UNDERSTANDING SILICATE DEPOSIT VARIABILITY AND ITS IMPLICATIONS FOR EVALUATING TBCs AND EBCs

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CMAS degradation of thermal and environmental barrier coatings (T/EBCs) is recognized as a fundamental barrier to progress in gas turbine technology; melting of the precursor silicate deposits, typically at ~1200°C, limits the temperature capability of the coatings and by extension the achievable engine efficiency. At its core, coating damage is essentially thermomechanical, driven either by stiffening of the strain-tolerant segmented architecture of TBCs or by conversion of the dense and nominally CTE matched EBCs into reaction products that are thermally mismatched with the CMC substrate. In that context, thermochemical interactions are crucial to the microstructural changes that eventually lead to the loss of strain tolerance in most coatings, but could be used to advantage in developing mitigation strategies to arrest melt penetration in TBCs.

A challenge to understanding the breath of thermochemical interactions between the thermal and/or environmental barrier oxides with silicate melts is the stochasticity of the mineral deposits from which the melts originate. It is acknowledged that these deposits can vary widely in composition but most studies in the literature are based on one or at most a few compositions, usually simplified to the main oxide components. Nevertheless, it has become evident in several studies that the response of a given oxide to silicate melts of different compositions, notably their Ca:Si ratio, can be substantially different with attendant implications for the damage mechanisms. There is, however, a need for a systematic approach to select silicate compositions representative of the breadth of behaviors to evaluate coating candidate compositions.

This presentation will describe the development of a protocol to identify exemplary silicate compositions for coating evaluation from a large database including a wide range of engine deposits and potential sources of mineral debris. A database comprising about 180 CMAS-relevant composition was compiled from literature sources, including engine deposits, naturally occurring siliceous minerals like volcanic ashes, sands and dusts, as well as synthetic compositions used in research or testing studies. These were reduced to 90 compositions by eliminating all the synthetic compositions as well as obvious duplicates from mineral debris from a single origin, e.g. a volcanic eruption or a specific desert. A subsequent distillation included only the major oxides, i.e. those of Ca, Mg, Fe, Al and Si, adding to 90% or more of the specific debris or deposit. The database was then organized into classes with similar chemical characteristics, independent of mineral origin, using principal component analysis (PCA) and k-means clustering. Four distinct classes were identified, each with a centroid and a range in Ca:Si as well as in the sum of the relatively minor oxides (Mg, Fe, Al). Liquidus and solidus temperature, as well as the composition of the equilibrium melts at relevant temperatures, were calculated using Thermocalc and the TCOX10 thermodynamic database. The melt compositions were then used to estimate properties critical to melt penetration and thermomechanical response, notably viscosity, glass transition temperature, thermal expansion and basicity from reasonably established models in the literature. The reaction paths between prototypical coating oxides and the centroids and other compositions of interest within the individual classes were calculated, reflecting the range of scenarios that may evolve from these interactions. Exemplary compositions for testing may then be selected from consideration of properties and reaction paths, as discussed in the presentation.

This work is not intended to identify specific compositions for standardize CMAS testing by the community, although that could certainly be one outcome, but rather to provide a structure and statistical tools that can be applicable to similar compilations of deposit compositions. Ideally this would be continued through collaborations with industry, expanding the database of engine deposits to ensure broader geographical and engine technology coverage.