Investigation of thermophysical properties of ZrO2-Sm3TaO7 ceramics

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ABSTRACT
This study investigates rare earth RE3TaO7 ceramics and shows that these materials may be optimal for thermal barrier coatings. ZrO2-Sm3TaO7 ceramics were prepared through a solid-state reaction. X-ray diffraction and structural refinement revealed a phase structure with an ordered orthorhombic phase for the Ccmm space group. The degree of structural disorder increased with increasing ZrO2 content. Gaussian function fitting of the oxygen 1s X-ray photoelectron spectra showed that the Sm3+ and Ta5+ ions were replaced by Zr4+ ions. At high temperatures, 8 mol% ZrO2-Sm3TaO7 has a high thermal expansion coefficient (10.9 × 10−6 K−1). The thermal conductivities of ZrO2-Sm3TaO7 (1.17 – 1.75 W m−1 K−1) are lower than those of 7–8 wt.% yttria-stabilized zirconia, while those of 2% ZrO2-Sm3TaO7 are the lowest. The oxygen vacancies maintain the charge in equilibrium and enhance phonon scattering while decreasing the thermal conductivity. These results indicate that Sm3TaO7 can be used as a TBC.

1. Introduction
As a thermal barrier coating (TBC) material, yttria-stabilized zirconia (YSZ) is the most extensively used material for high-temperature applications. In addition to YSZ, the most studied TBCs are RE3Zr2O7 (rare earth (RE) ceramics. RE3Zr2O7 ceramics have low thermal conductivity, demonstrating their potential as effective TBC materials [1,2]. Several potential zirconate ceramics, such as Sm2Zr2O7, La2Zr2O7, Gd2Zr2O7, and Nd2Zr2O7, have been studied for this purpose [3–5]. For example, the thermal diffusivity and thermal conductivity of 4 mol% ZrO2-Y2O3 decreased with increasing Gd2O3 content. The fluoride-type Gd2Zr2O7 phase was formed as the second phase in the tetragonal ZrO2 matrix [6]. However, compared to YSZ, the thermal expansion coefficients of these zirconate ceramics are considerably low. As reported in [7], RE3Sm2O7 is also a type of pyrochlore oxide ceramic; however, its high thermal conductivity is not suitable for TBCs. RE3MO7 [8–11] (M+ = Mo5+, Sb5+, Nb5+, Ta5+, etc.) ceramics have excellent magnetic and dielectric properties. Based on previous investigations [12–15], RE3MO7 ceramics have low thermal conductivity because of lattice defects in their crystal structure that can enhance phonon scattering. In previous studies, ordered orthorhombic phases were observed in RE3 TaO7 ceramics (RE = Y–Dy), whereas the rest of the ceramics exhibited disordered cubic phases [16,17]. Because of their fluoro-type crystal structure, RE tantalate RE3TaO7 ceramics may be promising candidates for TBCs.

Recent studies have shown promising results that RE3TaO7 is an excellent ceramic material for use as a TBC. Using solid-state reactions, a series of RE3TaO7 (RE = Y, Eu, La, Gd, Nd, and Dy) ceramics were synthesized, and the results demonstrated that the RE3+ and Ta5+ ions are ordered in orthorhombic phases in RE3 TaO7 ceramics [18,19]. La2TaO7 and Nd3TaO7 ceramics exhibited an increase in thermal conductivity above 600°C. The minimum thermal conductivities of Y3 TaO7 [20] and ZrO2-Eu3TaO7 [21] ceramics reached 1.5 W m−1 K−1 and 1.37 W m−1 K−1, respectively. The thermal conductivities of TiO2-Sm3TaO7 ceramics were lower than those of Sm2YbTaO7, Sm2YTaO7, and Sm2 Zr2O7 ceramics [22,23]. Because of the unique properties of Sm [24], the application temperature of Sm2Zr2 O7 ceramics as TBCs exceeds 1500°C. Our previous study [25] on SmTaO4, Sm3TaO7, and SmTaO9 ceramics showed that Sm3TaO7 has a lower thermal conductivity than those of SmTaO2 and SmTa3O7 ceramics. The lower thermal conductivities of the Sm3TaO7 ceramics are related to their complex crystal structure and vacancy concentration (as high as 66.7%). Phase diagrams of ZrO2-SmO1.5-TaO2.5 are shown in Figure 1, which displays the isothermal section of the phase diagram at 1700°C derived from the ZrO2-YO1.5-TaO2.5 phase diagram at 1500°C [26]. The black spots in the blue area indicate the composition of the study.

In this study, ZrO2-Sm3TaO7 ceramics were prepared using a solid-state reaction. X-ray diffraction (XRD) and X-ray photoelectron spectroscopy (XPS) were conducted to investigate the crystal structure and solid-
solution mechanism of ZrO$_2$-Sm$_3$TaO$_7$. The Rietveld refinement method was used to correctly identify the crystal structures. The microstructure and mechanical properties of the samples were also studied, and the thermal properties (thermal expansion, thermal conductivity, predicted thermal conductivity, and mean phonon free path) were characterized. This research aims to understand the solid-solution mechanism and thermal physical properties of ZrO$_2$-Sm$_3$TaO$_7$ ceramics to promote the application of Sm$_3$TaO$_7$ ceramics in TBCs.

2. Experimental process

The x mol% ZrO$_2$-Sm$_3$TaO$_7$ ceramics (x = 0, 2, 4, 6, 8, and 10) were prepared using the solid-state reaction method. Sm$_3$O$_9$, Ta$_2$O$_5$, and ZrO$_2$ powders were ball-milled for 10 h at 360 rpm in C$_6$H$_{12}$. The powder was dried at 70°C for 10 h to obtain dehydration and alcohol removal. The dried powder was pressed into columns under 10 MPa for 5 min, and the columns were sintered at 1700°C for 10 h to synthesize ZrO$_2$-Sm$_3$TaO$_7$ ceramics.

The crystal structures were characterized by XRD (Rigaku, MiniFlex600, Japan) and General Structure Analysis System software. The lattice parameters were obtained using Rietveld refinement. The oxidation states were investigated using XPS (PHI 500, Thermo Fisher, USA). The bulk density ($\rho$) was measured using the Archimedes principle, and the porosity ($\varphi$) was calculated according to the following formula:

$$\varphi = 1 - \frac{\rho}{\rho_0}$$  \hspace{1cm} (1)

The microtopography of ZrO$_2$-Sm$_3$TaO$_7$ was observed using a secondary electron scanning electron microscope (SEM, JEOL, JSM-7001F, Japan). The longitudinal acoustic velocity ($u_l$) and transverse acoustic velocity ($u_t$) were measured using a UMS advanced ultrasonic modulus measurement system (TECLAB, France). The average acoustic velocity ($u_m$), shear modulus ($B$), Young’s modulus ($E$), and Poisson’s ratio ($\sigma$) were calculated according to the following formulas:

$$v_m = \left[1 + \frac{1}{2} \left(\frac{1}{\nu_1^2} + \frac{2}{\nu_3^2}\right)\right]^{-\frac{1}{2}}$$  \hspace{1cm} (2)

$$E = \rho u_t^2 \left(3u_t^2 - 4u_l^2\right) \left(u_t^2 + u_l^2\right)^{-2}$$  \hspace{1cm} (3)

$$B = \rho \left(u_t^2 - \frac{4}{3} u_l^2\right)$$  \hspace{1cm} (4)

$$\sigma = \frac{1 - 2 \left(\frac{\nu_1}{\nu_3}\right)^2}{2 - 2 \left(\frac{\nu_1}{\nu_3}\right)^2}$$  \hspace{1cm} (5)

The thermal expansion properties of ZrO$_2$-Sm$_3$TaO$_7$ were analyzed using a thermobalometric analysis instrument (NETZSCH TMA 402, Germany). The specific heat ($C_p$) was calculated by applying the Neumann-Kopp rule, using the specific heat of the constituent oxide and the bulk density. The thermal diffusivities were measured using a laser flash instrument (NETZSCH, LFA 457, Germany). The thermal conductivity was calculated based on the thermal diffusivity ($\alpha$), bulk density ($\rho$), and specific heat ($C_p$):

$$k' = \alpha \rho C_p$$  \hspace{1cm} (6)

Because the ZrO$_2$-Sm$_3$TaO$_7$ ceramic samples contain pores, the thermal conductivity ($k$) was corrected for the porosity of the sintered specimen ($\varphi$) using the following equation:

Figure 1. Sm$_{0.5}$Ta$_{0.5}$-ZrO$_2$ phase diagram at 1700°C. T = tetragonal, F = fluorite, C = cubic, and O = orthorhombic [26].
3. Results and discussion

3.1. XRD and solid solution mechanism

The XRD patterns of ZrO$_2$-Sm$_3$TaO$_7$ were compared with the JCPDF #36-0672 standard, as illustrated in Figure 2(a). The lack of evidence to suggest the precipitation of any secondary phase indicates that ZrO$_2$-Sm$_3$TaO$_7$ is comprised of a pure phase. The XRD 2θ peaks between 28° and 30° are shown in Figure 2(b), consistent with the JCPDF #36-0672 standard, as there are two peaks at 28.7° and 29°. As shown in Figure 2(b), the peak did not move to the right or left with increased ZrO$_2$. If Zr$^{4+}$ exists as an interstitial ion causing lattice expansion, the main peaks shift to the left. This indicates that ZrO$_2$ is completely solid-soluble in the lattice. The crystal structure of Sm$_3$TaO$_7$ is composed of TaO$_6$ octahedra and SmO$_6$ and SmO$_8$ polyhedra [27]. Therefore, the phase structures of ZrO$_2$-Sm$_3$TaO$_7$ are in agreement with those reported in the literature, demonstrating an ordered orthorhombic phase, and the space group is categorized as Ccmm. Figure 2(b) shows that diffraction peaks (202) and (022) broadened considerably with increasing ZrO$_2$ content. Based on the theory of X-ray diffraction, small crystal particles and a low degree of crystallinity lead to a wide diffraction peak. According to the literature, the change in crystal structure is in accordance with the ionic radius. The order–disorder status of Ln$_3$TaO$_7$ (Ln = La, Gd, Y, or Lu) is mainly determined by the ratio of the ion radius of lanthanum to that of tantalum, or $r$(Ln)/$r$(Ta) [28]. A similar phenomenon may occur in ZrO$_2$-Sm$_3$TaO$_7$ because the ionic radius of Sm$^{3+}$ is larger than that of Zr$^{4+}$. Doping Zr$^{4+}$ ions into Sm$_3$TaO$_7$ ceramics increases the value of $r$(Sm)/$r$(Ta), driving the structure toward disorder and reducing crystallinity.

Based on Ref [16,17], RE$_3$TaO$_7$ ceramics are ordered in the orthorhombic phase for RE = Y–Dy, while the rest is a disordered cubic phase. For RE$_3$TaO$_7$ ceramics with a disordered cubic phase, the cation consists of 1/4 Ta$^{5+}$ and 3/4 RE$^{3+}$, and no framework structure exists, which differs from that with an ordered phase. Therefore, Sm$_3$TaO$_7$ has three types of ordered orthorhombic phases with different space groups: the cation site is occupied by 1/4 Ta$^{5+}$ and 3/4 Sm$^{3+}$. The results of the Rietveld refinement XRD are shown in Figure 3(a–f). The results agree with the crystal structure of ZrO$_2$-Sm$_3$TaO$_7$ ceramics, which confirms the correctness of the structural model. The curve of the calculated lattice parameters with changing the doping content of ZrO$_2$ is shown in Figure 4. The lattice parameters changed little with increasing ZrO$_2$ doping, which is consistent with the XRD analysis. This is related to the solid-solution mechanism of Zr ions in the Sm$_3$TaO$_7$ substrate.

According to the effective ion radius (Sm$^{3+}$ = 1.08, Ta$^{5+}$ = 0.64, and Zr$^{4+}$ = 0.72), the ionic radii of Zr$^{4+}$ are smaller than those of Sm$^{3+}$, but larger than those of Ta$^{5+}$. If the Zr$^{4+}$ ions substitute only Sm$^{3+}$ ions, some oxygen will be absorbed to maintain the charge balance, and the change in the ionic radii will be increased by 0.68 Å. If the Zr$^{4+}$ ions only substitute Ta$^{5+}$ ions, the

\[
\frac{k'}{k} = 1 - \frac{3

\[\frac{\varphi}{2}
\]

(7)

\[\text{PDF-36-0672}
\]

Figure 2. XRD patterns of ZrO$_2$-Sm$_3$TaO$_7$ compared with PDF #36-0672: (a) 10° ≤ 2θ ≤ 70°; (b) 28° ≤ 2θ ≤ 30°.
change in the ionic radii will be decreased by 1.24 Å. The position of the X-ray diffraction peaks will significantly move if Zr$^{4+}$ substitutes for the sole Sm$^{3+}$ or Ta$^{5+}$ ions. Therefore, the Zr$^{4+}$ ions substitute in pairs for both Sm$^{3+}$ and Ta$^{5+}$ ions in the unit cell, and the difference in ionic radii is 0.28 Å, leading to the position of diffraction peaks remaining almost unchanged. Therefore, Zr$^{4+}$ ions mainly substitute in pairs for both the Sm$^{3+}$ and Ta$^{5+}$ ions in the unit cell.

The solid-solution mechanism of ZrO$_2$-Sm$_3$TaO$_7$ was investigated using XPS. The Gaussian function fitting of the O 1s XPS profiles is shown in Figure 5. According to Table 1, the binding energy of O is 529–532 eV. The O 1s profiles of ZrO$_2$-Sm$_3$TaO$_7$ can be matched with two Gaussian functions. The L1 and L2 curves represent the O1 and O2 positions of O 1s, respectively. With an increase in ZrO$_2$ content, there was a minor binding energy change in L1 and L2, as shown in Table 1. The same numbers of Sm$^{3+}$ and Ta$^{5+}$ ions are replaced by Zr$^{4+}$ ions, and the O1 and O2 positions of the O 1s spectra demonstrate the lowest electronegativity. Therefore, the XRD results, lattice refinement, and oxygen 1s XPS profiles revealed that the Sm$^{3+}$ and Ta$^{5+}$ ions were replaced by Zr$^{4+}$ ions. The solid solution mechanism of ZrO$_2$-Sm$_3$TaO$_7$ involves substitution, and the corresponding crystal cohesive energy can be expressed as Eq. (8):

$$4\text{ZrO}_2 \rightarrow \text{Zr}_4^{4+} + 3\text{Sm}^{3+} + 8\text{O}_2$$  \hspace{1cm} (8)

3.2. Microstructure and mechanical properties

The microstructures of ZrO$_2$-Sm$_3$TaO$_7$ are shown in Figure 6(a–f). The grain size of the samples was approximately 1–6 μm, and the grain boundaries...
were clearly visible. The densities and porosities of the samples are listed in Table 2. The densities of the samples were in the range of 7.31–7.92 g·cm⁻³. The porosity of ZrO₂-Sm₃TaO₇ increased with increased ZrO₂ content, and the porosity could be regulated by controlling the ZrO₂ content. Therefore, the realistic thermal properties of ZrO₂-Sm₃TaO₇ can be estimated by adding the appropriate ZrO₂ content. Compared with Sm₃TaO₇, ZrO₂-Sm₃TaO₇ has a lower density. The shear modulus and Young’s modulus of ZrO₂-Sm₃TaO₇ are shown in Figure 7(a). The ZrO₂ content in Sm₃TaO₇ reduces the Young’s and shear moduli of

Table 1. O 1s XPS data for ZrO₂-Sm₃TaO₇

| ZrO₂-Sm₃TaO₇ | L₁ binding energy (eV) | L₂ binding energy (eV) |
|-------------|------------------------|------------------------|
| 0%          | 531.19                 | 529.82                 |
| 2%          | 529.17                 | 531.01                 |
| 4%          | 529.21                 | 531.03                 |
| 6%          | 529.19                 | 531.02                 |
| 8%          | 529.23                 | 531.05                 |
| 10%         | 529.15                 | 530.94                 |

Figure 5. Oxygen 1s XPS profiles and corresponding Gaussian function-fitted curves of ZrO₂-Sm₃TaO₇.

Figure 6. Secondary electron microstructures of ZrO₂-Sm₃TaO₇.

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3.3. Thermophysical properties

Figure 8(a) shows the thermal expansion of ZrO$_2$-Sm$_3$TaO$_7$. All the ceramic data showed good linearity, except for those for 2 mol% ZrO$_2$ doping, caused by a phase transition. According to previous research on TiO$_2$-Sm$_3$TaO$_7$ ceramics [22], the phase transition of Sm$_3$TaO$_7$ ceramics is eliminated via the TiO$_2$ alloying effect. The TEC of the samples increased with increasing temperature and ZrO$_2$ content, as shown in Figure 8(b). The 8 mol% ZrO$_2$-Sm$_3$TaO$_7$ sample has a high TEC of $10.9 \times 10^{-6}$ K$^{-1}$, which is close to that of 7–8 wt.% YSZ ($10 \sim 11 \times 10^{-6}$ K$^{-1}$) and higher than that of Sm$_3$TaO$_7$ ($9.8 \times 10^{-6}$ K$^{-1}$, 1200°C). As discussed in previous studies [30,31], the thermal expansion of the samples can be explained by the asymmetric anharmonic vibrations of chemical bonds. Generally, the macrothermal expansion of bulk ceramics originates from the crystal binding energy. The longer the bond, the lower the bond energy. The bond lengths of ZrO$_2$-Sm$_3$TaO$_7$ decrease with increasing ZrO$_2$ content, and the thermal expansion is mainly affected by the Sm–O bond. The measurement of the Young’s modulus shows that the value decreases with increasing ZrO$_2$ content.

The specific heat of ZrO$_2$-Sm$_3$TaO$_7$ was calculated at various temperatures and is shown in Figure 9(a), increasing slowly with temperature and ZrO$_2$ content. The specific heat exhibited a nearly linear temperature dependence. Figure 9(b) shows that the thermal diffusivity curves follow a typical T$^{-1}$

Table 2. Density and porosity of ZrO$_2$-Sm$_3$TaO$_7$ ceramics.

| ZrO$_2$-Sm$_3$TaO$_7$ | 0% | 2% | 4% | 6% | 8% | 10% |
|----------------------|----|----|----|----|----|-----|
| Density (g/cm$^3$)   | 7.92 | 7.31 | 7.45 | 7.33 | 7.32 | 7.32 |
| Porosity (%)         | 1.4 | 9.0 | 7.2 | 6.9 | 8.7 | 8.9 |

ZrO$_2$-Sm$_3$TaO$_7$, which are lower than those of 7–8 YSZ (205 GPa) [29]. It is inferred that the ZrO$_2$ alloying effect may result in a decrease in the covalent interaction and a decrease in the elastic modulus of Sm$_3$TaO$_7$ ceramics. The Poisson ratio of the samples is shown in Figure 7(b), as it decreases with increasing ZrO$_2$ content, showing a similar trend to the Young’s and shear moduli.

Figure 8. Thermal expansion of ZrO$_2$-Sm$_3$TaO$_7$. (a) deformation variables and (b) thermal expansion coefficients.
The profile, indicating anharmonic phonon scattering. The thermal conductivities of the curve as a function of temperature and ZrO₂ amount are shown in Figure 9(c,d), respectively. The thermal conductivity decreased as the ZrO₂ content increased. At 900°C, 2 mol% ZrO₂-Sm₃TaO₇ has a lower thermal conductivity (1.17 W·m⁻¹ K⁻¹) than that of Sm₃TaO₇ (1.34 W·m⁻¹ K⁻¹) and 7–8 wt% YSZ (2.7 W·m⁻¹ K⁻¹) [32,33]. The thermal conductivities of 6%, 8%, and 10% ZrO₂ first decreased with increasing temperature and then increased. The thermal conductivity changes gradually at high temperatures when it becomes weakly independent of temperature. Because the Sm³⁺ and Ta⁵⁺ ions are replaced by Zr⁴⁺ ions, 1/8 of the oxygen vacancies will maintain the charge in equilibrium. According to previous research on ZrO₂-Dy₃TaO₇ ceramics [34], the stronger local non-harmonic vibration of phonons is caused by the oxygen vacancies, making the thermal conductivity of ZrO₂-Sm₃TaO₇ ceramics show weak temperature dependence at high temperature. Because of the large amount of doping, ZrO₂ not only enters the lattice, but also reduces the number of oxygen vacancies. To reduce the existence of oxygen vacancies, the thermal conductivity of the materials increases. Therefore, the 2% ZrO₂-Sm₃TaO₇ ceramics exhibited the lowest thermal conductivity at 900°C.

Based on the phonon–phonon scattering theory, the thermal conductivity can be calculated using the Debye temperature (θ_D), Boltzmann constant (B), proportionality constant (C), Planck constant (h), average acoustic velocity (υ_m), and absolute temperature (T):

$$\kappa = \frac{\theta_D B^2}{2\pi^2 C h \nu_m T}$$  \hspace{1cm} (9)

According to defect scattering theory, ceramics show low thermal conductivity owing to the existence of oxygen vacancies, which can enhance phonon scattering and reduce phonon mean free paths. The calculation results for the phonon mean free paths are shown in Figure 10(a). Because the Sm³⁺ and Ta⁵⁺ ions are replaced by Zr⁴⁺ ions, 1/8 of the oxygen vacancies will maintain the charge in equilibrium. The decrease in thermal conductivity is usually attributed to oxygen vacancies.

To investigate the influence of the doping process on the thermal conductivity, the predicted and experimental thermal conductivities were analyzed. Based on the Debye model, the thermal diffusivity can be calculated using the phonon mean free path (λ) and the average sound velocity (υ_m) as follows:

$$\alpha = \frac{1}{3} \nu_m$$  \hspace{1cm} (10)
Parameter $\Lambda$ can be calculated by defect scattering ($\Lambda_{\text{defect}}$), Umklapp phonon-phonon scattering ($\Lambda_{\text{phonon}}$), grain boundary scattering ($\Lambda_{\text{boundary}}$), and other types of scattering:

$$\frac{1}{\Lambda} = \frac{1}{\Lambda_{\text{phonon}}} + \frac{1}{\Lambda_{\text{defect}}} + \frac{1}{\Lambda_{\text{boundary}}} + \sum_{i} \frac{1}{\Lambda_i} \quad \text{(11)}$$

According to crystalline solid theory, $\Lambda$ can be estimated using the pre-exponential factor ($\Lambda_0$), Debye temperature ($\Theta_D$), and number of atoms per primitive cell ($n$) [35]:

$$\Lambda = \Lambda_0 \left[ \exp \left( \frac{\Theta_D}{b T^{1/3}} \right) \right]^{1/2} \quad \text{(12)}$$

where $b$ is a constant of $\approx 2$. At high temperatures ($T > \Theta_D/bn^{1/3}$), $\Lambda$ can be approximated by the following simple formula [36]:

$$\frac{1}{\Lambda} = \frac{1}{\Lambda_0} \left[ \exp \left( \frac{\Theta_D}{b T^{1/3}} \right) - 1 \right]^{-1} + D$$

$$= \frac{b T}{\Theta_D} C \left[ 1 + \frac{1}{2} \left( \frac{\Theta_D}{b T^{1/3}} \right) + \frac{1}{12} \left( \frac{\Theta_D}{b T^{1/3}} \right)^2 + \ldots \right] + D$$

### Table 3. Fitted inverse thermal diffusivities of ZrO$_2$-Sm$_3$TaO$_7$.

| ZrO$_2$-Sm$_3$TaO$_7$ | $\alpha^{-1}_{\text{experiment}}$ | C | D |
|-----------------------|---------------------------------|---|---|
| 0%                    | 1156 T + 1505050                | 71,268 | 1,540,684 |
| 2%                    | 1046 T + 1461430                | 63,488 | 1,493,174 |
| 4%                    | 743 T + 1500100                 | 46,802 | 1,523,501 |
| 6%                    | 745 T + 1696830                 | 43,847 | 1,718,754 |
| 8%                    | 729 T + 1718550                 | 46,375 | 1,741,737 |
| 10%                   | 765 T + 1708020                 | 47,664 | 1,731,852 |

$$\approx C \left[ \left( \frac{b T^{1/3}}{\Theta_D} \right)^{b T} \right]^{-1} + D = \left( \frac{b C^{1/3}}{\Theta_D} \right) T + \left( D - \frac{1}{2} C \right)$$

Therefore, Equation (13) can be modified as

$$= \left( \frac{1}{\Lambda_0} \right) \left( \frac{b C^{1/3}}{\Theta_D} \right) T + \left( D - \frac{1}{2} C \right) \quad \text{(14)}$$

We can linearly fit the data of $1/\alpha$ with respect to temperature and calculate parameters $C$ and $D$ from the values of slope and intercept. Equation (14) shows that $1/\alpha$ is linear in temperature. The temperature dependence is dominated by Umklapp phonon-phonon scattering. The linear fitting of $1/\alpha$ is shown in Figure 10(b), and the fitted parameters $C$ and $D$ are listed in Table 3. The experimental and predicted values are presented in Figure 10(c). Thermal radiation is increased at high temperatures, resulting in a higher experimental thermal conductivity than the calculated data. As the doping of ZrO$_2$ increased, the thermal...
conductivity decreased, and the experimental and calculated values tended to be close to each other, as the Sm$^{3+}$ and Ta$^{5+}$ ions were replaced by Zr$^{4+}$ ions, while the oxygen vacancy maintained the charge in equilibrium, increased phonon scattering, and reduced thermal conductivity.

4. Conclusion

In this study, ZrO$_2$-Sm$_3$TaO$_7$ ceramics were prepared using a solid-state reaction method. The XRD profiles and structural refinement results show that the phase structures of ZrO$_2$-Sm$_3$TaO$_7$ possess an ordered orthorhombic phase with the Ccmm space group. The degree of structural disorder increased with increasing ZrO$_2$ content. The Gaussian function fitting of the O 1s XPS profile shows that Sm$^{3+}$ and Ta$^{5+}$ ions are substituted by Zr$^{4+}$ ions. Meanwhile, 8 mol% ZrO$_2$-Sm$_3$TaO$_7$ has a high TEC of 10.9 × 10$^{-6}$ K$^{-1}$ at high temperatures. The thermal conductivities of ZrO$_2$-Sm$_3$TaO$_7$ range from 1.17 to 1.75 W m$^{-1}$K$^{-1}$, which is lower than that reported for 7–8 wt% YSZ, while 2% ZrO$_2$-Sm$_3$TaO$_7$ had the lowest thermal conductivity. The solid-solution mechanism revealed that the Sm$^{3+}$ and Ta$^{5+}$ ions are replaced by Zr$^{4+}$ ions. The oxygen vacancies maintain charge in equilibrium and enhance phonon scattering. Meanwhile, the decrease in the thermal conductivity was attributed to oxygen vacancies. These results indicate that Sm$_3$TaO$_7$ can be used as a TBC.

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Disclosure statement

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