RE2O3 dissolution kinetics and mechanisms in CMAS silicate melts: influence of the rare-earth

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Abstract

Fine particles of sand, dust or volcanic ashes ingested by aircraft engines are well-known to damage Thermal Barrier Coatings (TBC, ZrO2-8wt. %Y2O3). In service, these particles deposit on hot TBC surface ($\geq$ 1200∘C) as molten silicate and infiltrate porous microstructure of coating. They are mainly constituted of CaO-MgO-Al2O3-SiO2 (CMAS) in variable proportions and also contain metallic oxides. Gd2Zr2O7 TBC has shown efficiency to mitigate synthetic CMAS infiltration due to its reactivity with CMAS. Indeed, the dissolution reaction leads to rapid formation of a sealing-layer in the topcoat mainly constituted of crystalline Ca2Gd8(SiO4)6O2 apatite. However, this phase is not always stable in contact with CMAS and many rare-earth silicates may compete with apatite crystallization. Several rare-earth oxides RE2O3 can be considered to replace yttria in ZrO2-based TBC but little is known on reaction kinetics and thermodynamics involving RE2O3 and multi-component CMAS system.
In this study, a simplified CMAS was selected with eutectic (1170∘C) 65SiO2-26CaO-9Al2O3 (mol. %) composition. Investigation on RE2O3 (RE = Nd, Sm, Gd, Dy and Yb) dissolution mechanism in CAS-melt was then performed at 1200∘C. For this, CAS-glass beads containing dispersed RE2O3 solid powder in adequate amount to reach oversaturation were elaborated. Beads were then annealed in air for several durations and quenched. SEM observations coupled with XRD analysis gave information about dissolution/precipitation sequences and phase equilibria. The evolution of RE content dissolved in CAS over time and chemical evolution of CAS-melt was measured by EPMA.
Reaction mechanism in CAS of RE2O3 was identified to be incongruent dissolution leading to precipitation of different RE-rich silicates. Dissolution of Nd2O3, Sm2O3, Gd2O3 and Dy2O3 in CAS results in formation of apatite, and then precipitation of cyclosilicate. Disilicate Yb2Si2O7 was also obtained in Yb2O3 bead-sample. The silicate precipitations induced significant variations of local CAS composition resulting in CaAl2Si2O8 anorthite formation. The RE2O3 basicity has a direct influence on silicates solubility limits in CAS and on reaction kinetics.

Keywords: Molten silicates, Rare earth oxides, Dissolution, Precipitation

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