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Original Article

Material-genome perspective towards tunable thermal expansion of rare-earth di-silicates

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ABSTRACT

Optimization of thermal expansion coefficient (CTE) mismatch among constituent rare-earth silicate layers is a critical challenge for the optimal multilayer thermal/environmental barrier coating (T/EBC) architecture for SiC_f/SiC_m CMCs. In this study, thermal expansion properties for β-, γ- and δ-$RE_2Si_2O_7$ polymorphs are investigated via DFT calculations. The interaction between rare-earth (RE) atoms and neighboring bridging oxygen (O_B) atoms, as well as the structure of [$O_3Si—O_B—SiO_3$] pyrosilicate units are the characteristic "gene" that controls the positive or negative contribution from low-frequency vibration patterns to the overall phonon anharmonicity, and eventually determine the significantly different CTE of $RE_2Si_2O_7$ polymorphs. Inspired by the concept of "genome modification", γ-$(Dy_{0.15}Y_{0.85})_2Si_2O_7$ solid solution is designed and synthesized, which shows notable enhancements of CTE as compared with γ-$Y_2Si_2O_7$. Such tunable CTE could presumably be explained by doping-induced localized lattice distortion around [$O_3Si—O_B—SiO_3$] pyrosilicate units, leading to switchable magnitude of negative contribution from low-frequency phonons to thermal expansion.

1. Introduction

In the future gas turbine technology, efforts to improve the engine efficiency initiate the development of advanced propulsion materials with higher temperature capability, allowing for increased gas inlet temperature [1,2]. Silicon carbide fiber reinforced ceramic matrix composites (SiC_f/SiC_m CMC) are the most promising candidate due to their low density, excellent high-temperature strength and reliability [2,3]. However, upon several thousand hours of exposure to combustion environments, SiC_f/SiC_m composites suffer from rapid recession, originated from water vapor reaction of the surface protective silica scale [4,5]. This problem could be alleviated by applying environmental barrier coating (EBC) systems, which protect the SiC components from oxidation and water vapor corrosion [6,7].

Over the years, a great variety of multilayer EBC systems have been proposed, where individual protection needs are integratedly addressed to realize improved coating performance [6–11]. Rare-earth (RE) mono- and di-silicates are important candidates due to their excellent capability in harsh combustion environments, as well as good thermochemical and thermomechanical compatibility with SiC_f/SiC_m CMC substrates [9–13]. In addition to the current advanced multilayer EBC structure (Si/Mullite/RE-silicate), an alternative promising concept is to design multilayer thermal/environmental barrier coating (T/EBC) system. Such an integrated T/EBC structure consists of a Si bond coat to promote coating adhesion and oxygen absorption, a $RE_2Si_2O_7$ EBC layer to ensure proper thermal expansion compatibility with SiC substrates, a RE_2SiO_5 EBC layer to provide good high-temperature chemical and thermal stability, and optionally an outer layer of RE-zirconate/hafnate to realize additional functions of both TBC and calcium-magnesium-aluminosilicate (CMAS) resistance.[10,11]

However, there would be huge gaps in coefficient of thermal expansion (CTE) from inner to outer coating layers. In particular, CTEs of $RE_2Si_2O_7$ EBC layer are around 4 to $5 \times 10^{-6}\,K^{-1}$, in good compatibility with SiC_f/SiC_m CMC substrates ($4.5–5.5 \times 10^{-6}\,K^{-1}$)[14]; whereas CTEs of RE_2SiO_5 (6 to $10 \times 10^{-6}\,K^{-1}$) EBC layer are much higher.[15] Significant CTE mismatch will cause huge residual stress upon coating processing and thermal cycling, and finally lead to catastrophic coating failure. For example, Richards[16] et al. has reported that gradually decreased CTEs from Yb_2SiO_5 topcoat down to SiC substrate resulted in several hundred MPa of tensile residual stress in each layer and lead to the formation of mud cracks as terrible high-diffusivity pathway for oxidation and silica volatilization. To stress this issue, engineering thermal expansions of $RE_2Si_2O_7$ inter-layer with bottom-up graded-tuned (i.e. enhanced) CTE would be of great

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significance. Such effort might enable improved thermal expansion compatibility with both upper RE_2SiO_5 layer and inner SiC_f/SiC_m CMC substrates. And hence the integrity of multilayer EBC or T/EBC is expected to be guaranteed due to optimization of thermal stresses. In forwarding this concept, understanding the mechanisms of thermal expansion behaviors for $RE_2Si_2O_7$ is in urgent necessity.

As important EBC candidates, rare-earth di-silicates exhibit interesting polymorphisms. β-, γ- and δ-$RE_2Si_2O_7$ have generally similar crystal structures,[17] yet their thermal expansion behaviors differ enormously. CTEs for β- and γ-$RE_2Si_2O_7$ range from 3.7 to $5.5 \times 10^{-6}\,K^{-1}$, whereas those for δ-$RE_2Si_2O_7$ range from 7.3 to $8.1 \times 10^{-6}\,K^{-1}$.[18–27] And hence, these three polymorphic types constitute an ideal model system for comparative studies. Becerro et al. reported that CTEs of $RE_2Si_2O_7$ are strongly dependent on polymorphic types but insensitive to RE^{3+} ionic radius [25]. Our recent work has found that CTEs of yttrium silicates depend on the anharmonicity for low-frequency phonons, in relation with the local environments and vibration flexibility of Si—O units [15]. These findings cast light on the intrinsic relationship between CTE of $RE_2Si_2O_7$ polymorphs and their crystal structures. However, questions remain as to what "gene-wise" factor and how it functions in controlling the CTE of $RE_2Si_2O_7$ polymorphs. Moreover, it is reported that for the series of RE cations, their belonged polymorphic types are distinguished by different RE-O coordination numbers, corresponding to different degree of participation of 4f-orbital in bonding [28]. Accordingly, $(RE^{I}_{x}RE^{II}_{1-x})_2Si_2O_7$ solid solution in imperfect isomorphs could be formed by doping different types of RE atoms, where the end-materials ($RE^{I}_2Si_2O_7$ and $RE^{II}_2Si_2O_7$) exhibit different polymorphic structures. Intuitively, the introduced localized lattice distortion might initiate novel structural, vibrational and thermal expansion behaviors. This concept might open a brand-new picture on optimal modification of CTE for $RE_2Si_2O_7$ derivatives and is expected to promote the proper design of advanced T/EBC systems for SiC_f/SiC_m CMC.

In this paper, lattice dynamics calculations are performed on β-, γ-, and δ-$RE_2Si_2O_7$ compounds by considering different RE^{3+} ions, i.e. RE = Y, Ho, Er, Tm, Yb, Lu for β phase; RE = Y, Ho, Er for γ phase; and RE = Y, Gd, Tb, Dy, Ho for δ phase. Phonon dispersions, Grüneisen parameters and coefficient of volumetric thermal expansion (α_V) are obtained via quasi-harmonic approximations (QHA). The gap in CTE values between β/γ and δ phase is explained in terms of differences in overall phonon anharmonicity and interatomic bonding strength. The nature of anharmonicity for low-frequency phonons is discussed with respect to specific structures of [$O_3Si—O_B—SiO_3$] pyrosilicate units and their interaction with surrounding RE atoms. This structural information is proposed as the characteristic "gene" that controls the CTE of $RE_2Si_2O_7$ compounds. To forward this understanding, γ-$(Dy_{0.15}Y_{0.85})_2Si_2O_7$ solid solution is synthesized, and thermal expansion behaviors are studied using high-temperature XRD measurements from room temperature up to 1673 K. The desired enhancement of CTE as compared with γ-$Y_2Si_2O_7$ is discussed, in regard with doping-induced localized lattice distortion around [$O_3Si—O_B—SiO_3$] pyrosilicate units.

2. Method

2.1. Theoretical calculations of thermal expansions

Based on the theory of thermodynamics, coefficient of volumetric thermal expansion (α_V) for a solid could be expressed as elastic response to thermally induced stress, as described by [29,30]:

$$\alpha_V = \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_P = \left(\frac{\partial P}{\partial T}\right)_V \cdot \left[-\frac{1}{V}\left(\frac{\partial V}{\partial P}\right)_T\right] = \left(\frac{\partial P}{\partial T}\right)_V \cdot \frac{1}{B} \tag{1}$$

where V is the unit-cell volume, T is the temperature, P is the external pressure, and B is the bulk modulus. As is shown by the two terms in Eq. (1), thermal expansion behaviors could be imagined as occurred in two steps: 1) change of thermal pressure under temperature increment at a fixed volume; and 2) change in volume as an elastic response to internal thermal pressure [30]. Furthermore, the first term could be sequenced into two factors:

$$\left(\frac{\partial P}{\partial T}\right)_V = \left(\frac{C_V}{V}\right) \cdot \gamma \tag{2}$$

Herein, C_V is the constant-volume heat capacity, and parameter $\left(\frac{C_V}{V}\right)$ determines the energy gain during a temperature increment; and γ is the thermodynamic Grüneisen parameter, which determines the effectiveness of that energy in changing thermal pressure. Combining Eqs. (1) and (2) gives the following expression, which enables one to distinguish different contributive factors to thermal expansion behaviors.

$$\alpha_V = \frac{1}{B}\gamma\left(\frac{C_V}{V}\right) \tag{3}$$

In this study, equilibrium lattices of $RE_2Si_2O_7$ polymorphs are obtained via density functional theory (DFT) calculations; and bulk modulus (B) is derived from second-order elastic constants for a static volume, following the same method as in our previous work.[31] Vibration effects are calculated within the framework of quasi-harmonic approximations (QHA), by considering the contribution from each phonon mode of a system.[29] γ value in Eq. (3) is taken as the overall Grüneisen parameter ($\bar{\gamma}$), defined as the average Grüneisen parameter for mode (j,**q**) weighed by the heat capacity:

$$C_V = \sum_{j,\mathbf{q}} C_{Vj}(\mathbf{q}) \tag{4}$$

$$\bar{\gamma} = \frac{\sum_{j,\mathbf{q}} \gamma_j(\mathbf{q}) C_{Vj}(\mathbf{q})}{\sum_{j,\mathbf{q}} C_{Vj}(\mathbf{q})} \tag{5}$$

Here, index j and **q** denote phonon branch and wave vector, respectively, which run over all points in Brillouin zone (BZ). $C_{Vj}(\mathbf{q})$ is mode-contribution to the constant-volume heat capacity of a given unit cell; and $\gamma_j(\mathbf{q})$ is mode Grüneisen parameter, calculated from the dependency of phonon frequencies in response of slight perturbation around equilibrium volume:[32]

$$C_{Vj}(\mathbf{q}) = k_B\left(\frac{\hbar\omega_j(\mathbf{q})}{k_B T}\right)^2 \frac{exp(\hbar\omega_j(\mathbf{q})/k_B T)}{[exp(\hbar\omega_j(\mathbf{q})/k_B T)-1]^2} \tag{6}$$

$$\gamma_j(\mathbf{q}) = \frac{\partial \ln\omega_j(\mathbf{q})}{\partial \ln V} \tag{7}$$

where k_B is the Boltzmann constant; $\hbar$ is the reduced Planck constant; $\omega_j(\mathbf{q})$ is the (angular) phonon frequency, derived from DFT-based phonon calculations. One thing to notice is that total heat capacity for a solid could be seen as summed contributions from phonons and electrons, and the latter part is important for metallic materials. For $RE_2Si_2O_7$ dielectric solids, the electron heat capacity could be neglected and only the phonon heat capacity is considered in our calculations.

2.2. Computational details

Density functional theory (DFT) calculations are performed using projected augmented wave (PAW) method [33] as implemented in Vienna *Ab Initio* Simulation Package (VASP) code [34]. The Perdew-Burke-Ernzerhof (PBE) [35] form of the generalized gradient approximation (GGA) is employed to treat the electronic exchange-correlation effect. The kinetic energy cutoff for the plane-wave basis is set as 800 eV in all calculations; and the Brillouin zone (BZ) is sampled on Γ-centered 5 × 3 × 7, 7 × 3×5 and 3 × 7 × 5 Monkhorst-Pack [36] k-mesh for β-, γ-, and δ-$RE_2Si_2O_7$, respectively. During structural optimization, lattice parameters and internal atomic positions are fully relaxed until the total energy and maximum ionic Hellmann-Feynman force converge to 1×10^{-10} eV and 5×10^{-4} eV/Å. As is listed in Table 1, calculated lattice parameters for all studied $RE_2Si_2O_7$

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