Journal of the European Ceramic Society 37 (2017) 811–821

Contents lists available at www.sciencedirect.com

Journal of the European Ceramic Society

journal homepage: www.elsevier.com/locate/jeurceramsoc

Effect of neck formation on the sintering of air-plasma-sprayed thermal barrier coating system

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ARTICLE INFO

Article history:
Received 4 June 2016
Received in revised form 15 August 2016
Accepted 28 August 2016
Available online 9 September 2016

Keywords:
Sintering neck
Sintering stress
Young's modulus
Thermal conductivity
Thermal barrier coating

ABSTRACT

Sintering neck is a featured microstructure that may have significant effect on the sintering behaviour of air-plasma-sprayed thermal barrier coating system (APS TBCs). Based on experimental observations, a multi-necking wedge-shaped model for the sintering of APS TBCs was proposed by considering the sintering stress as surface tension and by employing the thermal-elasto-viscoplastic constitutive relation. Deformation pattern, stress distribution, sintering induced shrinkage, stiffening behaviour and temperature field were analysed by using finite element method. It is shown that the formation of sintering neck significantly affects thermal and mechanical properties related to sintering. Mechanisms of thermal and mechanical degradation induced by sintering were further elucidated.

1. Introduction

Thermal barrier coating system (TBCs) provides thermal protection and insulation to hot components of gas turbines. TBCs is mainly composed of three layers [1]: ceramic top-coat (TC), metal bond coat (BC) and superalloy substrate. During high temperature service, the TC of TBCs suffers a degradation process called sintering [2]. As a result, the elastic modulus of TC increases and its strain tolerance decreases, which promotes crack propagation and accelerates the failure [3]. Meanwhile the thermal conductivity also increases and weakens TBCs' thermal insulation ability. In view of these two important effects, sintering is attracting more and more attention [4–7]. Investigation of the microstructural change provides essential information of sintering induced degradation mentioned above.

For industrial gas turbine, air-plasma-spraying (APS) technology is widely adopted on account of process maturity and cost performance. The microstructure of TC in APS TBCs is lamellar determined by the spraying technique [8]. Based on experimental observations, the most significant microstructural change induced by the sintering process is the neck formation between splats [9]. Different from the sintering neck in powder metallurgy, the sintering neck studied herein is not the interconnection between powder particles but the interconnection between splats. Nevertheless, the physical essence remains the same that the neck formation is driven by surface energy in both cases.

A considerable amount of experiments on sintering neck of TBCs has been carried out. Siebert et al. [10], Vassen et al. [11] and Shinmi et al. [12] suggested that Young's modulus increases rapidly due to the necking between splats. Meanwhile, Rätzer-Scheibe et al. [13], Di Girolamo et al. [14] and Yu et al. [15] concluded from their experimental observations that sintering neck may enhance the heat flow from splat to splat, thus increasing thermal conductivity. Besides these two main effects, the formation of sintering neck changes the geometry of microstructure by bridging the crack faces [16] and by creating more contact sites [17]. It also increases the viscosity by raising the threshold load for the onset of splat sliding [18] and promotes main crack propagation despite the healing of some finer cracks [19]. In addition, attention is also devoted to the cracking of sintering neck [20]. Apart from these experimental investigations, there is limited theoretical and numerical work considering the effect of sintering neck formation and the details are still not clear. Based on a microstructure of parallel splats, Cipitria et al. [21,22] developed a sintering model, which is essentially a brick model. Although the brick model was able to predict shrinkage, surface area reduction, porosity reduction and even thermal conductivity, they ignored the fact that TBCs is rather viscous than elastic at high temperature during sintering. This fact might be the reason why the shrinkage predicted by the brick model was smaller than the

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http://dx.doi.org/10.1016/j.jeurceramsoc.2016.08.033

experimental result. On the other hand, it is difficult to consider the formation of sintering necks in the brick model. A similar model was adopted to simulate the heat flow by Golosnoy et al. [23]. Fleck et al. [24] established a multi-scale constitutive model assuming that splats were separated by penny-shaped cracks and crack faces sintered together at contacting asperities. These contacting asperities developed into sintering necks during thermal treatment. However, these asperities were treated as mechanical connections with elastic and plating (sintering) displacement. Consequently, the model was inapplicable to the analysis of stress distribution in sintering necks.

For the purpose of comprehensively understanding the role of neck formation in the sintering of TBCs, it is necessary to investigate the formation process of sintering neck and to evaluate its effect on shrinkage rate, Young's modulus, thermal conductivity, etc. Herein, experimental observation of sintering neck formation is stated in Section 2. A multi-necking wedge-shaped model developed on the basis of experimental observation, the constitutive equations and finite element analysis are presented in Section 3. Effects of neck formation on the shrinkage of TBCs, sintering stress, Young's modulus and thermal conductivity are discussed in Section 4. Conclusions are drawn in Section 5.

2. Experimental

Sintering neck formation was studied by experimental examination of the evolution of splat surfaces. Samples were prepared by plasma spraying system (GP-80, Jiujiang, China, 80 kW class) with fuse-crushed 8YSZ powder (Fujimi, Aichi, Japan). Details of spraying conditions can be found elsewhere [25]. Microstructure of as-sprayed samples was characterized by scanning electron microscope (SEM, TESCAN-MIRA3 LMH). The microstructure of APS TBCs is lamellar as a result of the spraying technique, as shown in Fig. 1(a). Observations at higher magnification indicated that intersplat voids are wedge-shaped rather than rectangular in most cases, as shown in Fig. 1(b).

Annealing treatment was then conducted at 1050 °C for 50 h. The evolution of microstructure in cross section was captured by high resolution transmission electron microscope (HRTEM, JEM-2100F). From a series of cross sectional TEM images, we clearly observed the evolution of surface morphology of splats in the lamellar microstructure. Splat surfaces of as-sprayed sample were quite smooth after ion milling process, as shown in Fig. 2(a). It is seen from Fig. 2(b) that surfaces become rough with multi-scale convex during annealing treatment. In sintering process, splat surfaces interconnected firstly in the vicinity of the corner of wedge-shaped pores, as shown in Fig. 2(c). The wedge-shaped geometry of intersplat voids facilitated neck formation at splat surfaces. Sintering process accelerated once sintering neck formed, leading to fast interface closure (Fig. 2(d)). More necks were observed at farther sites from the corner until fully sintered.

3. Formulation of the problem

3.1. A multi-necking wedge-shaped model for the sintering of TBCs

To characterize the problem, a multi-necking wedge-shaped model is developed on the basis of experimental observations, as shown in Fig. 3. In this model, the microstructure of APS TBCs is simplified as flat wedge-shaped splats with sintering necks formed between them. Detailed explanation of this model can be found in Section 3.3.

The variation of surface morphology is driven by surface energy and grain boundary energy [21] or interfacial energy [24]. These energies of yttria-stabilized zirconia (YSZ-8mol%) were summarized by Tsoga et al. [26] as linear functions of temperature T:

$$\begin{cases} \gamma_{sv}(\mathrm{Jm^{-2}}) = 1.927 - 0.428 \times 10^{-3}T \\ \gamma_{ss}(\mathrm{Jm^{-2}}) = 1.215 - 0.358 \times 10^{-3}T \end{cases} \quad (1),$$

where γ_{sv} and γ_{ss} are the surface energy and the grain-boundary energy, respectively.

This relation was obtained from experimental measurements over the temperature range 1573 K–1873 K but could be extrapolated to 1473 K–1973 K. As a consequence, the surface energy of 8YSZ-TBCs at 1200 °C can be estimated to be 1.3 J/m^2. This estimation has been validated by Fleck et al. [24]. In their study, the surface energy of zirconia was approximately equal to 1.0 J/m^2. The same value is taken as the surface energy of coatings herein. The changes of surface energy and grain-boundary energy finally lead to the change of free energy of TBCs. Herein, we assume that splat surfaces remain flat and the reduction of free energy is mainly contributed by the formation of sintering neck. Given that microstructural evolution is largely controlled by surface energy during the sintering process of TBCs, we substitute the change of surface energy by surface tension. From the viewpoint of mechanics, this surface tension can be treated as an equal-biaxial surface stress. Exerted on the splat surfaces in the multi-necking wedge-shaped model, this so-called sintering stress can be expressed in following equations:

$$\begin{cases} \tau_{zx}^{\mathrm{sinter}} = \sigma_0 \\ \tau_{zy}^{\mathrm{sinter}} = \sigma_0 \\ \sigma_{z}^{\mathrm{sinter}} = 0 \end{cases} \quad (2),$$

where σ_0 is assumed to be a constant value that guarantees the surface energy to be 1.0 J/m^2.

Note that each surface in the model has its own local coordinate system. In this way, the microstructure evolution of TBCs can be divided into three stages with different amount of sintering necks, i.e. 0 neck represents the initial as-sprayed state, 1 and 2 necks denote the early and late stages of sintering process, respectively. The change in interface morphology can be regarded as the formation of sintering necks.

3.2. Constitutive equations

To describe the microstructure evolution in sintering process, the constitutive relation should be developed. In our previous work, based on the theoretical derivation of relative density evolution, a modified thermal-elasto-viscoplastic constitutive model has been proposed and applied to the macroscopic sintering model of TBCs by considering the sintering stress as a volumetric force [27],

$$\{d\varepsilon\} = \{d\varepsilon^{\mathrm{E}}\} + \{d\varepsilon^{\mathrm{Vp}}\} + \{d\varepsilon^{\mathrm{sint}}\} + \{d(\alpha T)\} \quad (3)$$

where $d\varepsilon^{\mathrm{E}}$ represents the elastic strain increment, $d\varepsilon^{\mathrm{Vp}}$ denotes the viscous plastic strain increment, $d\varepsilon^{\mathrm{sint}}$ refers to the free sintering volumetric strain increment and $d(\alpha T)$ is the thermal strain increment.

The equivalent macroscopic sintering stress takes the form [28]:

$$\sigma_s = \rho^N \frac{2\gamma}{r^*}, \quad (4)$$

where ρ, γ and r^* are the relative density, surface tension and effective pore radius, respectively. N is a constant number determined by experiment.

Nevertheless, sintering stress in the microscopic model can not be treated as volumetric, seeing that the pores can not be simplified as uniformly distributed and they are not spherical. In this circumstance, the thermal-elasto-viscoplastic behaviour of TBCs can still