Title: Improving Jet Engine Turbine Thermal Barrier Coatings via Reactive Element Addition to the Bond Coat Alloy
Authors: Emily A. Jarvis and Emily A. Carter
Contact: University of California, Los Angeles, Department of Chemistry and Biochemistry, Box 951569, Los Angeles, CA 90095-1569
E-mail: E. Carter (eac@chem.ucla.edu) and E. Jarvis (asche@chem.ucla.edu)
URL: www.chem.ucla.edu/carter/
MHPCC Resources: IBM SP3
Other resources: DEC Alpha ES40s

Introduction:
Thin ceramic coatings on jet engine turbine blades can shield the temperature to which the underlying superalloy is exposed by hundreds of degrees Celsius. The higher temperature operation afforded by the ceramic top coat of the thermal barrier coating allows the engine to operate with greater power and fuel efficiency. After repeated thermal cycling, the ceramic film spalls, exposing the superalloy to the harsh temperature and the oxidative/corrosive gases of the combustion chamber. Previously, we characterized ideal interfaces within the thermal barrier coating. Improved understanding of the atomic-level interactions at heterogeneous interfaces gained in these studies allowed us to extend our research into largely uncharted territory over this past year. Specifically, we are attempting to serve in a predictive role by suggesting chemical modifications that should inhibit the failure of these jet engine turbine thermal barrier coatings.

Research Objective:
Our research aim is twofold. First, we characterized ideal interfaces in a typical thermal barrier coating to investigate fundamental interactions at heterogeneous interfaces and atomic-level culprits in materials failure. In particular, we explored the interactions occurring at ceramic/Ni interfaces since nickel is the primary component of the turbine blade metal alloys. Armed with understanding gained through our ceramic/Ni studies, the second phase of our research is to search for means of limiting or eliminating thermal barrier coating failure. Our focus thus far has been on suggesting chemical means by which interface adhesion could be enhanced by forming stronger covalent and ionic bonding interactions across the metal-ceramic interface.

Methodology:
We perform planewave, pseudopotential density functional calculations, employing the generalized gradient approximation (PW91) to the exchange-correlation potential, using the Vienna Ab Initio Simulation Package. The 3D periodic boundary conditions allow simulation of effective bulk crystals, surfaces, and interfaces neglecting long-range relaxation effects. Interface adhesion energies are calculated by subtracting the total energy of the combined substrate and coating from the sum of the energies of isolated

substrate and coating slab calculations and normalizing this value by the area of the interface periodic cell. For several metal/ceramic interfaces, we also performed high temperature annealing and quenching to simulate the harsh temperature environment of the jet engine and ascertain that the relaxed ionic coordinates were not trapped in an unfavorable local minimum energy configuration.

**Results/Significance:**
A significant source of thermal barrier coating failure is related to the Al$_2$O$_3$ scale that grows via bond coat alloy oxidation during thermal cycling of the jet engine. Our calculations of the clean Al$_2$O$_3$/Ni interface showed that the interfacial bonding is very weak for thick Al$_2$O$_3$ films. Our calculations indicate that introducing early transition metals to the bond coat alloy results in significantly increased interface adhesion at these metal-ceramic interfaces. These elements have open $d$-shells; thus, they are able to form stronger bonds to the closed-shell ions of the highly ionic Al$_2$O$_3$ than the late transition metals, which have a mostly filled valency leading to closed-shell repulsions.

Furthermore, our investigation of the interface bonding behavior of nickel with SiO$_2$, an oxide with a strong covalent component to its bonding, displays that this interface does not suffer from the unfavorable adhesion properties exhibited by Al$_2$O$_3$/Ni. It is hoped that modifying the composition of the bond coat alloy to include certain early transition metals will provide a straightforward means to improve operational lifetimes of thermal barrier coatings. Likewise, our calculations of the SiO$_2$/Ni interface imply that ultimately it may be preferable to alter the bond coat chemistry such that its oxidation product has a stronger covalent character than Al$_2$O$_3$.

Figure Caption: This figure shows the valence electron density localization of a cross-sectional view of the SiO$_2$/Ni interface. Red is zero and purple indicates the highest localization. The violet arrow indicates the region of localized bonding between the SiO$_2$ and the nickel substrate. The other blue peaks correspond to the oxygen ion lone pairs and the nickel ions are located at the rows of lime-colored bumps.