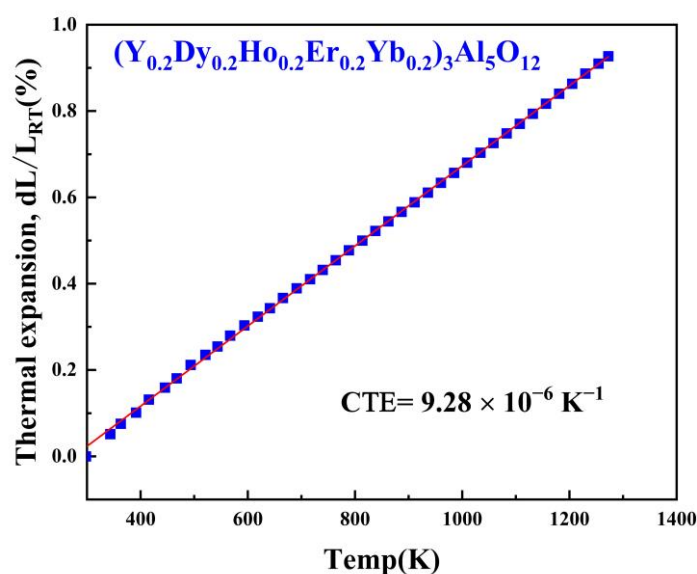
Figure 6. The power of (a) the high-resolution TEM image and (b) the corresponding SAED pattern.

Thermal conductivity is essential for assessing the thermal insulation properties of insulating materials. Table 2 shows the thermal conductivity of the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$, $(Y_{0.2}Yb_{0.2}Lu_{0.2}Eu_{0.2}Er_{0.2})_3Al_5O_{12}$, and $Y_3Al_5O_{12}$ at 300 K. The thermal conductivity of the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ ceramics was found to be 3.48 W/(m·K) at 300 K, approximately 25.48% and 8.66% lower than that of $Yb_3Al_5O_{12}$ (4.67 W/(m·K)) and $(Y_{0.2}Yb_{0.2}Lu_{0.2}Eu_{0.2}Er_{0.2})_3Al_5O_{12}$ (3.81 W/(m·K)), indicating that $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ is a potential thermal barrier material [32]. The thermal conductivities of rare-earth aluminates are slightly higher than commercial TBC material YSZ, which is 2.50–3.20 W/(m·K) [38]. Five equimolar rare-earth cations are present in $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ at crystallographic sites randomly occupying the same lattice. The mismatch between the masses and radii of these cations leads to large lattice distortions and intense phonon scattering in the material. Among the four core effects of high-entropy materials is lattice distortion, which may play a significant role in the decrease in the thermal conductivity of garnets [39].

Table 2. The thermal conductivity of the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ and $Yb_3Al_5O_{12}$ at 300 K.

	$(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$	$Yb_3Al_5O_{12}$	YSZ
Thermal conductivity (W/(m·K))	3.48	4.67	2.50–3.20

Thermal barrier materials need to possess a coefficient of thermal expansion similar to that of a high-temperature alloy matrix because a thermal expansion mismatch can cause cracking of the matrix. Figure 7 shows the curves of the linear thermal expansion of $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$, determined from room temperature to 1273 K. The relative changes in the length of the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ samples were recorded with the increasing temperature.

**Figure 7.** The linear thermal expansion coefficient of polycrystalline $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$.

In addition, the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ was designed using a pressureless sintering method, with a coefficient of thermal expansion of $9.28 \times 10^{-6} K^{-1}$ at 673–1273 K, approximately 18.52% higher than that of $Yb_3Al_5O_{12}$ ($7.83 \times 10^{-6} K^{-1}$, 673–1273 K), and approximately 8.67% higher than that of $(Y_{0.2}Yb_{0.2}Lu_{0.2}Eu_{0.2}Er_{0.2})_3Al_5O_{12}$ ($8.54 \times 10^{-6} K^{-1}$, 673–1273 K); thus, this material can be applied to various substrates, such as typical high-temperature nickel-based alloys [40].

A low grain growth rate is very important for TBC materials, as it improves the crack resistance due to thermal stress and prevents an increase in thermal conductivity. Figure 8 shows the average grain sizes of the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ ceramic specimens after annealing at 1550 °C for 7 days. As seen in Figure 8, the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ ceramics have a lower average grain rate than the $Yb_3Al_5O_{12}$ and $Er_3Al_5O_{12}$. The grain growth rates of the $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$, $Yb_3Al_5O_{12}$, and $Er_3Al_5O_{12}$ samples were similar during the annealing period of 0–5 days at 1550 °C. However, when the annealing time was extended to 7 days, the grain growth rate of the high-entropy aluminate was less than that of the $Yb_3Al_5O_{12}$ and $Er_3Al_5O_{12}$ under the same conditions. As a result of sluggish diffusion in HECs, $(Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})_3Al_5O_{12}$ ceramics grow slowly because fine particles are retained under high temperatures [39].

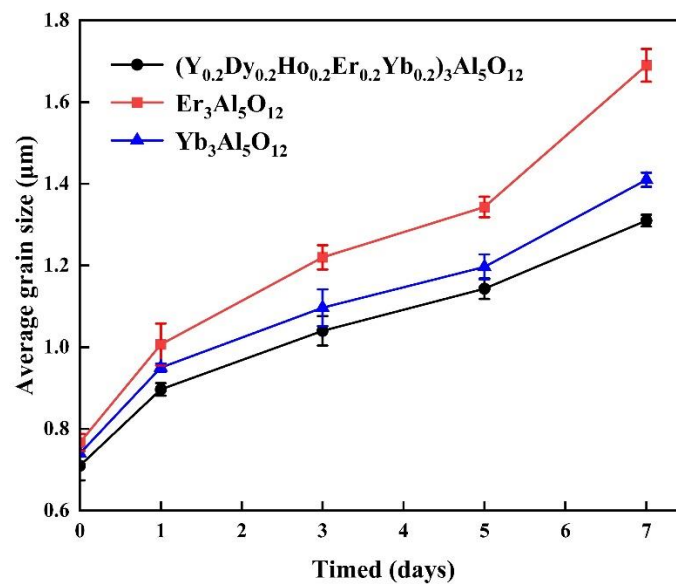


Figure 8. Average grain size of (Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})₃Al₅O₁₂, Er₃Al₅O₁₂, and Yb₃Al₅O₁₂ ceramics after annealing at 1550 °C for 7 days in air.

Thermal barrier materials must have excellent chemical inertness. Figure 9 shows the normalized elemental leaching rates of Al in (Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})₃Al₅O₁₂ and Yb₃Al₅O₁₂ over 5 days. The elemental aluminum leaching rate of the (Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})₃Al₅O₁₂ remained low during 1–5 days of corrosion in an acidic environment. However, the single-component garnet-structured aluminate ceramics exhibited a large leaching rate of elemental aluminum. This result indicated that (Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})₃Al₅O₁₂ ceramics have better chemical stability than Yb₃Al₅O₁₂. Therefore, high-entropy aluminate ceramics are promising materials for TBCs.

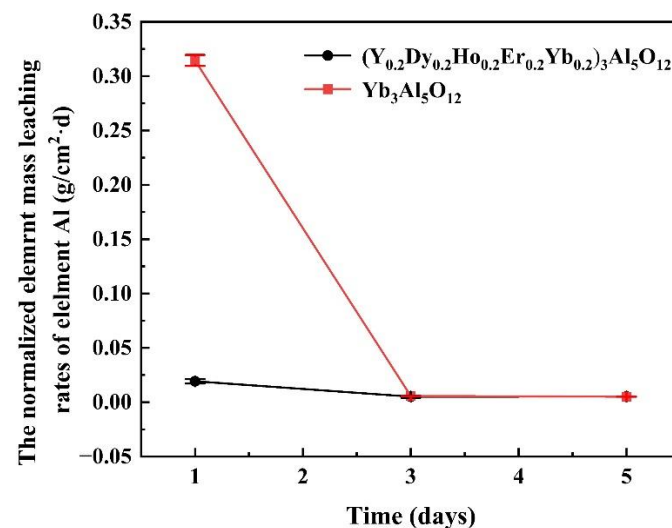


Figure 9. Normalized elemental leaching rates of Al in (Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})₃Al₅O₁₂ and Yb₃Al₅O₁₂ for 5 days.

The mechanical property of a TBC is also important as mechanical damage such as impact or wear can apply to the TBC. The mechanical property data of (Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})₃Al₅O₁₂ are also listed in Figure 10 as a comparison. As can be seen in Figure 10, the hardness of (Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})₃Al₅O₁₂ is significantly higher than that of Yb₃Al₅O₁₂. The hardness of (Y_{0.2}Dy_{0.2}Ho_{0.2}Er_{0.2}Yb_{0.2})₃Al₅O₁₂ is determined to be 13.3 GPa, which is higher

than that of $\text{Yb}_3\text{Al}_5\text{O}_{12}$ (10.7 GPa), and slightly lower than YSZ (14.0 GPa). The excellent mechanical property data suggest that $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ can be used as a potential thermal insulation material.

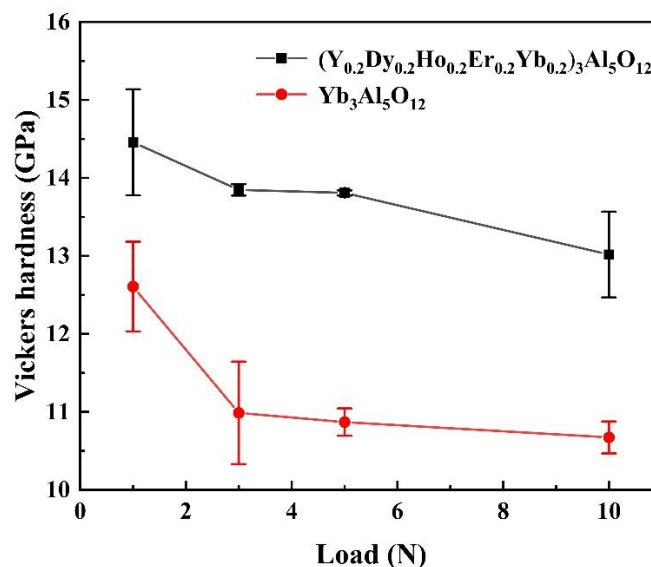


Figure 10. Vickers hardness of $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ and $\text{Yb}_3\text{Al}_5\text{O}_{12}$ versus load.

4. Conclusions

In summary, high-entropy rare-earth aluminate thermal barrier materials with garnet structures, $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ ceramics, were prepared by a solid-state reaction method. The results of XRD, SEM, and TEM analysis show that the synthesized $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ ceramic powder consists of a pure phase with a homogeneous rare-earth element distribution, and the lattice dimensions are $a = b = c = 11.992 \text{ \AA}$. According to the refined dimensions of the cell, the theoretical density of the $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ was calculated to be 6.02 g/cm^3 . The sintered density calculated by Archimedes' approach is 5.88 g/cm^3 , which is 97.67% of the theoretical value. The thermal conductivity of $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ ceramics is $3.48 \text{ W/(m}\cdot\text{K)}$ at 300 K, approximately 25.48% and 8.66% lower than that of $\text{Yb}_3\text{Al}_5\text{O}_{12}$ ($4.67 \text{ W/(m}\cdot\text{K)}$) and $(\text{Y}_{0.2}\text{Yb}_{0.2}\text{Lu}_{0.2}\text{Eu}_{0.2}\text{Er}_{0.2})_3\text{Al}_5\text{O}_{12}$ ($3.81 \text{ W/(m}\cdot\text{K)}$). The thermal expansion of $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ is $(9.28 \times 10^{-6} \text{ K}^{-1}, 673\text{--}1273 \text{ K})$, approximately 18.52% higher than that of $\text{Yb}_3\text{Al}_5\text{O}_{12}$ ($7.83 \times 10^{-6} \text{ K}^{-1}, 673\text{--}1273 \text{ K}$) and approximately 8.67% higher than that of $(\text{Y}_{0.2}\text{Yb}_{0.2}\text{Lu}_{0.2}\text{Eu}_{0.2}\text{Er}_{0.2})_3\text{Al}_5\text{O}_{12}$ ($8.54 \times 10^{-6} \text{ K}^{-1}, 673\text{--}1273 \text{ K}$). After annealing the $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ at $1550 \text{ }^\circ\text{C}$, its average grain size was found to be lower than that of $\text{Yb}_3\text{Al}_5\text{O}_{12}$ and $\text{Er}_3\text{Al}_5\text{O}_{12}$, due to the sluggish diffusion effects. As the annealing time was extended to 7 days, the average grain size of the high-entropy aluminate became much smaller than that of the $\text{Yb}_3\text{Al}_5\text{O}_{12}$ and $\text{Er}_3\text{Al}_5\text{O}_{12}$ under the same conditions. In addition, $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$ has excellent chemical stability compared with $\text{Yb}_3\text{Al}_5\text{O}_{12}$. The results show that $(\text{Y}_{0.2}\text{Dy}_{0.2}\text{Ho}_{0.2}\text{Er}_{0.2}\text{Yb}_{0.2})_3\text{Al}_5\text{O}_{12}$, in comparison with $\text{Yb}_3\text{Al}_5\text{O}_{12}$, is a potential thermal barrier material.

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