

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

F. Perrudin^{1,2}, M.H. Vidal-Sétif¹, C. Petitjean², P.J. Panteix², M. Vilasi²

¹*Department of Metallic Materials and Structures (DMSM), ONERA - The French Aerospace Lab, F92322, CHATILLON cedex, FRANCE*

²*Institut Jean Lamour - Université de Lorraine (UMR 7198), Dépt CP2S, Eq SIRCM - Faculté des Sciences et Technologies, B.P 70239, VANDOEUVRE les NANCY cedex, FRANCE*

E-mail francois.perrudin@onera.fr

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 ($\geq 1200^{\circ}\text{C}$) 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.