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Environmental/Thermal Barrier Coatings for Ceramic Matrix Composites: Thermal Tradeoff Studies

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Summary

Recent interest in environmental/thermal barrier coatings (EBC/TBCs) has prompted research to develop life-prediction methodologies for the coating systems of advanced high-temperature ceramic matrix composites (CMCs). Heat-transfer analysis of EBC/TBCs for CMCs is an essential part of the effort. It helps establish the resulting thermal profile through the thickness of the CMC that is protected by the EBC/TBC system. This report documents the results of a one-dimensional analysis of an advanced high-temperature CMC system protected with an EBC/TBC system. The one-dimensional analysis was used for tradeoff studies involving parametric variation of the conductivity; the thickness of the EBC/TBCs, bond coat, and CMC substrate; and the cooling requirements. The insight gained from the results will be used to configure a viable EBC/TBC system for CMC liners that meet the desired hot surface, cold surface, and substrate temperature requirements.

Background

The development of environmental/thermal barrier coating (EBC/TBC) systems is being pursued under the NASA Ultra-Efficient Engine Technology (UEET) project. The primary objective is to enable the use of ceramic matrix composites (CMCs) in propulsion system components that are subjected to extremely harsh thermal environments. Turbine vanes and combustor liners are examples of such components that need protection from hot combustor gases as well as environmental effects due to moisture, debris, oxygen, and other substances (refs. 1 and 2). A novel concept is under development at the NASA Glenn Research Center for protecting materials against these environmental effects. EBC/TBC material systems developed previously under the High-Speed Civil Transport and Enabling Propulsion Materials programs can perform at material surface temperatures up to 1315 °C. New EBC/TBCs being developed under the NASA UEET project are expected to deliver environmental and thermal protection with assured reliability and durability under very high operating gas temperatures. These coating systems are expected to see surface temperatures around 1480 °C. The bond-coat/substrate interface temperature is required to survive temperatures at least as high as 1315 °C with assured reliability. A pictorial representation of the current state-of-the-art EBC is shown in figure 1. It consists of a CMC substrate, a silicon layer bond coat, a composite layer of mullite and BSAS (barium strontium aluminum silicate), and a pure BSAS layer as the EBC on the top.

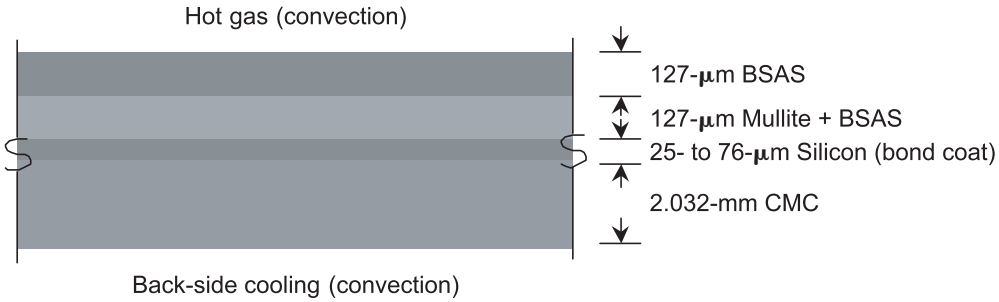

Figure 1.—Environmental barrier coating system under development at Glenn.

Introduction

With the ever-increasing need for EBC/TBC systems in more critical high-temperature applications, the life and durability of coatings has become an important issue. Experimental investigations of EBC/TBC systems often lead to an understanding of the underlying failure mechanisms. However, extensive experimental testing is needed to arrive at an optimum configuration for the system because of the many design parameters involved. Consequently, analytical modeling of EBC/TBC life and durability is required to minimize the number of experimental investigations needed to arrive at an optimum EBC/TBC system that satisfies all the requirements. Such efforts eventually lead to the development of computational design tools that can be used for the optimum design of coating systems for combustor liners. A brief survey of the relevant literature was reported recently (ref. 3). For completeness, results from the literature are summarized in the following paragraphs. Note that, since the EBC is a novel material development concept originated in-house at Glenn, research in the life prediction and cracking characteristics of EBCs is very limited, and the authors are not aware of any literature available on EBCs for CMCs outside of Glenn sources. Therefore, this section focuses primarily on Glenn coating material development design issues. Reference 4 summarizes various researchers' hypotheses regarding the cracking behavior and the attendant failure modes in TBCs that are of significant importance to the life and durability of the coatings as well as the coated components.

The temperature differences through the thickness of the EBC/TBC and the CMC, together with the mismatch in their expansion coefficients and the stiffnesses of the various layers, could lead to tensile stresses in some layers. The tensile stresses in the layers can cause cracks in the coating system and, thereby, expose the substrate to harsh environments. A number of research efforts are focusing on analytical and experimental studies of coating systems applied to metallic substrates (refs. 5 to 9). With the aid of conventional finite element analysis methods, Cheng et al. (ref. 5) quantified the residual stresses in an electron beam physical-vapor-deposited yttria-stabilized zirconia TBC system on a platinum-aluminum bond coat. An elastoplastic analysis of a circular disk specimen modeled with an actual interface surface with ridges and cavities showed significant areas of tensile stresses responsible for cracking.

Brindley (ref. 6) conducted experiments to observe the effect of increasing the NiCrAlY bond coat oxidation resistance on TBC life. He showed that besides the oxidation being a main driver, there are other issues that have a pronounced effect on life. Differences in the coefficients of thermal expansion of the various layers and the stress relaxation of the alloy in the bond coat applied on a metallic substrate affect the coating life significantly. Out-of-plane residual stresses in the bond coat increase during the stress relaxation phase of the plasma-sprayed coatings. These out-of-plane stresses result in material creep and delaminate the ceramic layer, decreasing the life.

DeMasi et al. (ref. 7) performed an extensive experimental investigation of a two-layer NiCrCoAlY TBC system in order to study the inherent failure mechanisms and develop coating life models. Their life model used a one-dimensional constitutive relationship and model developed by Walker (ref. 10) and an oxide growth rate that was verified with the experimental data.

Kokini et al. (refs. 8 and 9) studied the coating properties and behavior of CoCrAlY under high thermal gradients and thermal cyclic loads. Their initial study focused on the effects of surface temperature as well as thermal gradients on fracture mechanisms in TBCs. Subsequently (ref. 9), they studied the effects of laser heating, manufacturing processes, and coating thickness on the cracking behavior of TBCs. These studies indicated that the cooling-induced tensile residual stresses led to surface cracking. The studies are primarily experimental in nature, and comprehensive analytical modeling was not done to explain the experimentally observed behavior.

Thermal residual stresses, in addition to causing coatings to crack, sometimes cause the coating system to delaminate from the substrate. References 6, 11, and 12 focus on delamination-related failure mechanisms in the coating systems. Andritschky et al. (ref. 11) studied the mechanics of delamination on metal-based thick ceramic coatings. Delaminations initiated at the transverse crack sites caused by the sintering process, thermal stresses, and internal oxidation during prolonged exposure to high temperatures were experimentally and computationally investigated. Finite element method simulations were used in combination with analytical classical fracture mechanics to compute stress intensity factors. Andritschky et al.'s report concluded with the remark that finite element method analysis of the entire coating system can lead to better results because of the complexity of residual stresses, short cracks, multiple cracking, and other parameters.

Teixeira et al. (ref. 12) quantified the residual stresses induced by the processing and coefficient of thermal expansion mismatch experimentally and numerically in plasma-sprayed TBCs. Good agreement between the computed residual stresses and the experimental data was noted. Also, studies under isothermal and cyclic heat treatment were conducted. It was observed that residual stresses that developed in the coating led to the adhesive (delamination of the interface) or cohesive (microcracking or spalling within the ceramic coating) failure because of the coefficient of thermal expansion mismatch, thermal gradients, and the thermal history. In addition, the presence of free edges led to high interfacial shear and axial stresses, which promote microcracking parallel and adjacent to the interface. Also, during rapid thermal cycling, compressive stresses developed in the coating and caused cracks to form at the interface.

Cracks often originate at multiple sites in the coating system and, therefore, should be treated as such as opposed to analyzing a single crack with linear fracture-mechanics-based approaches, which have been quite commonplace in literature. Reference 13 addresses multiple cracking phenomena in functionally graded materials. Multiple cracking is the most common phenomena observed in both TBC and EBC coating systems. Nusier et al. (ref. 14) performed an experimental and analytical investigation of the damage process in TBCs subjected to different thermal cycle profiles.

Erosion due to foreign object impact, though not directly related to a thermal-gradient-induced failure mechanism, is often observed in conjunction with coating systems. Wellman and Nicholls (ref. 15) performed formal studies to explain the erosion process mechanism linked with the microstructure of the coatings.

As indicated by the literature, most of the work has focused on TBC systems on conventional metallic substrate material. Modeling and analysis of EBC/TBC systems on CMCs has been very limited. In order to design a viable coating system that meets the necessary survival requirements, it is highly desirable to establish thermal gradients through various layers in addition to computing the thermally induced stresses. These tasks can be achieved via heat-transfer and stress analyses. The present effort consists of the first part, namely heat-transfer analysis as well as a set of tradeoff studies. The coating system that is considered in the following sections is the one proposed and pioneered by Lee (refs. 1 and 2). Currently, this system is being investigated experimentally to evaluate its survivability and durability under harsh combustion environments. As mentioned earlier, it consists of a CMC as the substrate, a silicon layer as the bond coat, a composite layer of mullite and BSAS, and pure BSAS layer as the EBC at the top. The coating system must survive a surface temperature of 1480 °C and a bond-coat/substrate interface temperature of at least 1315 °C with assured reliability.

Heat-Transfer Analysis

In the present effort, the primary purpose of heat-transfer analysis is to predict and establish the thermal profile in the EBC system and the CMC with reasonable accuracy. This is important because it is the first step in assessing the durability of the coating system. As shown in figure 1, the system consists of four layers of different materials. In general only the temperatures at the top and bottom surfaces can be measured experimentally. The temperatures within the layers have to be established by some analytical means. A judicial combination of both methods will lead to an experimentally validated analytical procedure for a parametric analysis showing the effect of variations in the different design parameters on the thermal profile.

The one-dimensional linear heat-transfer analysis used accounts for hot gas convection; conduction in the EBC, mullite, bond coat, and CMC layers; and convection due to impingement cooling. Radiation effects are not explicitly considered in the present analysis. For the tradeoff studies, the amount of heat load on the hot side based on an equivalent film coefficient is deemed sufficient. Similarly on the cold side, the calculated heat loads are based on an equivalent convection cooling that represents typically an impingement-type cooling. For steady-state heat transfer, the heat flow through the hot gas, the EBC layers, and the cooling gas should remain constant. By applying this condition, equations to compute temperatures in different layers can be derived as shown in the following equations. Figure 2 and the following definitions should be referred to for the notations used in the equations:

h_1, h_2	film coefficients on the hot (EBC) and cold (CMC) sides
t_1, t_2, t_3, t_4	EBC, mullite, bond coat, and CMC layer thicknesses, respectively
k_1, k_2, k_3, k_4	EBC, mullite, bond coat, and CMC layer thermal conductivities, respectively
T_1, T_2, T_3, T_4, T_5, T_6, T_7	hot gas, EBC top, mullite top, bond coat top, CMC top, CMC bottom, and cold-side gas temperatures, respectively.
Q	total heat flow
A	unit surface area

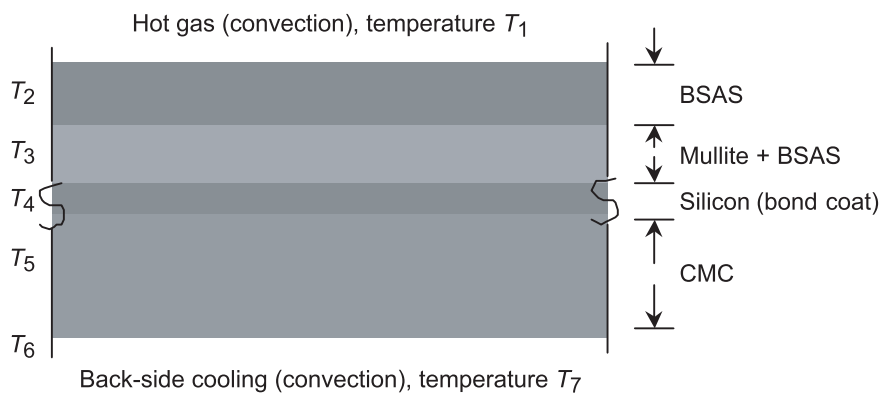

Figure 2.—Modeling for one-dimensional heat-transfer analysis.

The equation for the steady-state linear convection heat transfer within the hot gases over EBC can be written as (ref. 16)

$$Q = h_1 A(T_1 - T_2) \tag{1}$$

The one-dimensional heat-conduction equation according to Fourier's law is given by reference 16:

$$Q = kA\partial T/\partial x \tag{2}$$

Equation (2) states that the total heat flow Q in direction x, which is the thickness in the EBC system, is proportional to the gradient of the temperature in direction x. This equation can be applied successively to each of the BSAS, mullite, bond-coat, and CMC layers to arrive at the following governing equations:

EBC (BSAS) layer:

$$Q = k_1 A(T_2 - T_3)/t_1 \tag{3}$$

Mullite layer:

$$Q = k_2 A(T_3 - T_4)/t_2 \tag{4}$$

Bond-coat layer:

$$Q = k_3 A(T_4 - T_5)/t_3 \tag{5}$$

CMC substrate:

$$Q = k_4 A(T_5 - T_6)/t_4 \tag{6}$$

For conduction through the EBC and CMC, the following equation can be derived:

$$Q = \frac{(T_2 - T_6)A}{\frac{t_1}{k_1} + \frac{t_2}{k_2} + \frac{t_3}{k_3} + \frac{t_4}{k_4}} \tag{7}$$

The steady-state linear convection transfer through the back-side cooling gases is given by

$$Q = h_2 A(T_6 - T_7) \tag{8}$$

As mentioned earlier, the hot-side heat load due to the radiation and convection effects was combined with the equivalent film coefficient h_1.

Parametric Tradeoff Studies

An initial evaluation of the thermal profile computed using the design parameter values as received, figure 1 and table I, indicated that the temperature difference between the EBC top surface and the CMC top surface was less than 37.8 °C. This is much less than the UEET design requirement of 165 °C, as mentioned earlier. We, therefore, decided to conduct a parametric study by varying design parameters such as the conductivities and thicknesses of the EBC, mullite, CMC layers, and film coefficients on the

TABLE I.—EBC SYSTEM BASELINE CONFIGURATION

Design parameter	Substrate	Bond coat	Mullite + BSAS	EBC (BSAS)
Layer thickness, mm	2.032	0.0508	0.127	0.127
Thermal conductivity, W/m-K	22.000	22.1000	1.950	2.200
Film coefficient, W/m^2-K	604.000	----------	--------	458.000

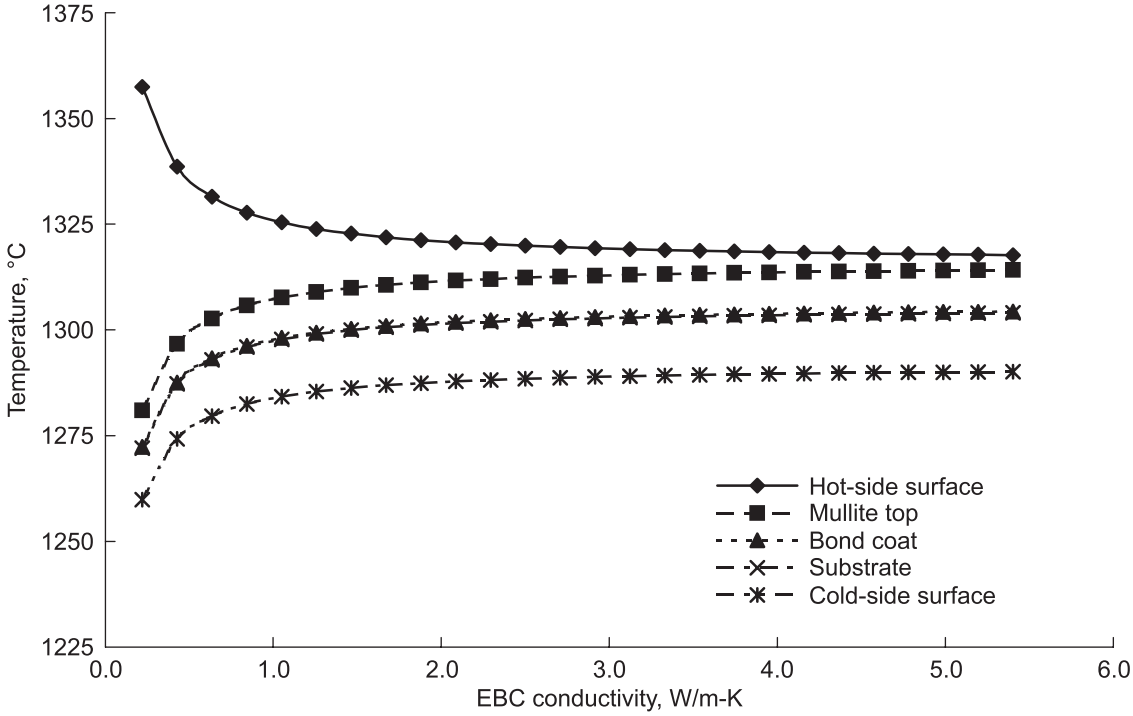

Figure 3.—Effect of variation in environmental barrier coating (EBC) conductivity.

hot- and cold-side surfaces. The EBC system configuration shown in figure 1 and table I was used as a baseline configuration. Design parameters were then varied one at a time, and temperature differences between the EBC and CMC top surfaces were computed for each variation. The design parameters were varied until the desired UEET requirement was met. The results of the analysis with gas temperatures of 1649 °C on the hot-side gas and 1038 °C on the cold side are shown in figures 3 to 9.

The effects on the overall thermal profile of varying the EBC (BSAS) and the mullite layer conductivities from 0.2 to about 6.3 W/m-K are shown in figures 3 and 4. It can be seen from figure 3 that, within the range of conductivities, 0.2 to 2.0, the increase in the thermal conductivity of the EBC has a significant effect on the thermal profile. The heat conduction, however, is also constrained by the conductivity of the mullite, bond coat, and CMC layers. Consequently, a further increase in EBC conductivity shows only a minimal effect on the thermal profile. Therefore, increases in EBC conductivity decrease the EBC layer temperature to a limit, but thereafter have no effect. The maximum temperature difference from figure 3 is about 90 °C, which is far below the UEET objective. Since the mullite conductivity and its thickness are close to those of the EBC, the variation in its conductivity also shows effects similar to those of the EBC conductivity, as shown in figure 4.

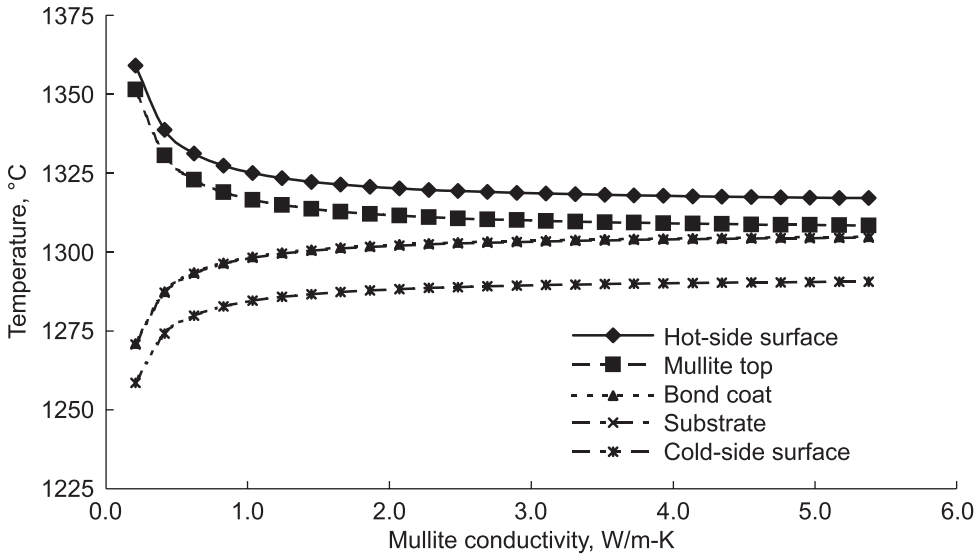

Figure 4.—Effect of variation in mullite conductivity.

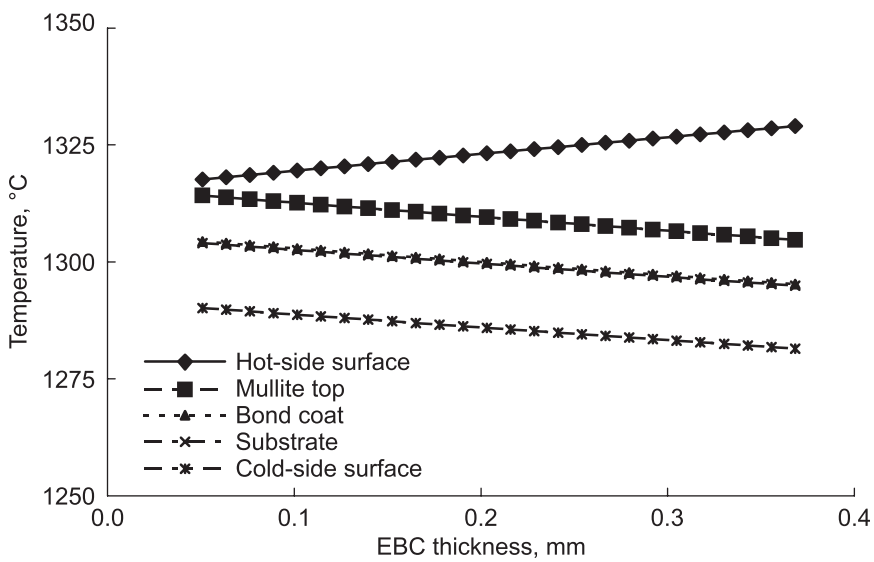

Figure 5.—Effect of variation in environmental barrier coating (EBC) thickness.

For the next set of tradeoff studies, geometry-related design variables were varied. Figures 5 to 6 show the effects of EBC and mullite thickness on the overall thermal profile. The thicknesses were varied between 0.063 and about 0.38 mm. The behavior basically follows the conduction law that an increase in thickness increases temperature differences for the same amount of heat flow. From this fact, it can be concluded that a desired temperature difference can be achieved by simply increasing the thickness of the EBC or mullite. However, the maximum temperature difference achieved between the EBC top and CMC top surfaces, within the range of EBC and mullite thicknesses analyzed, was only 40 °C, which is much lower than that observed from the EBC and mullite conductivity variation. Thus, a very thick coating system would be needed to achieve a higher temperature difference, which is not practical from a manufacturing viewpoint. In addition, there may be other design constraints as well as manufacturability issues that might limit the available thicknesses to a narrow window. Another geometry-related design

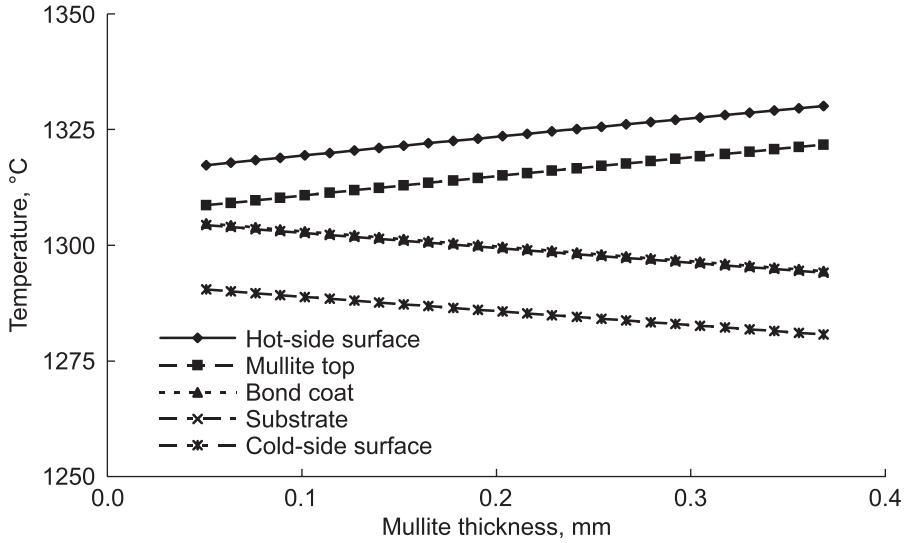

Figure 6.—Effect of variation in mullite thickness.

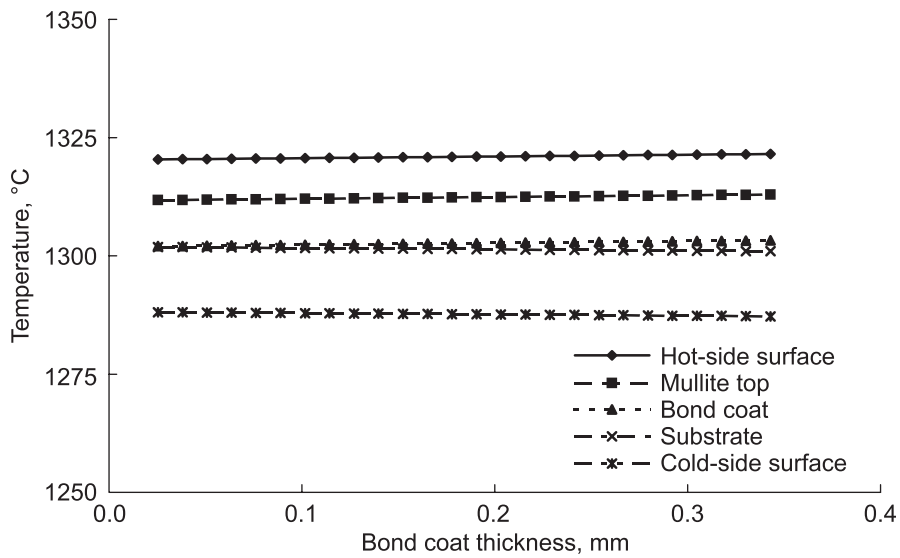

Figure 7.—Effect of variation in bond-coat thickness.

parameter is the bond-coat thickness, whose effect on the temperature profile is shown in figure 7. As seen in the figure, the bond-coat thickness has a negligible effect on the temperatures in the EBC system.

The last set of design variables considered in the tradeoff studies was related to the engine operation or the environment in which EBC systems are used. These variables are the film coefficients on the hot side and back side that control, via convection, the heat flow through the EBC and CMC. The hot-side convection is due to the hot combustion gases. The back-side convection is due to the impingement cooling that is generally implemented to cool the CMC substrate. Figure 8 shows a variation in hot-side film coefficients from 0.9 to 3.4 and the effect on the resulting thermal profile. From equation (1), it is clear that the hot-side film coefficient increases the heat flow through convection and yields the temperatures in the EBC system layers seen in figure 8. An increase in the cold-side film coefficient, on the other hand, results in greater heat flow out (eq. (8)), thereby leading to a significant drop in the EBC system layer temperatures, as shown in figure 9. These results show that the film coefficients, in general,

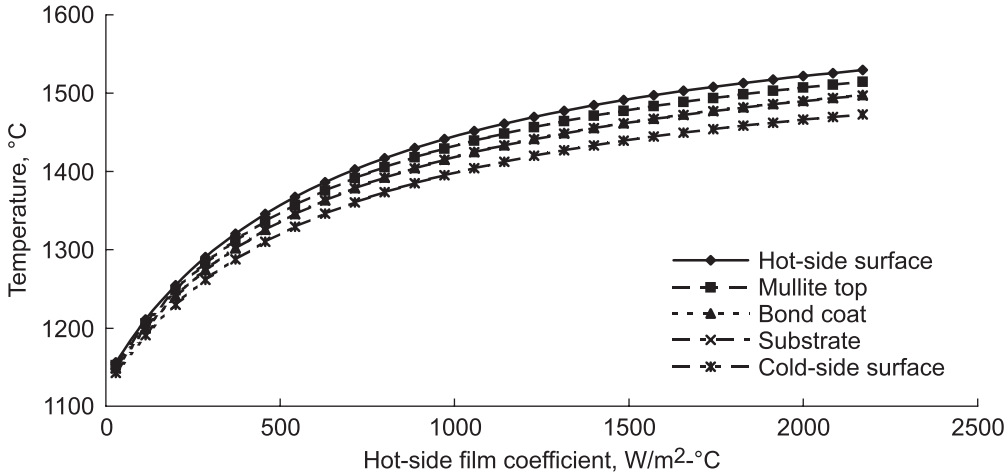

Figure 8.—Effect of variation in hot-side coefficient.

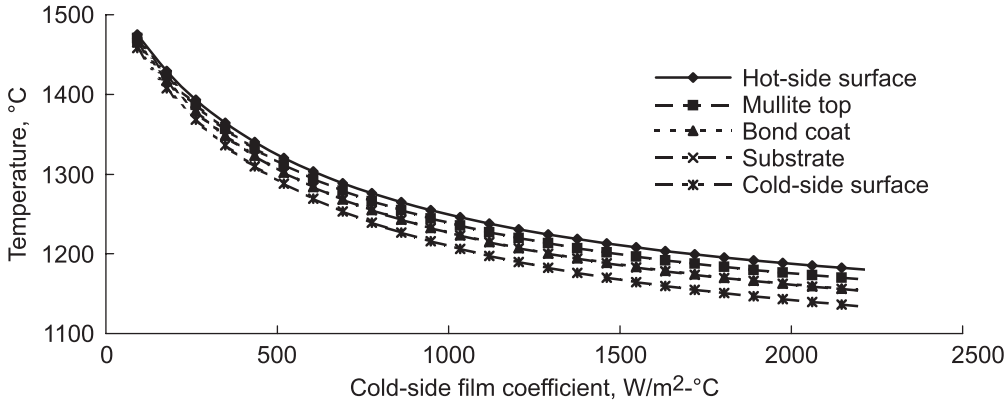

Figure 9.—Effect of variation in cold-side coefficient.

and the impingement-cooling-related coefficient, in particular, are design parameters that control the resulting thermal profile very effectively. However, the temperature difference computed between the EBC top surface and the CMC top surface is barely 30 °C, which is negligible.

For the selected range of variables, the tradeoff studies showed that the maximum difference achievable between the EBC top and CMC top temperature was only 90 °C. The tradeoff studies also showed the existence of more than one design configuration for a given temperature difference. In view of the multitude of possibilities and a number of competing design variables, we recommended that a formal optimization be performed in order to include the effect of simultaneous interactions of the design parameters and constraints. A sample optimization study was performed, and a typical set of results is given in tables II and III.

Although the optimization study revealed a configuration with a temperature difference of 149 °C, the unusually thick EBC required makes the configuration impractical. However, further optimization investigations with modified constraints may lead to a design configuration that is feasible to manufacture and achieve the design objectives.

TABLE II.—BOUNDS FOR SAMPLE OPTIMIZATION STUDY

Variable	Upper bound	Lower bound
EBC conductivity, W/m-K	3.115	1.66
Mullite thickness, mm	0.508	0.127
Mullite conductivity, W/m-K	3.115	1.66
Bond coat thickness, mm	0.102	0.051
EBC thickness, mm	0.508	0.127
EBC surface temperature, °C	1482	1479
Film coefficient of hot-side (EBC) surface, W/hr-m^2-°C	113.54	3.122

TABLE III.—OPTIMIZED CONFIGURATION

Thickness, mm		
EBC		0.35772
Mullite		0.39615
Bond coat		0.05853
Substrate		2.03200
Conductivity, W/m-K		
EBC		2.48108
Mullite		2.42314
Bond coat		22.10011
Substrate		22.00000
Film coefficient, W/hr-m^2-°C		
Hot side		3170.0
Cold side		2383.0
Temperatures	°F	°C
Hot-side gas	3000.000	1648.889
EBC surface	2695.000	1479.444
Mullite top	2555.615	1402.008
Bond-coat top	2397.562	1314.201
Substrate top	2395.001	1312.778
Substrate bottom	2305.708	1263.171
Back-side gas	1900.000	1037.778

Summary of Results

Collectively, the study results indicate that there are three major sets of design parameters that control the thermal profile through the coated CMC material. The first set is the conductivities of the BSAS (EBC) and mullite layers. Only the lower range of conductivities have a significant influence. An indefinite increase beyond this lower regime does not affect the thermal profile significantly. The second set of design variables is the thicknesses of the EBC and mullite layers. Here, a monotonic increase of the layer temperatures is seen as the thicknesses of the EBC and the mullite layers are increased. However, it should be noted that EBC layer temperatures in excess of 1482 °C are unacceptable because of the glass transformation of EBC. Furthermore, manufacturing of thick EBC poses additional geometrical limitations as well as higher residual stresses because multiple passes of plasma spray are needed. The change in the bond coat thickness has shown no effect on the thermal profile in the EBC system. The last set of design variables are related to engine operation and consist of hot-side and back-side film coefficients, which control the convective heat transfer. An increase of the hot-side film coefficient could lead to an unacceptably higher temperature in the EBC as well as CMC surfaces. However, the rate of increase in the EBC surface temperature is much higher than that of the CMC surface temperature. Thus, the temperature difference between the EBC and CMC surface increases with the hot-side film coefficient. On the other hand, an increase in the cold-side film coefficient reduces the EBC as well as the CMC temperatures significantly.

The tradeoff studies clearly show the existence of more than one design configuration for a given EBC top and CMC top temperature difference. Within the selected range of variable magnitudes, the maximum difference that can be achieved between the EBC top and CMC top temperatures is 90 °C. Because of the multitude of possibilities and the number of competing design variables, a representative optimization study was performed. On the basis of this study, we recommend that a formal optimization be performed to include the effect of the simultaneous interaction of the various design parameters and constraints. Also, because the temperatures are very high in the combustor, radiation effects may have to be accounted for in the analysis as well. One way to approach this problem is to embed the radiation effects into an equivalent hot-side film coefficient.

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14. ABSTRACT
Recent interest in environmental/thermal barrier coatings (EBC/TBCs) has prompted research to develop life-prediction methodologies for the coating systems of advanced high-temperature ceramic matrix composites (CMCs). Heat-transfer analysis of EBC/TBCs for CMCs is an essential part of the effort. It helps establish the resulting thermal profile through the thickness of the CMC that is protected by the EBC/TBC system. This report documents the results of a one-dimensional analysis of an advanced high-temperature CMC system protected with an EBC/TBC system. The one-dimensional analysis was used for tradeoff studies involving parametric variation of the conductivity; the thickness of the EBC/TBCs, bond coat, and CMC substrate; and the cooling requirements. The insight gained from the results will be used to configure a viable EBC/TBC system for CMC liners that meet the desired hot surface, cold surface, and substrate temperature requirements.

15. SUBJECT TERMS
Life prediction; Environment barrier coatings; Thermal barrier coatings; Spalling; Sintering; Oxidation; Cracking; Heat transfer analysis; Convective heat flow; Conduction; CMC substrate; Environmental effects

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