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Journal of the European Ceramic Society 28 (2008) 711–716

www.elsevier.com/locate/jeurceramsoc

Optimizing substrate and intermediate layers geometry to reduce internal thermal stresses and prevent surface crack formation in 2-D multilayered ceramic coatings

R. Valle *, D. Lévêque, M. Parlier

ONERA/DMSC, 29 avenue de la Division Leclerc, F-92322 Châtillon Cedex, France

Received 7 April 2007; received in revised form 16 July 2007; accepted 27 July 2007
Available online 18 October 2007

Abstract

In order to avoid the formation of normal cracks in the external layer of multilayered ceramic coatings during cooling, determination of the residual thermal stress field is essential. Concerning the effect of the intermediate layers (transition zone) on a possible reduction of stress in the external layer, it has been shown that in the case of a symmetrical (or constrained in-plane strain) system, reduction only occurs over a critical substrate thickness. In order to investigate the effect of the substrate geometry, the residual thermal stress field has also been determined in other configurations such as an asymmetrical arrangement (one side coating) allowing bending and a concentric-cylinder structure. In these last two configurations, a strong stress reduction is obtained. For practical applications such as gas turbine ceramic components, these results may provide guidelines for an optimum design of the geometry of substrate and intermediate layers of multilayered ceramic coatings.

Keywords: Coatings; Multilayered structures; Fracture; SiC; Al_2O_3; Mullite; Engine components; Thermal stresses

1. Introduction

Multilayered coatings are developed in order to protect mechanical parts from oxidation and/or hot corrosion. In the case of functionally graded coatings, the various layers possess specific thermo-mechanical and chemical properties. For example, a bond coat, directly deposited on the substrate may act as a thermal barrier, an outer ceramic layer is aimed at protecting the substrate from corrosion whereas an intermediate layer may play the role of a transition zone. Due to thermal expansion mismatch, residual thermal stresses develop during cooling of such a multilayered system. In order to avoid the formation of normal cracks in the external layer, determination of the stress field in each layer, and especially in the external layer, is essential. However, the residual thermal stresses which develop in each layer depend on the geometrical configuration of the system. In this respect, the most commonly found situation is that of a planar arrangement, i.e. a symmetrical or constrained in-plane strain system. Consequently, the effect of the presence of the intermediate layers on a possible stress reduction in the external layer will be investigated first in a symmetrical multilayered system. Concerning the effect of the substrate geometry on a possible stress reduction in the external layer, two other configurations will be taken into consideration: an asymmetrical arrangement allowing bending and a concentric-cylinder system.

All constituents are supposed to be ceramics; these three configurations will thus be analyzed through calculations in linear elasticity. The residual thermal stresses are determined in a region remote from the free edges; consequently, free edge singularities leading to interfacial peeling are not taken into consideration. A practical example will allow investigating the effect of the intermediate layers on a possible stress reduction in the external layer of the symmetrical system. These results will then be compared with those corresponding to asymmetrical and cylindrical configurations.

2. Analytical modeling of the residual thermal stresses

2.1. Geometrical configurations and boundary conditions

The various geometrical configurations of multilayered systems and the various boundary conditions lead to different

* Corresponding author. Tel.: +33 1 46 73 45 69; fax: +33 1 46 73 41 42.
E-mail address: roger.valle@onera.fr (R. Valle).

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doi:10.1016/j.jeurceramsoc.2007.07.017

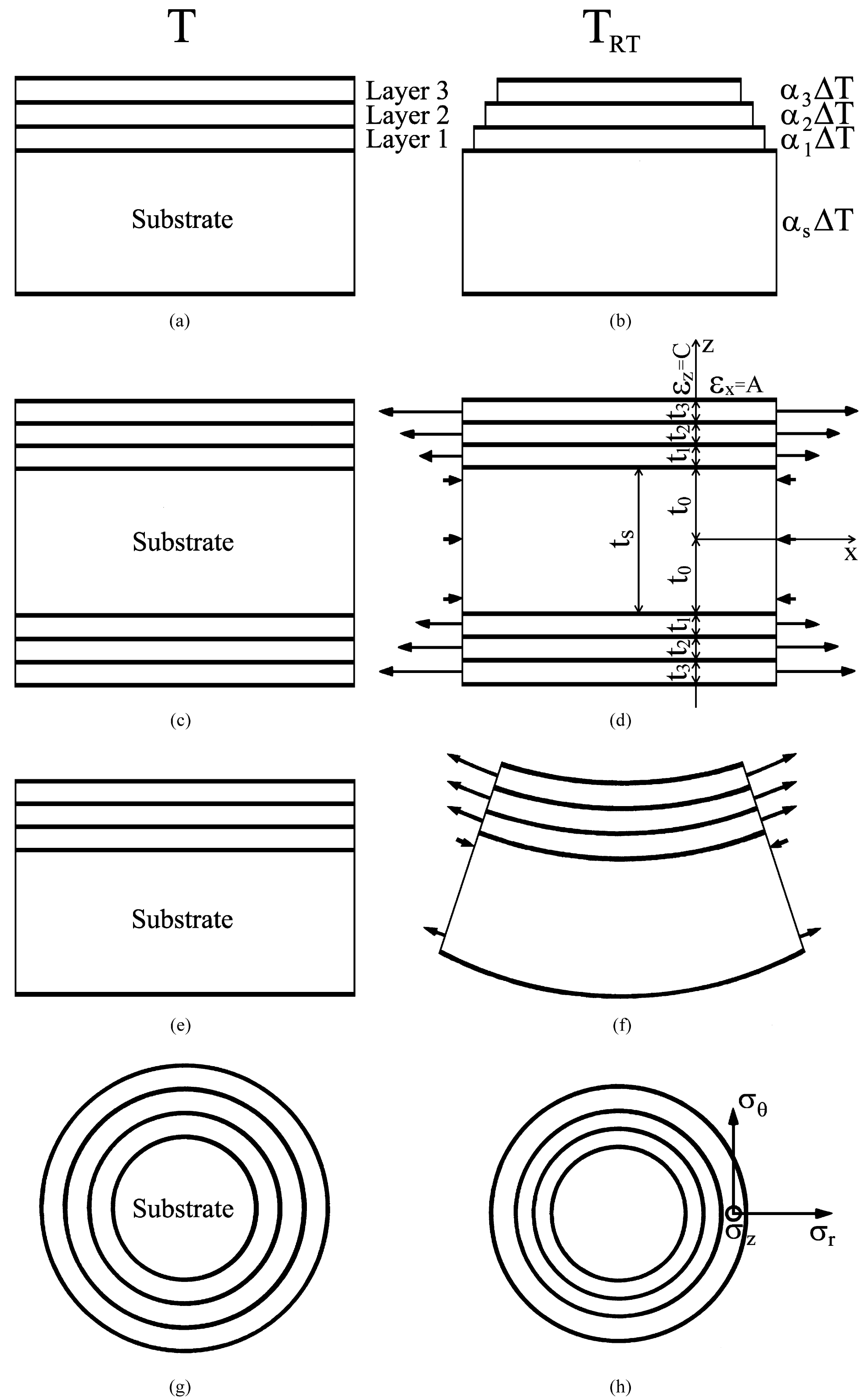

Fig. 1. Multilayered coatings on a planar (a–f) or cylindrical (g and h) substrate. Stress-free initial condition at the fabrication temperature T (a, c, e and g). After cooling down to the room temperature T_{RT}, unconstrained differential shrinkage $\alpha_i \Delta T$ in the case of free sliding between the plates (b), tensile and compressive stresses due to thermal expansion mismatch if perfect bonding is assumed (d, f and h), stress reduction through bending (f) or through radial compression of the intermediate cylindrical coating layers (h).

thermo-mechanical behaviours. At the fabrication temperature, all the systems are in a stress-free initial condition (Fig. 1a, c, e and g). In each case, the thermo-mechanical properties of the ith layer are represented by α_i, coefficient of thermal expansion (CTE), E_i, Young's modulus and ν_i, Poisson's ratio. Let us consider a four-layer system: the substrate and three coating layers. Each constituent is represented by a plate of a large size in the x–y plane and of a uniform thickness in the normal direction z. Slow cooling being assumed, the temperature is supposed to be uniform throughout the multilayered structure (i.e. there is no thermal gradient). During cooling from the fabrication temperature, T, to room temperature, T_{RT}, if free sliding between the plates is allowed, then each plate (i) experiences an unconstrained differential shrinkage during cooling ($\varepsilon_i = \alpha_i \Delta T$), where $\Delta T = T_{\mathrm{RT}} - T$ (Fig. 1b). However, if perfect bonding is assumed between the substrate and the various layers (i.e. no interface sliding), tensile and compressive stresses are imposed on the substrate and on the coating layers in order to achieve the displacement compatibility at the various interfaces between the plates. If a boundary condition is applied to the plates so that they have to remain flat during cooling, i.e. if bending is impeded, then the resulting solution is that of constrained in-plane strain. This situation is attained in the symmetrical configuration of an n-layered coating on each side of a substrate (Fig. 1c and d). In the case of an asymmetrical system (Fig. 1e), bending occurs in order to balance the bending moment induced by the asymmetric stress distribution (Fig. 1f). The resulting bending strains reduce the stress system due to thermal expansion mismatch. In a two-layer, one-dimensional system, the problem is that of the bimetal thermostat, first examined by Timoshenko.[1] In the case of a multilayered coating on a cylindrical substrate, the axial symmetry is maintained during cooling (Fig. 1g and h). If the Young's moduli of the intermediate layers of a three-layer coating are relatively low as compared to that of the external layer, then the circumferential stress component in the external coating is reduced through radial compression of the intermediate layers (Fig. 1h).

2.2. *Symmetrical system or constrained in-plane strain asymmetrical system*

The corresponding multilayer is modeled as an infinite 2-D multilayered plate. It should be noted that such a symmetrical $(2n+1)$-layer laminate (Fig. 1d) is equivalent to an asymmetrical n-layer coating on a substrate (layer 0, thickness $t_0 = t_s/2$), provided that the generalized plane strain condition ($\varepsilon_z = C$) is assumed. From a practical viewpoint, this constrained in-plane strain system corresponds to a one side coated substrate, sufficiently thick and rigid as compared to the coatings, that bending may be neglected.

For a layer in the x–y plane (Fig. 1d), a plane stress state ($\sigma_z = 0$) is assumed. Then, the three-dimensional stress–strain relations in a linear elastic isotropic medium subjected to a temperature change ΔT directly give the stress–strain relation for plane biaxial stress distribution in each layer (i)

$$\varepsilon_i - \alpha_i\,\Delta T = \frac{\sigma_i(1-\nu_i)}{E_i} \tag{1}$$

The system being infinite in the x and y directions, the generalized plane strain condition in planes perpendicular to the laminate surface is assumed (Fig. 1d)

$$\varepsilon_{0x} = \varepsilon_{0y} = \varepsilon_0 = \cdots = \varepsilon_{ix} = \varepsilon_{iy} = \varepsilon_i = \cdots = A = \varepsilon$$

Eq. (1) may thus be rewritten as

$$\varepsilon - \alpha_i\,\Delta T = \frac{\sigma_i(1-\nu_i)}{E_i} \tag{2}$$

hence

$$\sigma_i = \frac{E_i}{1-\nu_i}(\varepsilon - \alpha_i\,\Delta T) \tag{3}$$

The boundary condition on the biaxial stress component in the multilayer (stress equilibrium) is given by

$$\sum_{j=0}^{n} t_j\sigma_j = 0 \tag{4}$$

where t_j is the thickness of layer (j).

Reporting the expression (3) of σ_i in relation (4) yields

$$\sum_{j=0}^{n}\frac{t_jE_j}{1-\nu_j}\varepsilon = \sum_{j=0}^{n}\frac{t_jE_j}{1-\nu_j}\alpha_j\,\Delta T$$

Introducing the resulting expression of ε in relation (3) gives the stress component (σ_i) in each layer

$$\sigma_i = \frac{E_i}{1-\nu_i}\,\frac{\sum_{j=0}^{n}E_jt_j(\alpha_j-\alpha_i)/(1-\nu_j)}{\sum_{j=0}^{n}E_jt_j/(1-\nu_j)}\,\Delta T \tag{5}$$

For example, in the present case of a four-phase system (three coating layers of thickness t_1, t_2 and t_3 on each side of a substrate (s) of thickness $t_s = 2t_0$), the residual thermal stress in the external layer is given by

$$\sigma_3 = \frac{E_3}{1-\nu_3}\left[\frac{E_s}{1-\nu_s}t_0(\alpha_s-\alpha_3) + \frac{E_1}{1-\nu_1}t_1(\alpha_1-\alpha_3) + \frac{E_2}{1-\nu_2}t_2(\alpha_2-\alpha_3)\right]\frac{\Delta T}{D} \tag{6}$$

where

$$D = \frac{E_s}{1-\nu_s}t_0 + \frac{E_1}{1-\nu_1}t_1 + \frac{E_2}{1-\nu_2}t_2 + \frac{E_3}{1-\nu_3}t_3$$

In the case of a symmetrical system with $2n+1$ alternate layers of only two types, the stress components are given by much simpler expressions[2]; in a two-phase system (a coating layer of thickness t_1 on each side of a substrate of thickness $t_s = 2t_0$), the stress component in the coating layer is given by[3]

$$\sigma_1 = \frac{E_1}{1-\nu_1}\,\frac{\Delta\alpha\,\Delta T}{1+(t_1/t_0)(E_1/E_s)(1-\nu_s)/(1-\nu_1)} \tag{7}$$

where

$$\Delta\alpha = \alpha_s - \alpha_1$$

It should be noted that in the absence of flexure (multilayered symmetrical plate or constrained in-plane strain asymmetrical system), the stress component (σ_i) in each layer (i) is a constant throughout the layer thickness.

2.3. Asymmetrical system and concentric-cylinder structure

In the case of an asymmetrical multilayered plate (Fig. 1e and f), the situation is more complex than in the symmetrical system. In fact, due to bending, the stress component in each layer is no more a constant within the layer thickness, but a function of z. An analytical solution may still be obtained.[4] The strain distribution in the system is decomposed into a uniform component and a bending component, thus leading to analytical expressions of the uniform strain component, the position (z coordinate) of the bending axis and the radius of curvature.[4] Using these expressions may lead to exact analytical formulae of the stress components as a function of the geometrical and thermo-mechanical parameters: E_i, ν_i, α_i and t_i. However, these expressions of the stress components would be so complex that they would no more allow a direct evidence of the influence of the various thermo-mechanical parameters. Consequently, in the four-layer system, i.e. in the presence of the intermediate layers, the value of the stress component on the external surface of the external coating is determined through the laminate theory,[5–7] the calculations being performed by numerical resolution of the system of equations resulting from this theory. The stress component in the external coating layer is characterized by its value on the external surface. In the two-layer system (a coating layer of thickness t_1 on a substrate of thickness t_s), the analytical formulae obtained in the case of the bi-metal thermostat (one-dimensional strip)[1] may be extended to a two-layer plate (biaxial system) by replacing the Young's moduli E_s and E_1 by $E_s/(1-\nu_s)$ and $E_1/(1-\nu_1)$, respectively.[1] Although more complex than relations (6) and (7), the expression of the stress component in the external surface of the coating may be rewritten as

$$\sigma_1^{\text{ext}} = \frac{E_1}{1-\nu_1}\frac{(m^2+1/mn)-3m(1+m)}{3(1+m)^2+(m^2+1/mn)(1+mn)}\,\Delta\alpha\,\Delta T \tag{8}$$

where

$$m=\frac{t_1}{t_s},\quad n=\frac{E_1}{E_s}\frac{1-\nu_s}{1-\nu_1}$$

Furthermore, a symmetrical laminate being only a special configuration of an asymmetrical laminate, a practical numerical application will easily allow comparing, and thus confirming, the results given by both the very simple closed-form solutions (Eqs. (6), (8) and (9)) and the laminate theory.

The mechanical problem of a multilayered coating on a cylindrical substrate (s) is that of $n+1$ concentric cylinders subjected to a temperature change ΔT. This concentric-cylinder configuration verifies the (r, θ, z) axial symmetry and is supposed to be infinite in the axial direction (z); moreover, perfect bonding between the various cylinders is assumed. These assumptions lead to the generalized plane strain condition in the axial direction ($\varepsilon_{sz}=\varepsilon_{1z}=\cdots=\varepsilon_{iz}=\cdots=\varepsilon_z$). The stress components in the various phases may be determined by resolution of the system of equations resulting from the boundary conditions on the radial stress component (σ_r), the radial displacement (u_r) and the axial stress component (σ_z). In the case of only four phases, a completely analytical solution may be obtained[8]; the values of the stress components in the cylindrical configuration will be calculated through these formulae. In the two-layer system (a coating layer of external radius $r_1=t_1+t_s/2$ on a substrate of radius $r_0=t_s/2$), the analytical expressions of the stress components in the external surface of the coating may be rewritten as

$$\sigma_{1\theta}^{\text{ext}} = 2VE_sE_1(M+N)Q^{-1}\,\Delta\alpha\,\Delta T \tag{9}$$

$$\sigma_{1z} = VE_sE_1[M+N+(1+\nu_1)E_s]Q^{-1}\,\Delta\alpha\,\Delta T \tag{10}$$

where

$$V=\frac{r_0^2}{r_1^2},\quad M=VE_s+(1-V)E_1,$$
$$N=\nu_1VE_s+\nu_s(1-V)E_1,$$
$$Q=M[M-N+(1+\nu_1)E_s]-2N^2$$

It should be noted that the effect of the geometrical parameters r_1 and r_0 on the magnitude of the stress components is directly evidenced through the factor $V=r_0^2/r_1^2$ in the analytical expressions (9) and (10).

3. Practical application

In order to determine the influence of the intermediate coatings and of the geometrical configuration on a possible reduction in the residual thermal stresses, let us consider the practical example of a multilayered coating which may act as a protection layer against corrosion of a SiC/SiC composite (substrate). The bond coat will be a pure mullite coating (40% porosity), the outer layer an alumina coating (20% porosity) and the intermediate layer a 50% mullite/alumina coating (40% porosity).

Although complex models have been developed for the determination of the thermo-mechanical properties of multiphase materials, the Young's modulus and the coefficient of thermal expansion of the mullite/alumina layer will be determined through linear rule of mixture relations which are commonly used in the domain of ceramics.[9] Similarly, the influence of porosity (P) on the Young's modulus is taken into account through a power function of P.[9] The thermo-mechanical properties thus determined are reported in Table 1.

The residual thermal stresses near the external surface of the outer layer are calculated for the three investigated geometrical configurations and two situations: a three-layer coating and a one-layer coating (the external alumina coating). The results, as a function of the substrate thickness, are reported in Fig. 2.

In the case of a symmetrical coating (no bending), the presence of the intermediate layers (bond coat and transition layer) only slightly reduces the stress level in the outer layer (Fig. 2). This slight reduction may be explained as follows: the substrate and the two intermediate coating layers may be considered as a "composite" substrate whose Young's modulus is lower than

Table 1
Thermo-mechanical data of the substrate and of the coating layers (average values in the temperature range 20–1300 °C)

Material	CTE (10^{-6} °C^{-1})	Young's modulus (GPa)	Poisson's ratio
SiC/SiC substrate (Nicalon fibres)	4.5	250	0.16
Mullite ($3Al_2O_3$–$2SiO_2$)	5	150	0.27
Mullite coating (porosity = 40%)	5	58	0.27
1/2 mullite–1/2 alumina (mol.%) coating (P = 40%)	6	65	0.27
Alumina (Al_2O_3)	8	310	0.27
Alumina coating (P = 20%)	8	200	0.27

that of the isolated substrate and the coefficient of thermal expansion higher (rule of mixture). The thermo-mechanical mismatch between this "composite" substrate and the outer layer is thus smaller than that between the isolated substrate and the outer layer. However, this reduction in the stress level in the external layer only occurs over a critical substrate thickness t_{sc} (Fig. 2). For a lower substrate thickness, the presence of the intermediate layers no more leads to a reduction but to an increase in the stress level in the external layer. This phenomenon may be easily explained through the mathematical forms of the analytical expressions of the stress component in the external layer as a function of the substrate thickness t_s. In fact, relations (7) and (6) may be rewritten as

$$\sigma_{\text{2-layer}}^{\text{ext}} = K\frac{t_s}{1 + Lt_s}$$

and

$$\sigma_{\text{4-layer}}^{\text{ext}} = P\frac{1 + Rt_s}{1 + St_s}$$

where K, L, P, R and S are constants.

Consequently, the presence of the intermediate layers will not have any effect on the stress level in the external layer for a substrate thickness t_s corresponding to the point of intersection of the two curves

$$\sigma_{\text{2-layer}}^{\text{ext}} = \sigma_{\text{4-layer}}^{\text{ext}} = K\frac{t_s}{1 + Lt_s} = P\frac{1 + Rt_s}{1 + St_s} \quad (11)$$

i.e. when t_s is a solution of the resulting second degree equation

$$(KS - PLR)t_s^2 + (K - PL - PR)t_s - P = 0 \quad (12)$$

For the thermo-mechanical data corresponding to the present example (Table 1), $t_{sc} \approx 508$ μm (the other solution is negative).

On the contrary, in the asymmetrical system and in the concentric cylinders structure, it should be noted that, whatever the substrate thickness, the presence of the intermediate coatings leads to a marked reduction in the stress component in the external coating (Fig. 2).

In the cylindrical geometry, radial compression of the low modulus intermediate layers leads to a marked stress reduction which is however more important in the case of smaller diameter substrates (Fig. 2). This phenomenon may be explained as follows: for a given thickness of the intermediate layers, the volume fraction of these two layers increases when the substrate diameter decreases. In the present example, the maximum stress component near the surface of the external layer is the axial stress component whereas the maximum stress component, near the external layer–transition layer interface, is the tangential stress component. In the two-layer system, the role of the mechanical parameters on the prevalence of either the axial or tangential stress as a maximum stress component in the external surface is directly evidenced through the mathematical forms of the analytical expressions (9) and (10): term $(1 + \nu_1)E_s$ in (10) and factor 2 in (9). In the present example, the term $(1 + \nu_1)E_s$ is very slightly superior to $M + N$. Consequently, the axial stress component is just slightly higher than the tangential stress component and thus prevailing.

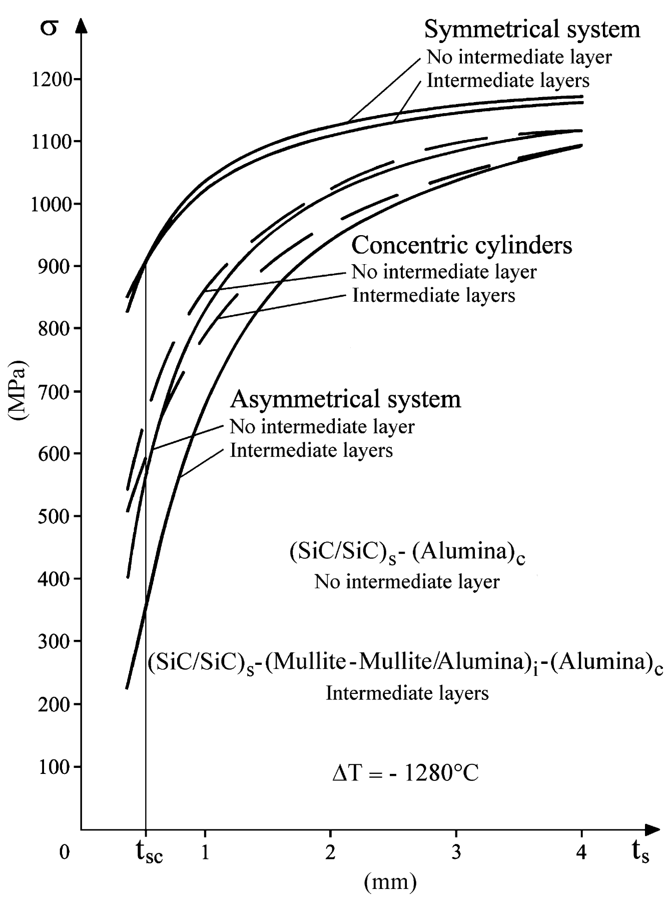

Fig. 2. Maximum stress near the surface of the external coating as a function of the substrate thickness. SiC/SiC substrate, three-layer coating (mullite, t = 150 μm, P = 40%; 1/2 mullite + 1/2 alumina, 100 μm, P = 40%; alumina, 100 μm, P = 20%) or one-layer coating (alumina, 100 μm, P = 20%). Manufacturing temperature: T = 1300 °C ($\Delta T = -1280$ °C). In the case of the symmetrical system, note the existence of a critical substrate thickness t_{sc} under which the presence of the transition layers increases the stress level in the external layer.

In the asymmetrical system, and in the case of a thick substrate (4 mm), the reduction due to the presence of the intermediate layers and to bending only attains the same magnitude as that given by the same intermediate layers on a cylindrical substrate (Fig. 2). However, bending of a thinner substrate (0.5 mm) allows a markedly higher reduction of the residual thermal stresses ($\sigma \approx 345$ MPa) as compared with an identical substrate with symmetrical coating layers impeding bending (≈900 MPa) or with an asymmetrical 4 mm thick substrate (≈1085 MPa).

4. Discussion and conclusion

In the most commonly found situation of a symmetrical (or constrained in-plane strain) structure, the analytical determination of the stress field in each layer of a two-dimensional multilayered coating leads to simple analytical expressions allowing a direct evidence of the influence of the various thermomechanical parameters. Concerning the effect of the presence of intermediate layers (transition zone) on a possible stress reduction in the external layer, it should be noted that the favourable effect attained in the case of thick substrates is rather small. Moreover, the existence of a critical substrate thickness t_{sc} is evidenced. For a substrate thickness lower than t_{sc}, the presence of the transition layers increases the stress level in the external layer. This phenomenon is easily explained through the mathematical forms of the analytical expressions of the stress components which allow the determination of t_{sc}.

Concerning the effect of geometry, it should be noted that, in the case of asymmetrical or concentric cylinders systems, the presence of the intermediate coatings leads to a markedly higher stress reduction in the external coating than in the symmetrical system. In the case of an asymmetrical system, the stress reduction is so strong, especially in the case of a thin substrate (a factor of 3 in the present example), that an undamaged external coating (no crack) may be expected in a three-layer coating on a thin asymmetrical system (bending), whereas the same coating on a symmetrical system would be densely cracked. In practical terms, it means that an experimental multilayered coating, successfully tested on a thin substrate (one side coating), would be densely cracked when used as a protection on a thick and rigid mechanical part. The reduction obtained on a cylindrical system is nearly as strong as that obtained with asymmetrical systems, thus evidencing the advantages of "convex" substrates.

All these results concerning the influence of the geometrical configuration of the substrate and of the role of the intermediate layers may help designing optimum substrate shapes and tailoring functionally graded coatings in order to avoid the formation of cracks in the external protective layer. For instance, the fact that a strong stress reduction occurs for thicknesses lower than ≈2 mm for both the asymmetrical and concentric cylinders systems, suggests the use of thin walled hollow substrates in order to combine these two possibilities of stress reduction (substrate bending and/or radial compression of the transition layers). In the case of a substrate having a complex external shape (e.g. resulting from fluid mechanics calculations for gas turbine components), the thickness of the wall of the hollow structure may be locally adjusted so that the stress level in the external coating would remain lower than the ultimate tensile strength of this coating.

Acknowledgements

The authors would like to thank Dr. N. Carrère, Dr. F.-H. Leroy, M.-H. Ritti and V. Moreno for fruitful discussions.

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