

# Effects of sintering and mixed oxide growth on the interface cracking of air-plasma-sprayed thermal barrier coating system at high temperature



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## ABSTRACT

Sintering and mixed oxide (MO) growth significantly affect the thermal and mechanical properties of thermal barrier coating system (TBCs) in gas turbine at high temperature. In this work, we numerically analyzed the effects of sintering and MO growth on the interface cracking of TBCs. A thermal-elasto-viscoplastic constitutive model was introduced, in which the effect of sintering was studied using a spherical shell model. Based on the same spherical shell model and our previous experimental observations, we theoretically derived the evolution of relative density and applied this constitutive model to the sintering of ceramic coating. The numerical results showed that viscosity, initial porosity of ceramic and the growth rate of MO had significant effects on interface cracking. In contrast, the influence of initial pore size of ceramic coating was neglectable. Suggestions were also made for the choice of material during TBCs design.

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## 1. Introduction

The increase of inlet gas temperature is one way to improve the performance of gas turbine. Thermal barrier coating system (TBCs) provides thermal protection and oxidation insulation for the elements working at high temperature. The air plasma sprayed (APS) technology is employed to manufacture the TBCs in the industrial gas turbine for power generation [1]. APS-TBCs is composed of three layers: the substrate, the bond-coat (BC), and the ceramic top coat (TC). High temperature sintering significantly affects the thermal and mechanical properties of TBCs, e.g., strain tolerance, durability and thermal insulation performance [2,3]. On the other hand, mixed oxide (MO) forms and grows at elevated temperature, which may result in the interfacial damage and speed up the debonding of coatings. Both sintering and MO growth affect the service life of TBCs.

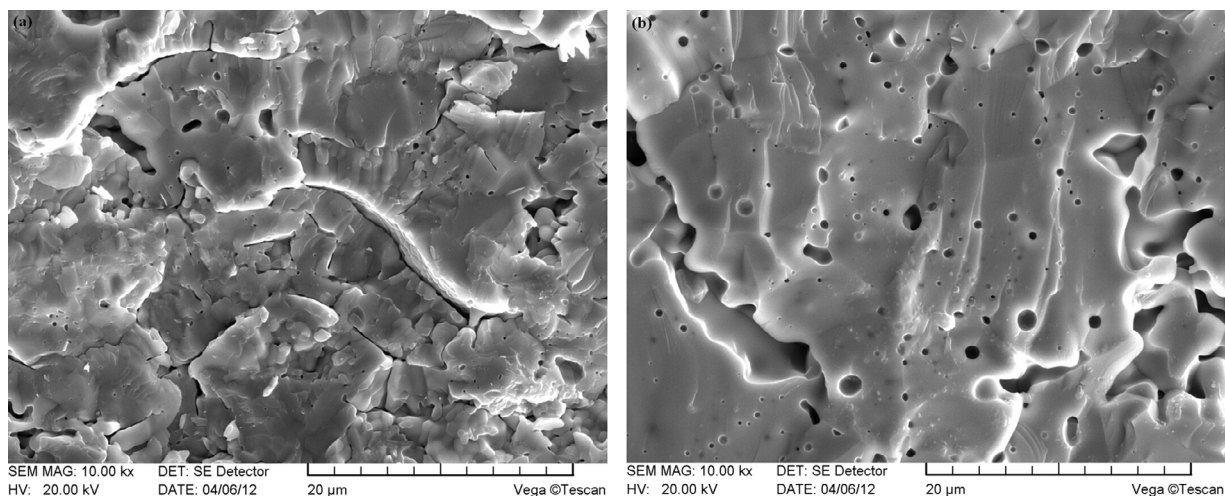
At elevated temperature, the sintering of TC is caused by the interconnection of ceramic splats driven by surface energy,

accompanying with macroscopic volumetric shrinkage. According to Coble's definition [4], sintering of solid can be divided into three stages depending on the change of pore shape. In the initial stage of sintering, neck growth leads to the increase of contact area of interparticle. The intermediate stage is characterized by the formation of equilibrium dihedral angles on the pore surface. In the final stage, pores are ultimately closed. Particularly in APS-TBCs [5,6], the first two stages are summed up to be a stage of inter-splat bonding improvement, which is dominated by surface diffusion and evaporation-condensation [7] and is relatively fast (<10 h). In the second quasi-stationary stage, macro-pore shrinks and tends to be more spherical. The later stage is dominated by sintering stress coming from surface energy [8,9] and is relatively long. Considering the long-term service of industrial gas turbine, sintering in the later stage is more crucial to the performance and lifetime of APS-TBCs, which attracts great interest in understanding the effect of sintering on APS-TBCs.

Theoretical and experimental investigations have been carried out to understand the effect of sintering. However, restricted by experimental conditions, limited parameters are considered in analysis, e.g. elastic modulus, thermal conductivity, diffusivity, porosity and impurity [6,10–12]. Besides, Ahrens et al. [13]

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**Fig. 1.** Cross-sectional microstructure of TBCs sintered at 1300 °C for (a) 50 h and (b) 500 h.

observed the time-dependent deformation behavior in a series of in situ three-point bending tests at different annealing temperatures. Based on variational principle, Cipitria et al. [14,15] proposed a sintering model and predicted the evolution of microstructure in TBCs. Fleck and Cocks [16] developed a constitutive model for TBCs incorporating the elastic response, local contact problem, sintering, diffusion and creep behaviors. They used a brick model for TBCs to study the sintering of splats [17]. However, the effect of sintering on the interface cracking has not been reported to the best of our knowledge.

MO growth is one of the major factors affecting interface cracking of TBCs. In the early service stage of TBCs, the thermally grown oxide (TGO) is mainly dense and uniform  $\alpha$ - $\text{Al}_2\text{O}_3$ . Then, MO (i.e. NiO,  $\text{Cr}_2\text{O}_3$  and spinel) forms. The main reason of MO formation is the depletion of aluminum [18] or the depletion of  $\text{Al}_2\text{O}_3$  [19]. The growth rate of MO is fast, typically three orders higher than that of  $\alpha$ - $\text{Al}_2\text{O}_3$ , and is approximately linear in time. More importantly, due to its porous and nonuniform features, MO may result in the concentration of local stress, interface damage and significant reduction of interface strength.

In the last decades, investigations concern the effect of TGO on the failure of TBCs, for example the effect of TGO on the stress distribution and interface morphology [20–26]. Also, attentions have been devoted to the effect of MO on the failure of TBCs [18,27]. Relatively, numerical analysis is not so much, e.g. Xu et al. [28] investigated the effects of MO spacing and its growth rate on interface cracking via finite element method, in which the effect of sintering is not considered.

The motivation of this work is to study the effects of sintering and MO growth on the interface cracking of TBCs at high temperature. In Section 2, the problem is stated, including constitutive models for sintering and MO growth. In Section 3, a finite element model is constructed based on typical TBCs structures, taking both sintering and MO growth into consideration. In Section 4, the effects of material and microstructural parameters are discussed in detail. Conclusions are drawn in Section 5.

## 2. Statement of the problem

### 2.1. Experimental observations

A detailed examination of the microstructure evolution was carried out for the comprehension of whole sintering process [29]. Coating material is 20 wt% yttria stabilized zirconia (YSZ)

powder (Xiandao, Yiyang, China) from 30  $\mu\text{m}$  to 70  $\mu\text{m}$  in size with a mean value of 47  $\mu\text{m}$ . Stainless steel substrates were employed for the deposition of YSZ coatings. We obtained the freestanding coatings by dissolving the substrate in hydrochloric acid. The 20 wt% YSZ coating underwent heat treatment at 1300 °C for different durations from 50 h to 500 h. Cross-sectional microstructures were analyzed by using scanning electron microscope (SEM) before and after high temperature exposure. Statistical data of apparent porosity of the coatings were obtained through image analysis. The apparent porosity decreased during sintering, especially in the early 50 h. For the purpose of revealing microstructural evolution, we examined the fractured cross-sectional morphologies of annealed coatings. The typical lamellar structure tended to disappear during sintering. Healing of intersplat microcracks and spheroidization of pores were observed during sintering, as shown in Fig. 1(a) and (b).

The effect of MO growth on the interfacial delamination was studied in our previous experiment [18]. TC was prepared by APS method with 8YSZ powder (Metco 204B-NS, Sulzer Metco Inc., New York, USA). BC was deposited by a cold spray system with NiCoCrAl-TaY powder (Amdry 997, Sulzer Metco Inc., New York, USA). The substrate was nickel-based superalloy (Mar-M247). As-sprayed TBCs went through heat treatment to introduce TGOs including MO to the interface between TC and BC. SEM (VEGA II-XMU, TESCAN, Czech) was employed for the observation of MO. The MO composed of spinel and NiO/ $\text{Cr}_2\text{O}_3$  presented at the interface exhibiting protrusion morphology due to much faster growth than  $\alpha$ - $\text{Al}_2\text{O}_3$ . The protrusion expanded and induced interfacial delamination with the growth of MO, as shown in Fig. 2.

### 2.2. Constitutive model for the sintering of TBCs

The TC of as-sprayed TBCs has a lamellar microstructure with random distributed pores and cracks. After heat treatment, the splats of TC sinter together accompanying with microcrack healing and pore spheroidization, which is confirmed by our experiment in Section 2.1 (see Fig. 1(a) and (b)). Therefore, the microstructure of sintered TBCs can be regarded as a homogeneous material with random spherical pores shown in Fig. 3(a). With the process of sintering, these pores tend to be homogenized (Fig. 3(b)). Keeping the same volume fraction, these cells can be equivalent to a representative volumetric element (RVE) of a spherical shell, as shown in Fig. 3(c), which has been proved applicable [30]. Based on the above simplification, we use a thermo-elasto-viscoplastic

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