MASS TRANSFER CONTROL IN MULTILAYER EBC SYSTEMS AT HIGH TEMPERATURES

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Environmental barrier coating (EBC) systems, which are required to have excellent environmental shielding capability and thermomechanical durability, typically have a multilayer structure that consists of complex oxides such as silicates and aluminates to achieve the required performance through the use of layers with different characteristics. Such coatings with highly dense layers exhibit excellent gas shielding performance when they are exposed to a high oxygen potential gradient ($d\mu_O$) at elevated temperatures. For example, in the case of $Yb_2Si_2O_7$ (YDS) and Yb_2SiO_5 (YMS) layers that constitute the oxide-EBC systems, the application of $d\mu_O$ results in the inward diffusion of oxide ions and outward diffusion of Yb ions along grain boundaries (GBs), according to the Gibbs-Duhem equation [1,2]. In the case of $Al_6Si_2O_{13}$ (mullite) layer, inward GB diffusion of oxide ions and outward GB diffusion of Al ions occur [3]. Cation transport induces decomposition of the complex oxides that comprise the layers, which leads to collapse of the multilayer structure. Therefore, suppression of the outward diffusion of cations under $d\mu_O$ is extremely important in the design of robust EBC systems.

In this study, an EBC design to improve both the structural stability and environmental shielding properties of multilayer structures was investigated based on mass transfer mechanisms in the individual layers. The oxygen and cation fluxes at the outflow side in single-layer EBC systems such as YMS, YDS, and mullite were significantly larger than those at the inflow side, in accordance with dominant cation transport under a high oxygen potential (μ_O) region near the surface, and dominant oxygen transport under the low-μ_O region in the vicinity of the interface between the oxide-EBC and Si-based bond coat or SiC/SiC substrate. This suggests that several different ions interdiffuse within multilayer EBC systems and can be separated into a single species according to the layer configuration by control of the GB densities and thicknesses of the layers. For instance, in the case of a double layer structure that consists of an upper YDS layer and a lower mullite layer, the transport of Yb ions and oxide ions is expected to be dominant in the YDS layer and the mullite layer, respectively.

In addition, the water vapor resistance of YMS is superior to that of YDS, and the thermal expansion coefficient of YMS is larger than those of SiC/SiC and YDS. Therefore, a YMS layer is typically arranged on a YDS layer for a two-layer structure. The chemical potential of Yb in YMS at 1400 °C is about 30 kJ/mol larger than that in YDS; therefore, the interface between the YDS and YMS layers acts as an energy barrier for the outward GB diffusion of Yb ions at this temperature [4]. The oxygen permeability of a two-layer structure, in which a dense YMS film was coated on a YDS wafer by electron-beam physical vapor deposition (PVD), was evaluated under $d\mu_O$, where Yb ions are predominantly transported via GBs. The results indicated that the bilayer structure exhibited excellent oxygen shielding properties, even though the oxygen permeability constant normalized by the GB density for the YMS single film was clearly inferior to that for the YDS single film. Moreover, YMS is thermodynamically more stable than YDS in a high-temperature water vapor environment. Hence, even if no YMS film is intentionally deposited, the P_{O2}(hi) surface of the YDS single wafer can be thermodynamically converted to YMS only by humidification. Accordingly, oxygen shielding and permeation reversibly proceeds for a YDS single wafer in response to humidified and dry environments on the P_{O2}(hi) surface [4].

The structural stability and oxygen shielding properties of a common three-layer EBC system consisting of a YMS top layer, a YDS intermediate layer, and a mullite bottom layer are thus expected to be significantly improved by unifying the main diffusion species in each layer and simultaneously utilizing an energy barrier against the outward diffusion of cations. Finally, an effect of mass transfer in EBCs on corrosion of them due to CMAS-attack will be also discussed.

[1] M. Wada et el, Acta Materialia, 135, 372-381 (2017).
[2] T. Matsudaira et el., J. Eur. Ceram. Soc., 41, 3150-3160 (2021)
[3] S. Kitaoka et el., Coatings, 9, 630-639 (2019)
[4] M. Wada et el., Acta Materialia, 201, 373-385 (2020