Coatings for Use at High Temperatures
Room Pacific E - Session A2-2-WeM

Thermal and Environmental Barrier Coatings II

Moderators: Dr. Vladislav Kolarik, Fraunhofer Institute for Chemical Technology ICT, Germany, **Dr. Pantcho Stoyanov**, Concordia University, Canada

8:00am **A2-2-WeM-1 On the Suitability of MoNbTaW Based Thin Films to Act as Diffusion Barriers***, **Georg C. Gruber**[1],* Montanuniversität Leoben, Austria*; A. Lassnig, S. Zak,* Austrian Academy of Sciences, Austria*; M. Kirchmair,* Montanuniversität Leoben, Austria*; S. Wurster, C. Gammer, M. Cordill,* Austrian Academy of Sciences, Austria*; R. Franz,* Montanuniversität Leoben, Austria

To further improve the performance of diffusion barriers between Cu and Si, several new approaches were developed, such as the use of high entropy alloys (HEAs). HEAs are alloys with a configurational entropy of at least 1.5R, where R is the gas constant. On the one hand, HEA thin films can be deposited with amorphous microstructure which can be beneficial for their use as diffusion barriers. On the other hand, the lattice distortion within HEAs leads to an increase of the strain energy as well as the cohesion energy leading to an increased activation energy for the Cu diffusion. Within the current study, six different HEAs, based on the MoNbTaW system and alloyed with Ti, V, Cr, Mn, Zr or Hf, have been deposited by high power impulse magnetron sputtering (HiPIMS) as diffusion barrier layers between Cu and Si. The thickness of the HEA layers on the Si substrate was 20 nm, followed by a 150 nm thick Cu layer also deposited by HiPIMS. X-ray diffraction (XRD) investigations found that the MoNbTaW layers alloyed with Ti and V showed a body-centered cubic microstructure while the other alloys showed an amorphous microstructure. Subsequently, the bi-layers were annealed at temperatures ranging from 550 to 800 °C in a vacuum furnace. After annealing, the bi-layers were investigated by XRD, resistance measurement and laser-scanning-microscopy, to check for potential barrier failure. The lowest onset temperature for barrier failure of 600 °C was found for amorphous MoNbTaWZr, whereas the highest of 700 °C was observed for CrMoNbTaW. The obtained results are discussed in terms of strain and cohesive energy as well as the enthalpy of mixing between the fifth alloying element (Ti, V, Cr, Mn, Zr or Hf) and Cu. Within the study, the great potential of these HEAs for future diffusion barriers was demonstrated.

8:20am **A2-2-WeM-2 Improvement of EBC Performance by Controlling Driving Forces for Mass Transfers in Oxides***, **Satoshi Kitaoka**,* JFCC, Japan*; T. Matsudaira, T. Ogawa, M. Wada,* Japan Fine Ceramics Center, Japan **INVITED**

Environmental barrier coating (EBC) systems 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 (dμO) at elevated temperatures.

In the case of Yb2Si2O7 (YDS) and Yb2SiO5 (YMS) layers that constitute the oxide-EBC systems, the application of dμO results in the inward diffusion of oxide ions and outward diffusion of Yb ions along grain boundaries (GBs), according to the Gibbs-Duhem equation. In the case of Al6Si2O13 (mullite) layer, inward GB diffusion of oxide ions and outward GB diffusion of Al ions occur. Cation transport induces decomposition of the complex oxides that comprise the layers, which leads to collapse of the multilayer structure. Hence, suppression of the outward diffusion of cations under dμO 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 EBCs 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 (μO) region near the surface, and dominant oxygen transport under the low-μO 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. In addition, the interface between the YDS and YMS layers acts as an energy barrier for the outward GB diffusion of Yb ions. Therefore, the structural stability and oxygen shielding properties even for an common three-layer EBC system consisting of a YMS top layer, a YDS intermediate layer, and a mullite bottom layer are considered 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.

Key Words: Diffusion, Grain boundary, EBC, Oxygen permeability, High temperature

9:00am **A2-2-WeM-4 Steam Oxidation Kinetics of Si / Modified $Yb_2Si_2O_7$ Environmental Barrier Coatings on SiC/SiC Ceramic Matrix Composites at 1250 °C – 1350 °C***, **Kang Lee**, J. Stuckner, M. Presby, B. Pulio,* NASA Glenn Research Center, USA*; W. Jennings,* HX5, USA

Environmental barrier coatings (EBCs) have enabled the implementation of SiC/SiC ceramic matrix composites (CMCs) in gas turbines by protecting CMCs from H_2O-induced volatilization. Improving the reliability of CMC components requires long-life EBCs and accurate EBC lifing. Steam oxidation-induced failure is the most frequently observed EBC failure mode. NASA previously reported that modifying Si / $Yb_2Si_2O_7$ EBC by adding Al_2O_3 or Al_2O_3-containing compound, such as mullite ($3Al_2O_3{\cdot}2SiO_2$) and YAG ($Y_3Al_5O_{12}$), in $Yb_2Si_2O_7$ reduces the EBC parabolic oxidation rates by up to about 20 times at 1316°C in steam. Oxidation is a thermally activated process and therefore the oxidation rates vs. temperature relationship typically follows the Arrhenius equation. This paper reports the results of follow-on studies that expanded the test temperature to 1250 °C and 1350 °C to understand the temperature dependency of oxidation rates, which is a key component for EBC lifing.

9:20am **A2-2-WeM-5 Oxygen Permeability, Failure Analysis and Life Prediction of Environmental Barrier Coatings Under Adverse Environments***, **Prakash Patnaik**,* Aerospace Research Centre, National Research Council Canada*; A. Kumar,* TECSIS Corporation, Canada*; K. Chen,* Aerospace Research Centre, National Research Council Canada

Environmental barrier coatings (EBCs) are typically used to protect ceramic matrix composites (CMCs) substrate against a harsh environmental attack such as high-temperature water vapour-induced recession in aero-engines achieving lower density, higher temperature tolerance capability and higher engine thrust-to-weight ratio. Under adverse service operations, the oxygen permeability of ytterbium disilicate (YbDS) topcoat and thermally grown oxide (TGO) silicon dioxide in EBCs plays a key role in determining EBCs durability and life span. Using physics-based model and thermodynamics calculations along with defect reaction formulae, oxygen permeabilities under dry oxygen and water vapour conditions, as well as different temperatures, partial pressures and top coat modifiers, are investigated. The results show that oxygen permeability for topcoat YbDS is an order of magnitude higher than for TGO, indicating that TGO hinders the oxidant diffusion stronger, proving to be the diffusion rate controlling layer. Moreover, water vapour strongly increases the oxidant permeation, with the defect reaction playing an important role. It is suggested that the mass transfer through the topcoat is primarily by outward ytterbium ion diffusion and inward oxygen ion movement, with the latter being dominant, particularly in water vapour environments.

The heterogeneous multi-layers designed for EBCs offer specific functions. High thermal strain and stress could develop due to materials mismatch and thermal gradient, leading to an accelerated degradation limiting EBCs durability and performance. Multi-physics/mechanics approaches were used to establish thermal and residual stress models for multi-layer EBCs considering the theory of composite elastic beam bending due to thermal gradient under force–moment equilibrium and stress-strain-curvature relationship. Residual stress distributions along the layer thickness were obtained for selected EBCs. Results are then compared with the test data, and the model exhibits a consistent variation in stress values across different coating layers. Specifically, thermal and residual stresses were evaluated within topcoats YbDS and ytterbium mono silicates (YbMS). Higher stress in YbMS makes it unsuitable as the topcoat despite its thermochemical compatibility between components and very low steam volatility. Finally, the lifetime prediction was explored for selected EBCs under specific water vapor contents and thermal cycle conditions based on the developed stress models.

[1] **Graduate Student Award Finalist**

9:40am **A2-2-WeM-6 Raman Spectroscopic Investigation of SiO_2 TGO Phase Transformation and Si and SiC Substrate Stress**, ***Michael J. Lance,*** Oak Ridge Natinal Laboratory, USA; *M. Ridley, T. Aguirre, B. Pint,* Oak Ridge National Laboratory, USA

SiC ceramic matrix composites (CMCs) are desired for use in combustion environments to achieve higher turbine operating temperatures, although CMCs require environmental barrier coatings (EBCs) for protection from the gas environment. EBC systems are known to primarily fail through coating delamination via growth of a thermally grown oxide (TGO) at the EBC – silicon bond coat interface. The TGO undergoes a phase transformation during thermal cycling, which results in stresses that may encourage EBC spallation. Uncoated Si and SiC substrates were exposed to air and steam to form cristobalite TGOs of a thickness that remained intact upon cooling to room temperature. These TGOs were then thermally cycled across the phase transformation temperature for cristobalite and characterized with Raman microscopy to map the α↔β transformation. The stress in the silicon and SiC was simultaneously measured with Raman microscopy. Finite element modeling was used to predict the CTE mismatch stress and transformation stress in the TGO which was compared to the measured Raman stress. This research was funded by the Advanced Turbine Program, Office of Fossil Energy and Carbon Management, U.S. Department of Energy.

11:00am **A2-2-WeM-10 Hot Section Coating Technology as an Enabler for Sustainable Propulsion**, ***Eli Ross,*** Pratt & Whitney, USA **INVITED**

Continued growth within the aviation sector is often seen as running counter to the shared objective of carbon-neutral flight. That growth trend brings not only higher demand and corresponding emissions, it also brings exposure to more aggressive and challenging operating environments for gas turbine engines. Accordingly, sustainable propulsion systems must build on recent learning from harsh environmental exposure while provisioning for a future state of more efficient, lower carbon engine technology sets encompassing a suite of higher thermal efficiency, hybrid-electric, and alternate fuel solutions. A common thread across this complex future turbine engine landscape is the need for continued improvement and development of thermal and environmental barrier coatings used in the hottest areas of the engine to enable both performance and durability. An overview and historical perspective of Pratt & Whitney work in this area will be presented, along with a system-level vision for the continued role of coating development in helping to deliver on a more sustainable net-zero future.

11:40am **A2-2-WeM-12 Development of Tantalum Coating by the Cold Spray**, ***Sheng-Wei Zeng,*** Department of Material and Mineral Resources Engineering, National Taipei University of Technology, Taipei, Taiwan; *Y. Chung, W. Li,* National Chung Shan Institute of Science and Technology, Materials and Electro-Optics Research Division, Long-tan, Taiwan; *Y. Yang,* Department of Material and Mineral Resources Engineering, National Taipei University of Technology, Taipei, Taiwan

In the coating technology, cold spray(CS) is different from the traditional thermal spraying technology. The cold spray coating is formed by plastic deformation without high temperature melting, which can keep the original characteristics of the material during the spraying process and have a denser coating. Refractory metals, tantalum(Ta), has good performance at high temperature, used in the electronics industry, aviation, defense and medical treatment. This study used cold spray to prepare refractory metals, tantalum(Ta) coating, discusses the cold spray under different process parameters (chamber pressure, chamber temperature, working distance) affected the microstructure changes of the coating and its mechanical properties, such as porosity, hardness, tensile strength, etc.

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