

## **THERMAL RESIDUAL STRESS IN ENVIRONMENTAL BARRIER COATED SILICON NITRIDE-MODELED**

Abdul-Aziz Ali  
Cleveland State University  
2121 Euclid Avenue  
Cleveland, OH 44115

Ramakrishna T. Bhatt  
Vehicle Technology Center  
US Army Research Laboratory  
Glenn Research Center at Lewis Field  
21000 Brookpark Rd.  
Cleveland, Ohio 44135

### **ABSTRACT**

When exposed to combustion environments containing moisture both un-reinforced and fiber reinforced silicon based ceramic materials tend to undergo surface recession. To avoid surface recession environmental barrier coating systems are required. However, due to differences in the elastic and thermal properties of the substrate and the environmental barrier coating, thermal residual stresses can be generated in the coated substrate. Depending on their magnitude and nature of the thermal residual stresses can have significant influence on the strength and fracture behavior of coated substrates. To determine the maximum residual stresses developed during deposition of the coatings, a finite element model (FEM) was developed. Using this model, the thermal residual stresses were predicted in silicon nitride substrates coated with three environmental coating systems namely barium strontium aluminum silicate (BSAS), rare earth mono silicate (REMS) and earth mono di-silicate (REDS). A parametric study was also conducted to determine the influence of coating layer thickness and material parameters on thermal residual stress. Results indicate that z-direction stresses in all three systems are small and negligible, but maximum in-plane stresses can be significant depending on the composition of the constituent layer and the distance from the substrate. The BSAS and REDS systems show much lower thermal residual stresses than REMS system. Parametric analysis indicates that in each system, the thermal residual stresses can be decreased with decreasing the modulus and thickness of the coating.

### **INTRODUCTION**

Monolithic silicon nitride is a candidate material for next generation small engine turbine components because of its low density, high temperature strength, and high creep resistance. In addition, the processing methodology for the fabrication of complex shaped silicon nitride components is also well developed. Despite these advantages, silicon nitride is not used for flight worthy hardware of turbines because of its poor impact resistance and structural instability (surface recession) in engine operating environments [1, 2]. While impact resistance can be mitigated by component design, avoiding surface recession requires development of compatible multilayered environmental barrier coating (EBC). Problem of surface recession is not unique for silicon nitride, it also occurs in all other silicon based ceramics and SiC/SiC composites when exposed to a combustion environment containing moisture at temperatures > 1100°C [3, 4]. To protect the SiC/SiC composites from surface recession, environmental barrier coatings (EBC) have been developed. One key example is a multilayered coating having a barium strontium aluminum silicate (BSAS) and rare earth silicate top coat [5, 6]. The BSAS and rare earth silicate based EBCs have upper temperature capabilities of ~1316°C [6] and ~1482°C [7], respectively for applications over 30,000 hours. In general, an EBC consists of two or more layers that each layer has its own functionality and characteristics. The layer on top of the substrate is referred to as bond coat is typically silicon, followed by an intermediate coat which is a mixture of Mullite and top coat, and then a top coat. Each one of these

coating layer plays a different role in the protection scheme such as minimizing the migration of sintering of additives from substrate to the coating, avoiding sintering between layers, and reduces permeability of moisture to the substrate. These coating layers can be deposited by a variety of methods such as plasma spray (PS), electron beam physical vapor deposition (EBPVD), or slurry coating.

Literature data indicate that plasma sprayed multilayered BSAS based EBC had no effect on strength properties of SiC/SiC composites, but caused ~50% loss in strength in silicon nitride substrates [8, 9]. Various factors such as coating process causing flaws greater than those existed in the as machined surface, large tensile thermal residual stresses in the coating the substrate surface were suggested as mechanisms for strength degradation

In general, when elastic and thermal properties of the coating and the substrate are different, thermal residual stresses are generated in the coated substrates depending on the thermal environment used during coating process. Depending on their nature and magnitude, thermal residual stresses can have significant effect on the strength of the substrate. To predict as well as to develop methods of lowering thermal residual stress in coated substrates the current study was started.

The objectives of this study were several: First, to develop an analytical model to predict the magnitude of thermal residual stresses in coated substrates based upon the boundary conditions used during coating process, specimen geometry, and the thermo-mechanical properties of the coating layers and substrate; Second, apply this model to predict stresses in silicon nitride substrates coated with BSAS, Rare Earth Mono Silicate (REMS), and Rare Earth Di Silicate (REDS) EBC systems by plasma spray; Third, to determine the influence of coating thickness and modulus on the magnitude of residual stresses.

## **ANALYTICAL APPROACH AND FINITE ELEMENT MODELING**

The finite element method is used to determine the build up of residual stresses for the proposed layered EBC. These analytical calculations included modeling of a beam specimen with a layer of EBC and Silicon Nitride substrate. The specimen dimensions were 4 by 3 by 45 mm, Figure 1 Three different EBC thicknesses and a substrate structure are modeled. Thermal boundary conditions effects imposed by the coating application methodology are introduced into the thermal model. MSC/Patran [10] and MARC [11] finite element modeling software are used for both the geometric modeling of the specimen and as a solver for the stress state. Material properties of both, the coating and the substrate, are input through the model under linear isotropic conditions. Three dimensional model of the specimen is generated and a fine mesh through both the coating layer and the substrate interface is established. Eight-node hex type element is employed with a total average model size for each coating thickness considered of 44,000 elements and 50,000 nodes. Additional details regarding the geometry of the beam model as well as the representative thicknesses of each coating employed in the analyses are reported in Figure 1.

The calculations were made under linear elastic conditions where the behavior of the material is defined by the two material constants; Young's modulus and Poisson's ratio. The coated test specimen consists of three sub layers of EBC and the silicon nitride substrate. Three EBC systems namely, System-1, System-2, and System-3 correspond to BSAS, REMS, and REDS, respectively are considered. Each EBC system consists of 3 sub layers, the first is silicon layer followed by a mixture of Mullite-top layer and then a top layer. For example BSAS EBC system consists of a layer of silicon, a mixture of Mullite +BSAS layer, and then a layer of BSAS. In each EBC system two different coating thicknesses referred to in the paper as analytical case -1 and 2 were analyzed. The analytical case-1 which represents total coating thickness of 225  $\mu\text{m}$  consists of 75  $\mu\text{m}$  silicon bond coat layer, 75  $\mu\text{m}$  intermediate coat layer, and 75  $\mu\text{m}$  top coat layer. The analytical case-2 which represents 325  $\mu\text{m}$  thick EBC consists of 75  $\mu\text{m}$  silicon bond coat layer, 125  $\mu\text{m}$  intermediate coat layer, and 125  $\mu\text{m}$  top coat layer.

Material property data of the silicon nitride substrate and the individual coating layer of environmental barrier coating systems required for the modeling were obtained from reference 12. Table I shows the properties for both the coating and the substrate. Material properties for the combined coatings were derived using the rule-of-Mixtures to calculate the effective values. Thermal and mechanical boundary conditions applied during the coating application procedure as well as the material properties effects were all introduced into the analytical model. The beam geometry was constrained to match the experimental conditions and to suppress

rigid body motions effects. Additionally, in order to eliminate factors such as over constraining, edge effects and other related issues; the entire specimen unit was modeled and meshed accordingly. This was a reasonably valid assumption since the analyses were performed under linear elastic conditions. Thermal environment was imitated by applying an initial temperature of 1200 °C to the entire structure followed by a cool down to room temperature of 21 °C. This represented the thermal setting experienced by the beam specimen while being plasma sprayed. Subsequently, residual stresses in the coating, the substrate and the combined structure are all determined and evaluated.

Table I. Physical, thermal and mechanical properties of silicon nitride substrate and stand alone plasma sprayed coatings [12]

Material	$\rho$ gm/cc	Bend Modulus, GPa	Bend Strength, MPa	Thermal Expansion, $\alpha$ E-6/°C	Poisson' Ratio, $\gamma$
<b>Substrate, Silicon nitride</b>	3.4	318	595	3.8	0.18
<b>Plasma sprayed coatings</b>					
<b>Mullite</b>	2.9	45	28	5.8	0.17
<b>Rare Earth-Mono Silicate (REMS)</b>	7.9	91	57	8.1	0.18
<b>Silicon</b>	2.3	97	40	4.5	0.21
<b>Rare Earth Disilicate (REDS)</b>	7.9	90	60	4.8	0.18
<b>Barium Strontium Aluminum Silicate (BSAS)</b>	3.2	32	28	5.6	0.19

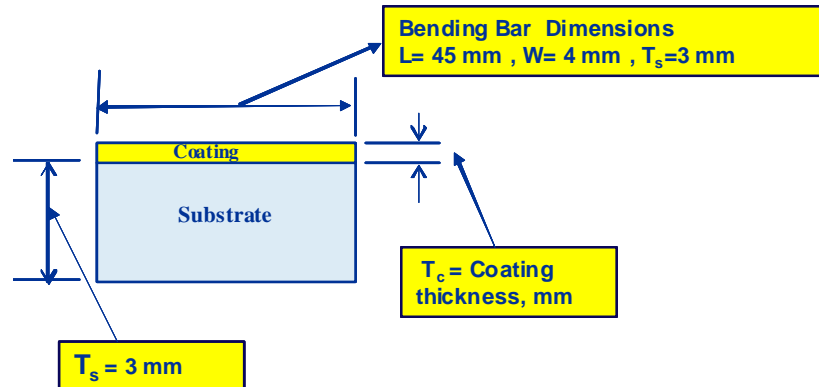


Figure 1. Test Bar Specimen Geometry

## RESULTS AND DISCUSSION

Results obtained from the finite element analyses are illustrated in Figures 2 to 6. Figure 2 shows the stresses in the bar specimen due to the thermal loading applied for both the coating and the substrate. These results are for system-1 with coating sequence as Silicon-(Mullite+BSAS)-BSAS and case-1 respectively. The coating thickness of each layer is 75  $\mu$ m. The coating and substrate thickness is designated by the symbols  $t_c$  and  $t_s$  and they represent the incremental thickness of the coating and the substrate respectively, while  $T_c$  and  $T_s$  are the total thickness of each entity. The location at the substrate/coating interface is represented by a ratio of zero for  $t_c/T_c$ , while a

ratio of unity represents maximum coating thickness. Similar convention is used concerning the ratio of the substrate thickness arrangement .i.e.  $t_s/T_s$  corresponds to maximum substrate thickness In figure 2(a) the stresses along the thickness, Z-axis, and along the X-Y axes are shown as a function of the normalized distance. The normalized distance is defined as the ratio of the length increment divided by the total thickness. It is reported from the data presented that the X-Y stresses are much higher than the through thickness stress; in fact the through thickness stresses are nearly negligible as projected. Figure 2(b) represents the stresses along the X, Y and Z axis through the substrate. Their magnitude is relatively small.

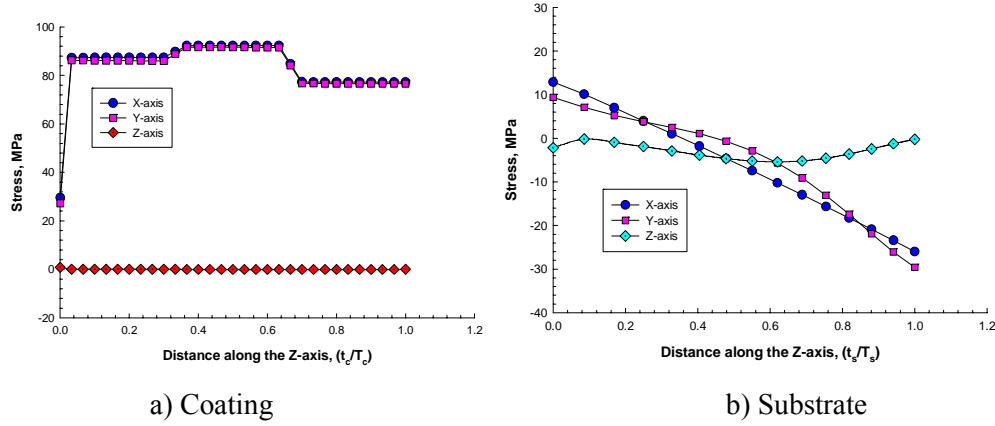


Figure 2. Predicted variation of thermal residual in-plane stress versus normalized distance for System 1, Case 1.

Similarly, the predicted stresses for the system-1, analytical case-2 are shown in Figures 3(a) and 3(b). The data shown in Figure 3(a) correspond to the in-plane and the through thickness stresses in the coating. In this case, the bond coat thickness is much smaller than the subsequent two coats. Comparison of figure 2(a) and 3(a) indicates that following. First, the X and Y stresses in the intermediate layer of the coating in both cases are relatively greater than either the silicon bond coat layer or top BSAS layer. Second, the X and Y stresses in the silicon bond coat for case 1 are nearly the same as that in the intermediate coat, but for case 2, X and Y stresses in the silicon bond coat are almost zero. Third, in general X and Y stresses for case 2 are  $\sim 25\%$  lower than case 1 which suggests that by manipulating relative thickness of individual layers as well as increasing overall thickness of the coating, it is possible to decrease thermal stresses.

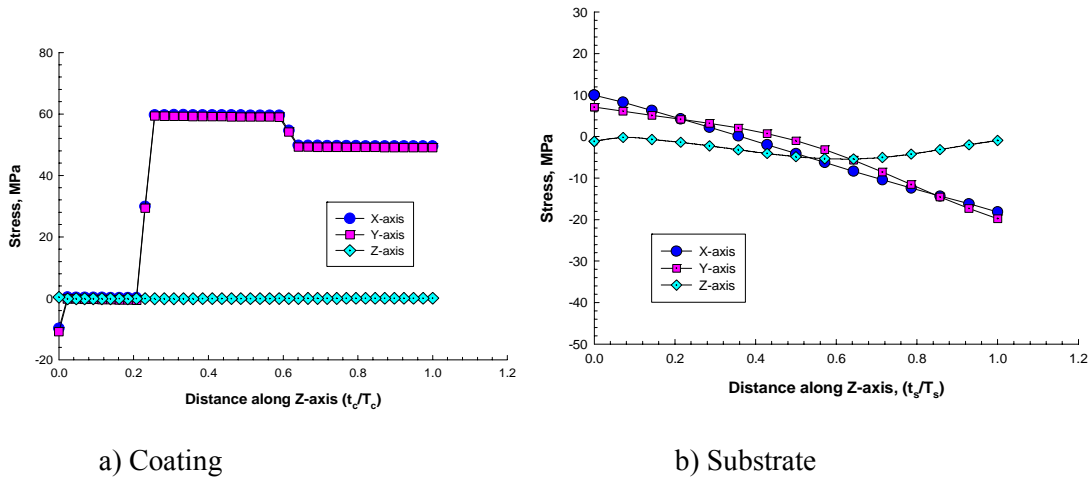


Figure 3. Predicted variation of thermal residual in-plane stress versus normalized distance for System 1, Case 2.

Figure 4(a) and 4(b) show the results obtained for System-2, case-1. It is very clear from Figure 4(a) that the in-plane stresses are substantially larger than those experienced by system-1 under the same conditions of case-1, Figure 2(a). They are an order of magnitude higher compared to case1/system-1 data. Similarly, the stresses through the substrate under system-2, case-1 also showed slightly higher stresses, nearly doubles in magnitude, compared to those obtained under system1, case1. However, their extents constitute no significance on the structural behavior of the silicon nitride, they remain relatively small.

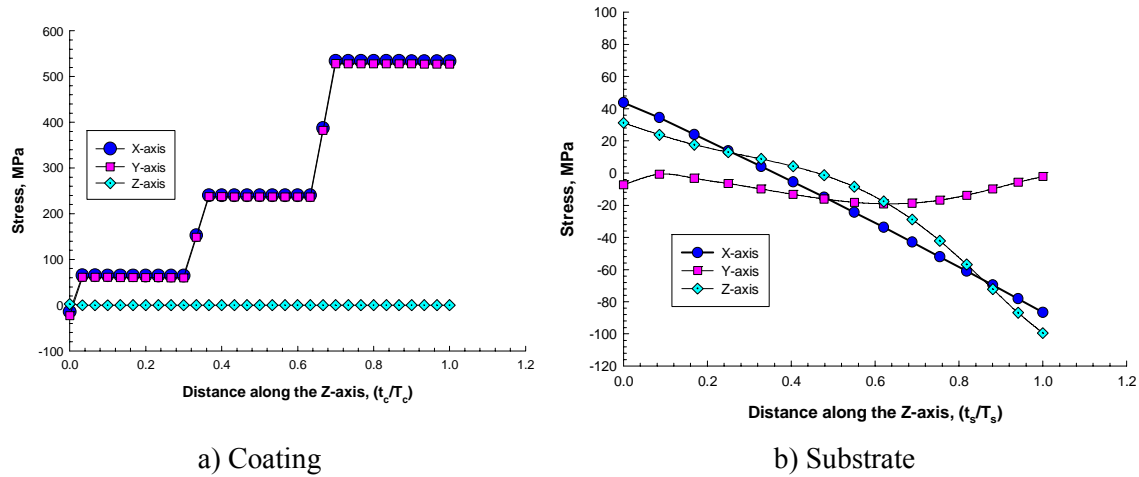


Figure 4. Predicted variation of thermal residual in-plane stress versus normalized distance, System-2, Case-1.

Figures 5(a) and 5(b) represent results obtained under the same thermal conditions for the System 3, Case-1. The elastic properties of system-3 are similar to that of System-2, but the thermal expansion of System-3 is nearly half that of System-2.

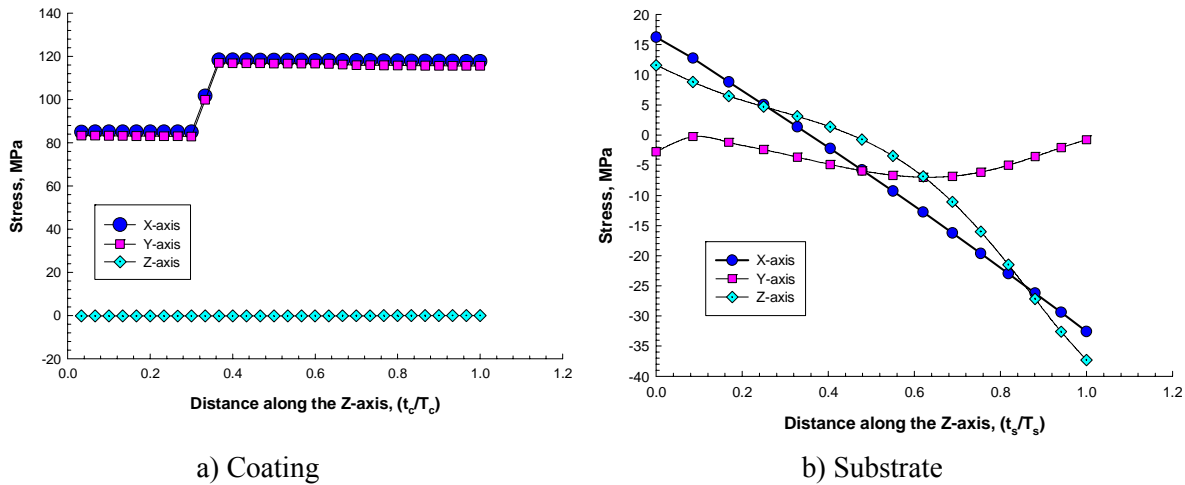


Figure 5. Predicted variation of thermal residual in-plane stress versus normalized distance for System-3, Case-1.

Figure 5 shows that X and Y stresses in this system are almost half that of System-2, Case-1 and are very comparable to the results obtained for the System-1, Case-1. Similarly, the stresses reported through the substrate are equally low and insignificant. Comparison of modeling data for all three systems suggests that both system-1 and System-3 are better systems for development of EBC for silicon nitride.

In plasma spray deposition, it is possible control porosity in the coating by varying processing parameters, thus varying the modulus of the coating. Influence of coating modulus on the X and Y stresses for System-1 and System-2 for Case-1 was modeled. The results plotted in figure 6 show that decreasing coating modulus decreases the stresses linearly. Figure 6 shows an exclusive chart illustrating the maximum tensile residual stress experienced by each system under the same operating conditions. The chart shows the stress versus the modulus ratio with E being the modified modulus and  $E_0$  the initial modulus of the coating respectively. This demonstrates that the modulus influence is obvious. It is all shown in figure 6; much higher tensile stresses are reported for the System-2 versus those reported for the System-1. It further confirms that variations in the material properties such as modulus and CTE will affect the thermal response produced by either the System-1 or the System-2. Lower thermal residual stresses are a product of a lower CTE.

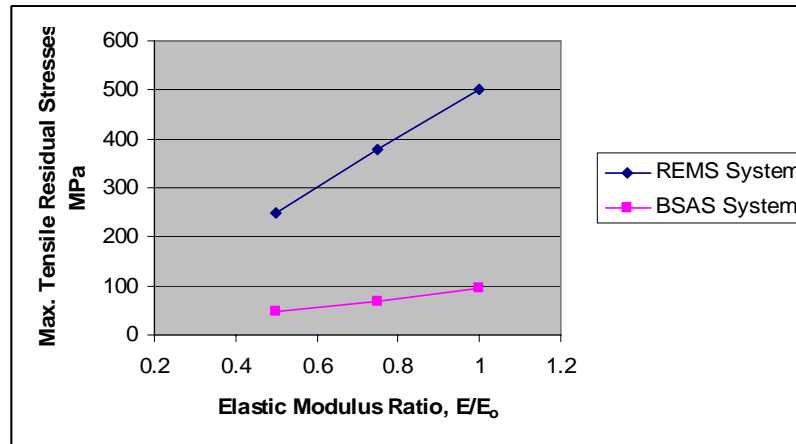


Figure 6. Influence of coating modulus on thermal residual stress for silicon nitride coated with REMS and BSAS systems, case-1.

## CONCLUSIONS

A finite element model was developed to predict in-plane and through-the-thickness stresses in a rectangular beam of coated ceramic substrate simulating the experimental environment conditions experienced during the coating process and using the constituent properties of the coating and the substrate. Thermal residual stresses developed in silicon nitride substrate coated with three environmental barrier coating systems- BSAS, REMS, REDS- (systems 1, 2 and 3) and for two overall coating thicknesses (analytical cases 1 and 2) were modeled. Influence of coating modulus and thermal expansion on the thermal stresses was studied. Major findings are the following.

- (1) The z-direction (through thickness) stresses in all three systems for case-1 are small and negligible, but maximum in-plane stresses can be significant depending on the composition of the constituent layer and the distance from the substrate.
- (2) The in-plane thermal residual stresses for System-2, Case-1 is much higher than those for System-1 or System-3, case-1.
- (3) Reducing the elastic modulus and the coefficient of thermal expansion will result in lowering stress response.

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