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Bond coat surface rumpling in thermal barrier coatings

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Abstract

Long term durability of thermal barrier coatings is determined by the mechanisms responsible for the nucleation and growth of interfacial cracks between the coating and the substrate. One such mechanism is the progressive roughening or “rumpling” of the bond coat surface upon thermal cycling in superalloy–bond coat systems. The present work is inspired by a recent rumpling experiment [1] pointing to the important role played by the bond coat stresses in the rumpling process. A thermodynamic analysis is presented in this paper based on a bond coat stress-driven surface diffusion mechanism to explain the rumpling process. The surface chemical potential variation required for such a diffusion process is induced by the changes in the surface energy and the elastic strain energy in the system during rumpling. It is found that the amplitude of surface fluctuations with a wavelength higher than a threshold value monotonically increases with thermal cycling, while the amplitude for fluctuations with wavelength lower than the threshold value monotonically decreases. A range of wavelengths is identified to have a high monotonic amplitude increasing rate, resulting in characteristic wavelengths of rumpled surface upon thermal cycling. Effect of various input parameters on these wavelength values is analyzed. It is shown that under certain conditions, the range of wavelengths obtained from the model agree with that observed in several experimental studies.

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1. Introduction

Thermal barrier coatings (TBCs) are presently used as protective heat resistant coatings on superalloy components in the hot sections of gas turbines [2–7]. The TBCs have been shown to reduce the operating temperature of the superalloys by as much as 180 °C, improving the creep and oxidation life of the corresponding gas turbine components.

A survey in early 1990s estimated that the efficiency gains by incorporating the TBC technology could potentially save up to 40 million gallons of fuel (approx. \$1 billion) a year for a fleet of 1000 aircraft [3]. In spite of these advantages, the TBC technology has been incorporated in the gas turbine industry rather slowly.

A TBC system (Fig. 1) consists of a ceramic topcoat and an intermediate (metallic) bond coat (BC) deposited on a substrate superalloy at high temperatures. Two common deposition processes are plasma spray and electron beam physical vapor deposition (EB-PVD). The bond coat is made of

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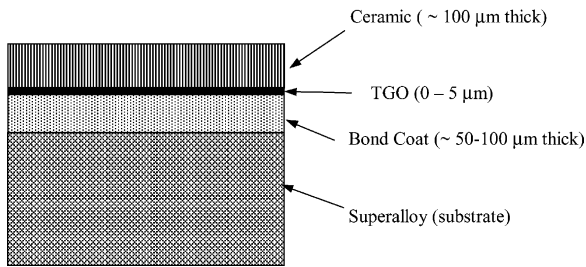


Fig. 1. TBC structure.

either platinum-aluminide or NiCoCrAlY alloys, while the ceramic layer is almost always made of 8% yttria-stabilized zirconia. A reaction product, thermally grown oxide (TGO), develops and grows at the BC–ceramic interface at high temperatures. The thermal expansion coefficients of these constituents of a TBC system are different, giving rise to thermal mismatch stresses as the temperature in the structure changes. In addition, growth stresses exist in the TGO due to continuous formation of TGO material at high temperatures.

One of the important problems limiting the use of TBCs is the long-term durability of these coatings. It has been observed that after a certain amount of service life, the coatings delaminate, exposing the underlying superalloy material. Since the local temperature in the hot section of the gas turbine may exceed the melting temperature of the superalloy, the delamination of TBCs can be very dangerous. The reasons behind this sudden delamination are poorly understood.

While it is generally known that the final TBC failure occurs by buckling and spalling of the ceramic layer, the origin of the required critical flaw is widely debated. Some analyses in the past have pointed to the role of the TGO formed at the BC–ceramic interface [8–11] and the grain boundary ridges on the BC surface [12] in inducing the fatal flaws. Another mechanism, “surface rumpling” of the BC, is also believed to be responsible for such flaws [13–17]. During “rumpling”, the BC surface along with the TGO becomes wavy with a characteristic wavelength upon thermal cycling. Several experiments have been conducted on the superalloy–BC system in the absence of ceramic topcoat to study this rumpling phenomenon [13–17]. Penefather and Boone [13] showed that platinum alu-

minide and NiCoCrAlY coatings of different compositions deposited on a superalloy substrate have a tendency to rumple with characteristic wavelengths ranging from 130–345 μm. The thermal cycle employed in their experiment consisted of cycling the BC–substrate between room temperature and 1050 °C. Deb et al. [14] also found that the platinum modified as well as unmodified aluminide coated superalloy specimens showed rumpling behavior with a wavelength of about 100 μm upon thermal cycling. The thermal cycles in their experiments consisted of a 60 minute exposure at 1100 °C followed by a 10 min air cooling prior to reheating. Tolpygo and Clarke [15] reported similar rumpling on platinum-modified nickel aluminide coatings. Each thermal cycle in their experiment consisted of heating and cooling of a superalloy substrate coated with platinum-modified nickel aluminide BC from room temperature to 1200 °C with a hold time of 1 h. After about 100 such cycles, the surface rumbled with a wavelength of 30–50 μm and an amplitude of about 10–15 μm. On the other hand, Tolpygo and Clarke observed that when the same system was subjected to isothermal oxidation (100 h at 1150 °C), no surface rumpling occurred [15]. They also observed that a minor difference in the surface morphology of BC, results in significant difference in the stresses in the TGO at room temperature, but little quantitative differences in the rumpling amplitude and wavelength. Fast heating and cooling rates (1050 °C to 300 °C temperature change in about a minute) applied to BC–superalloy system along with mechanical loading of the superalloy was also found to result in BC rumpling [16,17] coupled with other thermal shock effects such as TGO formation and subsequent spalling or “scaloping” [16]. In the presence of ceramic coating, the BC roughening caused by cyclic oxidation can give rise to cracks at the TGO–ceramic interface. These cracks can then coalesce to form a crack sufficiently large to initiate delamination failure.

Rumpling type instabilities for isothermal and cyclic thermal treatments have been addressed in the past by Suo [18], He et al. [19] and Tolpygo and Clarke [15]. Suo [18], while addressing the issue of the initiation of TGO separation from metallic surfaces under isothermal conditions, assumed

that diffusion of the substrate atoms along the TGO–substrate interface drives the substrate to become wavy. The driving force behind the diffusion would be provided by the growth stresses in the TGO. This diffusion theory was based on the assumption of a uniform and constant TGO thickness, essentially ignoring the TGO growth kinetics. Note that the theory is applicable to any general metal–oxide system, implying that the presence of BC in BC–superalloy systems is unnecessary for rumpling to occur for comparable diffusivities of BC and superalloy surfaces. Suo's theoretical predictions of the characteristic substrate wavelength overestimated those observed in metal–TGO systems under isothermal conditions [20] by nearly an order of magnitude. Suo [18] attributed this discrepancy to the absence of the growth kinetics and stress generation in his model and the subsequent assumption of a constant TGO thickness. Thus, he concludes that diffusion of substrate due to growth stresses in the TGO can cause a substrate to become wavy with experimentally observed characteristic wavelengths, a few times the TGO thickness [20].

He et al. [19], using a numerical model, proposed that under certain circumstances, the growth and thermal mismatch stresses in the developing TGO (on metallic substrates) can give rise to the rumpling of the metallic substrate during thermal cycling. The numerical analysis they carried out was based on a purely mechanical effect of 'ratcheting', i.e. cyclic yielding of the substrate upon each thermal cycle. The ratcheting led to a monotonic increase or decrease in the initial undulation amplitude of the substrate under thermal cycling. Note that for ratcheting to occur, the substrate material has to have relatively low yield strength [11]. Further, this theory is also applicable to general metal–TGO systems, i.e., the presence of BC is not necessary for rumpling to occur, as long as the mechanical properties of the superalloy and the bond coat are comparable. One of the consequences of the ratcheting analysis is the establishment of a critical initial amplitude of TGO waviness below which ratcheting does not occur. For rumple wavelengths observed in literature [1,13–17], the estimated critical amplitude is more than about a micron.

Tolpygo and Clarke [15] have speculated that rumpling in BC–superalloy systems occurs due to diffusion of various constituents of the system perpendicular to the BC surface. According to this conjecture, Al depletion in the BC and the subsequent decomposition of the BC constituents at high temperatures cause rumpling. However, no experimental observations of rumpled material [1,13–17] reported any periodicity in the Al content in the BC that corresponds to the observed rumpling wavelengths.

Recent experimental work by Panat and Hsia [1] has shed some light in identifying the dominant mechanisms for bond coat rumpling. They found that Pt–Al coatings with vastly different initial surface morphologies rumple to a similar characteristic wavelength of about 60–100 μm and an amplitude of 2–5 μm after 25 thermal cycles. Each thermal cycle in their experiment consisted of heating the specimens from 200 $^{\circ}\text{C}$ to 1200 $^{\circ}\text{C}$ in about 55 min, a 50 min hold followed by air cooling. Fig. 2(a, b) show the result for a Pt–Al BC on a superalloy [Fig. 2(a)] and that for a bulk superalloy without BC [Fig. 2(b)] thermally cycled under identical conditions. The BC surface has clearly rumpled, but not the superalloy, in spite of the fact that TGO on the superalloy surface is slightly thicker than that on the BC surface. Note that in the case of bulk superalloy, the TGO stresses alone are expected to be dominant; while for the BC on the superalloy, the stresses in the BC and in the TGO would be important. Fig. 2 demonstrates that the presence of BC is necessary for the rumpling (the long wavelength waviness in Fig. 2(a)) to occur [1].

Further evidence of the importance of thermal mismatch stresses in the BC was found by examining the free edge of the specimen, Fig. 3. Fig. 3(a) shows the top view of a rumpled sample that has a random distribution of the surface undulations. At the free edge, however [Fig. 3(b)], the rumpling ridges are distinctly aligned perpendicular to the edge. As schematically shown in Fig. 3(c), the normal stress σ_{xx} vanishes at the edge. This stress component will gradually build up to the maximum over a distance of the order of the film thickness. In Fig. 3(b), both the TGO ($\approx 2 \mu\text{m}$ thick) and the BC ($\approx 100 \mu\text{m}$ thick) are stress free at the edge.

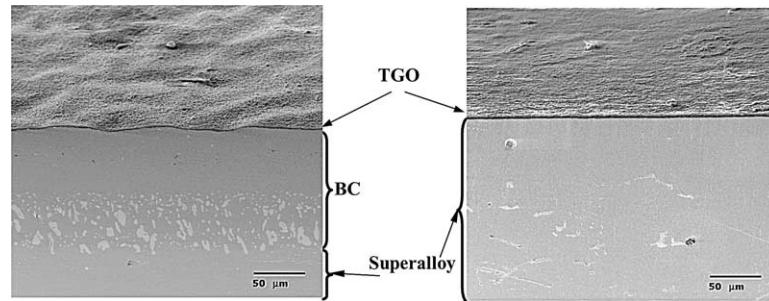


Fig. 2. Comparison of (a) a rumpled surface of the BC-substrate system with (b) an unrumpled surface of bulk superalloy upon identical thermal cycles (from Panat and Hsia [1]).

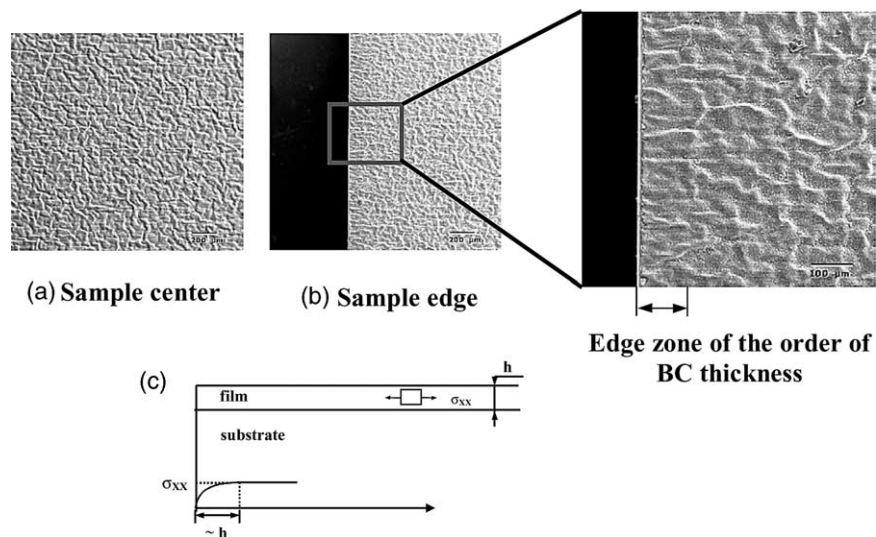


Fig. 3. Rumpling at the (a) specimen center and (b) at the specimen edge. (c) Expected variation of the residual stress in a thin film. (from Panat and Hsia [1]).

The extent of the ‘edge zone’ is, however, approximately equal to the BC thickness ($\approx 100 \mu\text{m}$). This result suggests that the bond coat stresses play a dominant role in the bond coat rumpling process.

Furthermore, Panat and Hsia [1] found that a very smooth initial BC surface with an undulation amplitude of about 25–50 nm rumpled to an undulation amplitude of about 2.5 μm and a characteristic wavelength of about 60–100 μm . According to the ratcheting model by He et al. [19], this initial amplitude is about two orders of magnitude smaller than the critical initial amplitude necessary for rumpling to occur.

These experimental results indicate that rum-

pling is not a generic phenomenon associated with metal-oxide systems, as would be expected from the ratcheting theory [19] and from Suo’s theory of TGO driven BC surface diffusion [18]. Furthermore, BC stresses play an important role in rumpling formation. In the present work, we analyze the rumpling process based on the premise that the BC stresses are the dominant factor during rumpling. Further, we analyze the rumpling process based on bond coat surface diffusion mechanism driven by the stresses in the bond coat. The results obtained from this analysis are compared with the experimental results in literature [1,13–17]. The effects of various input parameters on the rumpling

process are assessed. Finally, it is shown that the proposed mechanism of bond coat stress driven surface diffusion is a plausible mechanism for the BC rumpling process.

2. Governing equations

To study the bond coat stress driven surface diffusion mechanism, we employ the method used in the analysis of surface roughening in semiconductor thin films [21]. These films, while being processed at 600–700 °C, develop a characteristic roughness with a wavelength of hundreds of nanometers. This phenomenon was explained successfully by Freund and coworkers [22–25] by assuming that the observed surface morphology is a result of stress driven surface diffusion [18,26–30] of the thin film material along the film surface. In the present work, a similar approach is taken to study the phenomenon of BC surface rumpling.

We consider that the driving force for the BC surface diffusion is the thermal mismatch stress in the BC [1]. The diffusion at the BC–substrate interface is neglected since it is seen to maintain its shape during roughening of the free surface of the coating [1,13–17]. The thermodynamic quantity that drives the shape change of the BC surface is the chemical potential. The chemical potential, under certain conditions, changes only as a result of the change in the surface energy and the elastic strain energy of the system. We can define the chemical potential, χ , on the surface of the bond coat, as [22,24],

$$\chi = (U - \kappa U_s)\Omega \tag{1}$$

where κ is the BC surface curvature, Ω is the BC atomic volume, U is the elastic strain energy per unit volume in the BC, and U_s is the BC surface energy per unit area. Here U_s is taken to be a constant, γ . The variation in the chemical potential along the surface drives the diffusion of the surface atoms along the BC–TGO interface. The number of atoms passing a point on the surface per unit time per unit thickness, J , is proportional to the chemical potential gradient along the surface, and is given by the Nernst–Einstein relation [31] as,

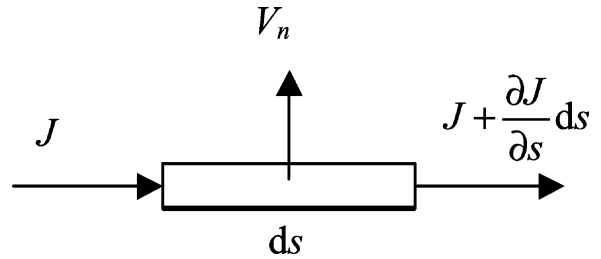


Fig. 4. Diffusion along surface element of the bond coat.

$$J = -\frac{D_s C_s \partial \chi}{kT \partial s} \tag{2}$$

where D_s is the surface diffusivity, C_s is the number of atoms per unit area, k is the Boltzman’s constant, and T is the absolute temperature. Note that D_s is a function of temperature [31] given by,

$$D_s = D_0 \exp(-q/kT) \tag{3}$$

where q is the activation energy and D_0 is the asymptotic value of diffusivity at very high temperatures. Conservation of mass at each point along the surface results in a velocity component of the local atoms normal to the surface, V_n (Fig. 4) given by,

$$V_n = -\Omega \frac{\partial J}{\partial s} = \frac{D_s C_s \Omega^2 \partial^2}{kT \partial s^2} [U - \kappa \gamma] \tag{4}$$

We now apply Eq. (4) to the problem of roughening of the BC on the superalloy. As stated before, the diffusion along the BC–substrate interface is assumed to be negligible as compared to the free surface diffusion. Thus, the coating can be modeled as a semi-infinite domain in the y direction, with thermal mismatch stress σ_T acting within the plane (Fig. 5). Note that σ_T changes with temperature, hence is a function of time in the present case of thermal cycling. This variation is given by,

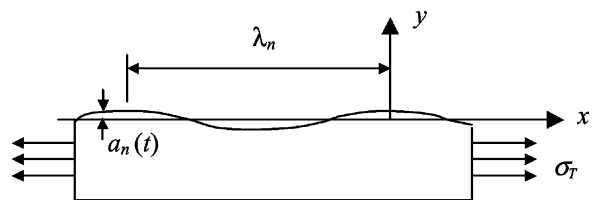


Fig. 5. Sinusoidal fluctuations of the free surface of BC.

$$\sigma_T = \alpha_m(T_p - T) \quad (5)$$

where α_m is a constant proportional to the thermal expansion mismatch between the BC and the substrate superalloy, and T_p is the coating processing temperature.

Assume that the initial bond coat surface is not perfectly flat. The evolution of the surface profile can be represented by a series of sinusoidal components such that,

$$y = h(x, t) = \sum_n a_n(t) \cos\left(\frac{2\pi x}{\lambda_n}\right) \quad (6)$$

where λ_n is the wavelength of the n th sinusoidal component and $a_n(t)$ is the time dependent amplitude corresponding to λ_n . It is assumed that the magnitude of each $a_n(t)$ is sufficiently small such that,

$$\frac{\partial}{\partial s} \approx \frac{\partial}{\partial x}, \quad \frac{dh(x,t)}{dx} < 1 \quad \text{and} \quad \kappa = \frac{\partial^2 h}{\partial x^2} \quad (7)$$

Substituting Eqs. (6) and (7) into Eq. (4), we get,

$$\frac{\partial h(x,t)}{\partial t} = \frac{D_s C_s \Omega^2}{kT} \frac{\partial^2}{\partial x^2} \left[U(x,t) - \gamma \frac{\partial^2 h(x,t)}{\partial x^2} \right] \quad (8)$$

To determine $U(x, t)$ on the coating surface, we need to find the variation of the stress σ_T due to slight sinusoidal fluctuations of the surface. This is derived by Gao [32], as,

$$U(x,t) = \frac{(1-\nu)\sigma_T^2}{4G} \left[1 - 8\pi \sum_n \frac{a_n(t)}{\lambda_n} \cos\left(\frac{2\pi x}{\lambda_n}\right) \right] \quad (9)$$

where ν and G are the Poisson's ratio and shear modulus of the coating material, respectively. Substituting Eqs. (3), (6) and (9) into Eq. (8), and noting the linear independence of each sinusoidal component in Eq. (6), we obtain a governing ordinary differential equation for rumpling of the bond coat surface,

$$\frac{da_n(t)}{dt} = \frac{\beta}{T\lambda_n^3} \exp(-q/kT) \left[\frac{(1-\nu)\sigma_T^2}{G} - \left(\frac{2\pi}{\lambda_n}\right)\gamma \right] a_n(t) \quad (10)$$

where

$$\beta = \frac{8\pi^3 D_0 C_s \Omega^2}{k} \quad (10a)$$

Solving Eq. (10) for given temperature history, we can evaluate the changing rate of the amplitude of an undulation with wavelength λ_n .

3. Case study

We consider particular cases of thermal cycling history applied to the BC–substrate system. In gas turbines, thermal histories experienced by TBC systems can vary widely according to the mode of application of the turbine. Systems used for propulsion and power peaking undergo multiple thermal cycles, while those used for power generation are subjected to very low frequency cycling [9]. Most of the BC rumpling experimental observations [1,13–17] are done with specimens subjected to multiple thermal cycles with heating and cooling times on the same order as the intermediate dwell times. In the present work, we consider two basic modes of thermal cycling history, triangular and sinusoidal. Rumpling behaviors for other complicated thermal histories can be deduced from these results.

3.1. Triangular temperature history

We consider the BC–substrate system experiencing triangular thermal cycles as shown in the insert in Fig. 6. The variation of the temperature in the film with time for the i th cycle is,

$$T = \begin{cases} T_{\min} + 2(T_{\max} - T_{\min})(\tau - i + 1) & i-1 < \tau \leq i-1/2 \\ T_{\min} + 2(T_{\max} - T_{\min})(i - \tau) & i-1/2 < \tau \leq i \end{cases} \quad (11)$$

where T_{\min} and T_{\max} are the minimum and the maximum temperatures, respectively, and $\tau = t/t_c$, where t_c is the period of the cycle.

Due to periodicity of the problem, we can analyze the evolution of the surface fluctuation amplitude for one cycle without loss of generality. It will be seen that surface evolution for further cycles can be deduced from the same result. To obtain a solution of Eq. (10), we take input parameters similar to that used in [1], with $T_{\min} = 473$ K, $T_{\max} = 1473$

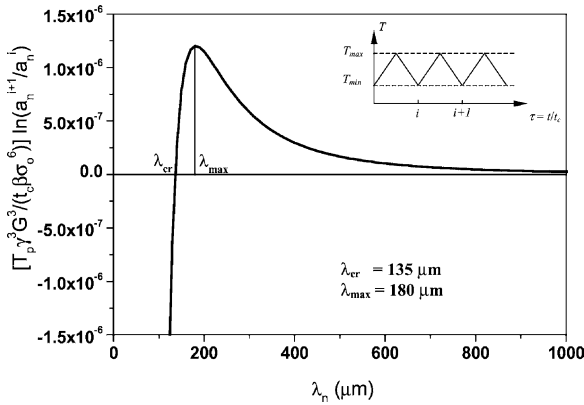


Fig. 6. Ratio of amplitude change after one thermal cycle as a function of wavelength for triangular temperature history.

K, and take the processing temperature $T_p=1273$ K. The BC properties are assumed to be, $\nu=1/3$, $\gamma=1$ J/m², $G=100$ GPa and $q/(kT_p)=5.8$ [24]. The mismatch stress in the BC, σ_T , varies with the temperature and depends on the thermal expansion mismatch between the BC and the superalloy. This residual stress is also likely to vary with different real systems and with time due to processing differences and relaxation. This mismatch stress will be the largest at room temperature. We denote the residual stress at room temperature by σ_o . In the present calculation, we take the BC stress at room temperature to be 450 MPa, a value used by Karlsson and Evans [11]. Note that the BC yield stress at room temperature is about 1000 MPa [11], similar to most superalloys. Solving Eq. (10) using numerical integration with these values, one has,

$$\ln\left(\frac{a_n^{i+1}}{a_n^i}\right) = \frac{t_c \beta \sigma_o^5 \hat{\lambda}_n^{-3}}{T_p \gamma^2 G^3} \left(M - \frac{N}{\hat{\lambda}_n}\right) \quad (12)$$

where a_n^i is the amplitude of the wave with wavelength λ_n after i th thermal cycle, $\hat{\lambda}_n = \lambda_n \sigma_o^2 / (\gamma G)$ is the dimensionless wavelength, and M and N are dimensionless quantities, dependent on T_{max} , T_{min} , and T_p and other BC properties. For the present case, $M=1.907 \times 10^{-1}$ and $N=5.187 \times 10^1$. The solution, Eq. (12) indicates that the amplitude for given temperature history and BC properties depends on the cycle period, wavelength, diffusivity, surface concentration of atomic sites, and the atomic volume. An interesting result from Eq. (12)

is that the ratio of the amplitude at the end of a thermal cycle to that at the beginning is independent of prior temperature histories, and depends only on the period (t_c) and the shape of temperature variation of the current thermal cycle.

The normalized amplitude growth ratio for one thermal cycle, $\ln(a_n^{i+1}/a_n^i)$, is plotted in Fig. 6 as a function of the wavelength, λ_n . The plot reveals different rumpling behaviors for fluctuations of different wavelengths. For components with wavelengths less than a critical value, $\lambda_{cr} = N\gamma G/M\sigma_o^2$, the amplitude decreases with each cycle. This value is about 135 μm in the present case. It can be seen that in this region ($\lambda_n < \lambda_{cr}$), the ratio of amplitude over each cycle decreases rapidly as the wavelength of waviness decreases. On the other hand, all components with wavelength greater than λ_{cr} increase in amplitude at varying relative rates per thermal cycle.

The result also shows that the maximum amplitude change occurs at a wavelength, $\lambda_{max} = 4N\gamma G/3M\sigma_o^2$, of about 180 μm in the present case. The components with wavelengths close to this peak point undergo rapid amplitude growth. The growth of these wavelengths can account for the observed rumpling of the BC surface as the number of cycles increases. These wavelength values are of the same order of magnitude as that observed in experiments [1,13–17]. Beyond λ_{max} , the rate of amplitude increase falls rapidly and becomes negligible for components with very long wavelengths.

3.2. Sinusoidal temperature history

We now consider a case of sinusoidal temperature history shown in the insert in Fig. 7. The temperature variation for the i th thermal cycle is given by,

$$T = T_{min} - (T_{max} - T_{min}) \sin[\pi(\tau - i)] \quad (13)$$

$$i-1 < \tau \leq i$$

Again, due to periodicity of the problem, one thermal cycle is analyzed for the evolution of $a(t)$. The surface evolution for further cycles can be deduced from the same result. For identical BC properties and the temperature range as before, the solution to Eq. (10) has the same form as Eq. (12) with $M=2.561 \times 10^{-1}$ and $N=8.089 \times 10^1$. A plot of

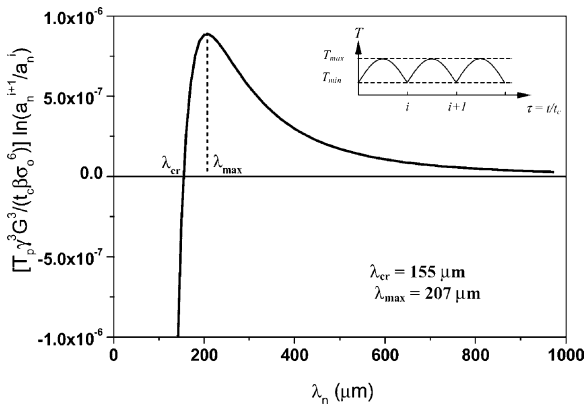


Fig. 7. Ratio of amplitude change after one thermal cycle as a function of wavelength for sinusoidal temperature history.

amplitude change as a function of wavelength in Fig. 7 shows the same trend as that for the triangular temperature history. The λ_{cr} and the λ_{max} for this case are 155 μm and 207 μm , respectively.

4. Discussion

The results presented here demonstrate that the analysis of BC surface rumpling based on stress driven surface diffusion can provide a plausible explanation for the experimental observations reported in literature [1,13–17]. According to the analysis, the amplitude of the initial surface waviness components with wavelength close to λ_{max} grow at high relative rates upon thermal cycling. These wavelength components will eventually dominate the surface features as the number of thermal cycles increases, and account for the observed nearly periodic rumpling behavior. This thermodynamic model also reveals a competition between the surface energy and the elastic strain energy in the system. The critical value of the wavelength, λ_{cr} , corresponds to a balance between these two energies.

Though the present analysis is for the BC–substrate system without the ceramic topcoat, it has strong implications in the TBC failure phenomenon. In presence of a TBC layer, rumpling of the BC surface may promote delamination between the TGO and the top ceramic coating and subsequent failure of TBC systems.

The present model is applicable to components with tensile as well as compressive BC stresses. Mumm et al. [33] have reported that the BC is under high tensile stress at room temperature. Note that in the analysis, the metallic BC layer is assumed to have negligible temperature gradient across the thickness during thermal cycling. In the case of strong temperature gradient through thickness of BC, the applicability of thermodynamics Eq. (1) must be re-examined.

Our parametric study reveals that the two most important parameters affecting the results are activation energy of the BC surface diffusion, q , and the thermal mismatch constant, α_m . The BC processing temperature, T_p , also affects the rumpling behavior. Note that α_m and T_p together determine σ_T , the residual stress in the BC as per Eq. (5). The activation energy for bulk diffusion is usually taken as a fraction of the chemical binding energy, and the activation energy for surface diffusion is a fraction of that for bulk diffusion. In the present analysis, the BC surface atoms are assumed to move along the BC–TGO interface which may increase the activation energy. On the other hand, q can decrease at high temperatures if defects on the BC–TGO interface interact with each other. To the authors' knowledge, q for BC materials has not been experimentally measured.

An important result of the present analysis is the prediction of λ_{max} , which can be easily compared with the dominant rumpling wavelength observed in experiments. The role of thermal mismatch, α_m , can be investigated in terms of the BC residual stress at room temperature, σ_0 . The possible values for σ_0 can vary from zero to the BC yield stress. Karlsson and Evans [11] have estimated the range of reasonable thermal mismatch values that correspond to a BC residual stress between 0 MPa and 450 MPa at room temperature. Fig. 8 shows the effects of variation of q and BC residual stress at room temperature on λ_{max} values for sinusoidal temperature history when $T_p=1273$ K. The values of activation energy, $q=0.1, 0.5, 1$ eV are considered, while the effect of a residual stress range of 0–1000 MPa at room temperature are investigated. Fig. 8 shows that for a BC residual stress higher than about 250 MPa at room temperature, the predicted wavelengths lie within those

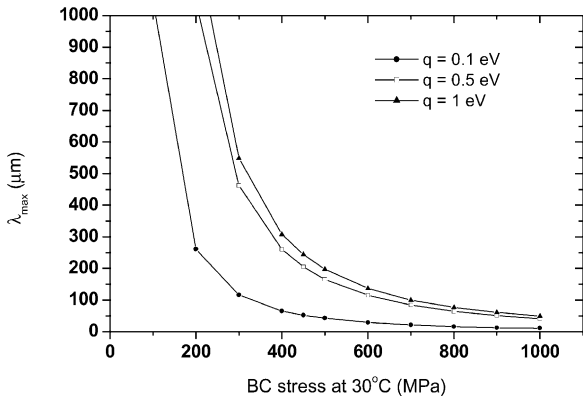


Fig. 8. Variation of λ_{\max} with q and the residual stress in the BC for sinusoidal temperature history.

observed in literature [1,13–17]. It can also be seen that at higher stresses, the sensitivity of the predicted wavelength to the activation energy decreases.

For fixed T_{\max} , T_{\min} , and a particular temperature history (e.g. sinusoidal as in Section 3.2), our analysis shows that the value of processing temperature, T_p , also affects the rumpling behavior. Such effect is shown in Fig. 9 in which λ_{cr} and λ_{\max} are plotted as a function of T_p for a sinusoidal temperature history with other input parameters identical to those in Section 3. Fig. 9 shows that, for a reasonable range of T_p between 900 °C and

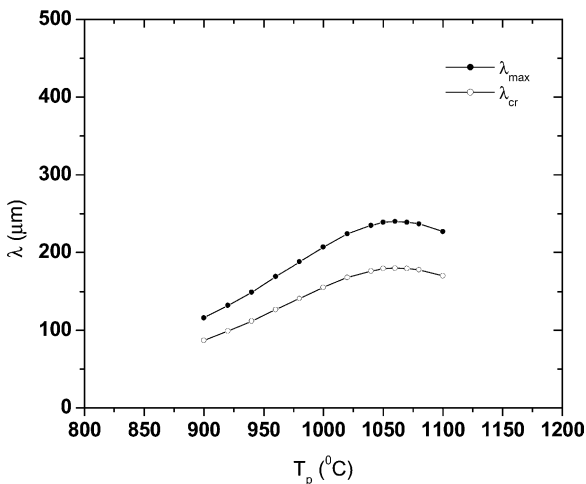


Fig. 9. Variation of λ_{\max} with T_p for sinusoidal temperature history.

1100 °C, the maximum wavelength λ_{\max} lies well within the range observed in experiments [1,13–17]. It is also interesting to note that the λ_{\max} versus T_p curve peaks at a value of T_p between T_{\max} and T_{\min} , indicating that the dominating rumpling wavelength will decrease both when T_p is approaching T_{\max} and when T_p is very low. A simple analysis of Eq. (10) confirms this trend. The result in Fig. 9 may potentially be used to estimate the processing temperature experimentally by measuring the dominant rumpling wavelength for different sets of T_{\max} and T_{\min} .

For isothermal temperature history ($T_{\max}=T_{\min}=T_{\text{isothermal}}$), we obtained a result similar to that by Freund et al. [22] with $\lambda_{\max} = 8\pi G\gamma/[3(1-\nu)\sigma_T^2]$. Fig. 10 shows the λ_{\max} values as a function of $(T_{\text{isothermal}}-T_p)$. The thermal expansion mismatch stress in the BC at temperatures near T_p is low, resulting in a high critical wavelength. For example, in case of $(T_{\text{isothermal}}-T_p) < 75$ °C, the critical wavelength is in millimeters. The figure also indicates that, contrary to the conclusion by He et al. [19], isothermal temperature history would result in BC rumpling with reasonable wavelength when $(T_{\text{isothermal}}-T_p)$ is large unless creep effects in the BC become dominant. In reality, long term exposure to high temperature may reduce the mismatch stress in the BC by relaxation and creep [34–37] thus resulting in a very

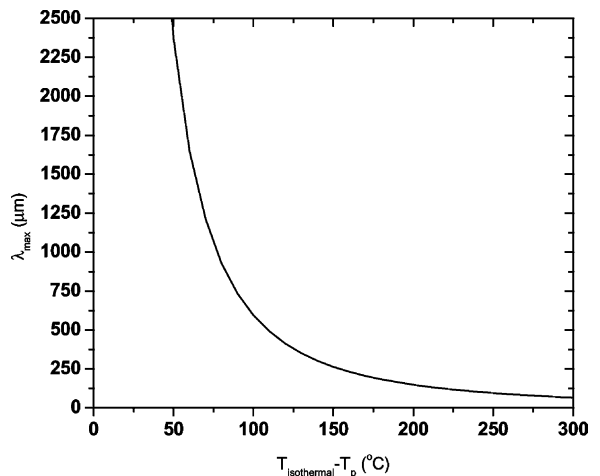


Fig. 10. Variation of λ_{\max} with $(T_{\text{isothermal}}-T_p)$ for isothermal temperature history.

long critical wavelength, making it impossible to observe the rumpling process. This could partly explain the lack of rumpling observed during isothermal oxidation of BC–substrate system [14,15]. Extensive isothermal experiments at various temperatures are required to determine the role of the temperature difference, $(T_{\text{isothermal}} - T_p)$, and the BC creep in the BC rumpling behavior.

A more typical temperature history in thermal cycling experiments [1,13–15] has a trapezoidal waveform with a substantial hold time at the maximum temperature between the heating and the cooling stages. Although the heating and the cooling stages can be approximated using the triangular waveform, the effect of holding needs to be considered separately. Fig. 10 shows that such effect strongly depends on the difference between the holding temperature and the processing temperature. Qualitatively, a small difference between T_{max} and T_p would tend to increase the dominant rumpling wavelength while large difference would tend to reduce the dominant rumpling wavelength.

The agreement between experiments and the analysis is rather good, in view of the assumptions used in the problem formulation. First, the BC is modeled as a homogeneous material, i.e., all the parameter values are taken to be identical everywhere. In reality, the BC has several phases across its thickness that evolve during thermal cycling. Secondly, the model does not take into account the effect of the TGO on the BC in the rumpling process. It is also worthwhile to mention the uncertainty involved in estimating D_s and C_s . According to Asaro and Tiller [38], D_0 can vary by as much as a factor of 10^5 depending on the environmental conditions.

The influence of the stresses in TGO on the BC surface rumpling is still not well understood. Tolpygo and Clarke [15] reported that significant differences in the levels of TGO stresses at room temperature result in no quantitative differences in the rumpling amplitude and wavelength. Suo's theory [18] of TGO stress driven BC surface diffusion, on the other hand, does not consider growth kinetics of TGO, which may be important in determining the stresses in TGO. Our experimental observations in [1] appear to indicate that the TGO stresses would be more likely to produce waviness

with a wavelength on the order of a few microns rather than that on the order of 100 microns. However, to fully understand the role of TGO, a comprehensive analysis taking into account of the growth kinetics of TGO as well as the TGO stresses must be done.

5. Conclusions

The present work gains an insight into the stress driven surface diffusion as a possible mechanism of the BC surface rumpling in the BC–superalloy systems upon thermal cycling. The analysis establishes the stress driven surface diffusion as a plausible mechanism of the bond coat rumpling observed in thermal barrier coatings upon thermal cycling. This analysis predicts continuous evolution of the bond coat surface with thermal cycling. Upon each cycle, it is shown that the amplitude of the surface fluctuations with wavelength higher than a critical value monotonically increases, while the amplitude of fluctuations with wavelength lower than the critical value monotonically decreases. The analysis also establishes a range of wavelengths that have a relatively high monotonic amplitude increase. This range of wavelengths accounts for the surface rumpling of the bond coat upon thermal cycling and is shown to be within the range of the experimental observations reported in literature. The analysis establishes parameters that influence the rumpling process. These parameters can be tailored to reduce rumpling and avoid its harmful effects on crack initiation and growth in TBCs.

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