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A comprehensive mechanism for the sintering of plasma-sprayed nanostructured thermal barrier coatings

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ABSTRACT

Nanostructured thermal barrier coatings (TBCs) are being widely researched for their superior thermal barrier effect and strain compliance. However, the sintering occurs inevitably in nanostructured TBCs that comprise both nanozones and lamellar zones, although the mechanism of sintering in such bimodal coatings is not yet clear. This study investigates the changes in microstructure and properties of nanostructured TBCs during thermal exposure with the aim to reveal the sintering mechanism operative in these coatings. Results show that the sintering process occurs in two stages. It was found that in the initial shorter stage (~0–10 h), the properties increased rapidly; moreover, this change was anisotropic. The main structural change was the significant healing of the intersplat pores through multiconnection. During the subsequent longer stage, the change in the properties was much smaller, where it was observed that the pores continued to heal, albeit at a much lower rate. Furthermore, the faster densification of the nanozones induced during sintering became significant, resulting in an opening at the interface between the nanozones and the lamellar zones. In brief, the pore healing at the lamellar zones affects the properties, especially in the initial stage. The presence of nanozones has a positive effect in that the performance degradation during the overall thermal exposure is slowed down. An understanding of this competing sintering mechanism would enable the structural tailoring of nanostructured TBCs in order to increase their thermal insulation and thermal cycling lifetime.

1. Introduction

Over the past few decades, thermal barrier coatings (TBCs) have been widely used in both aero and land-based gas turbines to protect their hot-section metallic components (e.g., combustion cans, blades and vanes) against high temperature. The TBCs significantly reduce heat transfer from the hot gas to the surface of metal alloy parts, and consequently higher operating temperatures can be employed resulting in notably improved efficiency of the gas turbines. A typical TBC system is a multilayer structure, consisting of a thermally-resistant top-coat and an oxidation resistant bond-coat applied over a metal substrate [1,2]. Owing to its unique porous structure and its excellent overall performance, plasma-sprayed yttria-stabilized zirconia (PS-YSZ) continues to dominate the processing application of the top-coat, especially in land-based gas turbines [3–5]. The lamellar structure of this top-coat with connected intersplat pores and intrasplat cracks results in a half more drop in the through-thickness thermal conductivity [6–10] and in the in-plane elastic modulus [5,11–13] with respect to the corresponding properties of the bulk YSZ material.

The continued requirement for increased gas turbine efficiency means that there is an urgent demand for the development of advanced TBCs with enhanced durability and resistance to degradation at higher operating temperatures. Consequently, doped-YSZ [14] and new candidate materials [15] for the application of top-coat are increasingly being investigated. Nanostructured materials are potentially attractive in view of the fact that the mechanical performance of these materials (hardness, strength, ductility and toughness) can be considerably enhanced if grain sizes are reduced from conventional microlevel to the nanoscale (i.e., < 100 nm) [16–18]. As a result, plasma sprayed nanostructured YSZ coatings have been widely studied and reported [19–22].

The achievement of the desired performance level of the nanostructured YSZ coatings depends crucially on their special microstructure that is inherent to plasma spraying. One of the biggest challenges is to retain the preexisting nanostructure of the feedstock [23]. Plasma spraying proceeds by the successive deposition of fully and partially molten particles on a substrate followed by lateral flattening, rapid solidification and cooling [24,25]. The fully molten nanostructured feedstock exhibits the traditional behavior of solidification, nucleation and growth [26], resulting in typical lamellar structure (lamellar zones)

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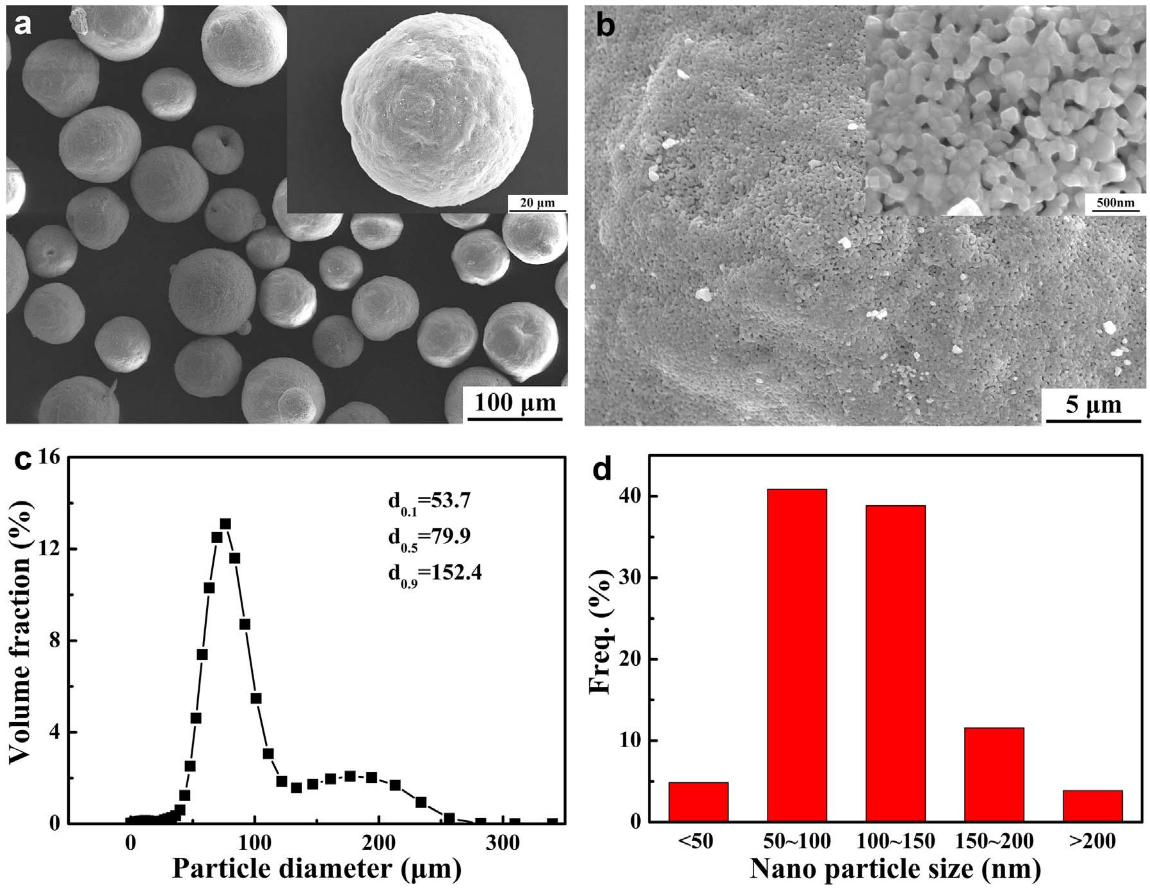

Fig. 1. Morphologies and size distributions of the agglomerates and the nanosized powders: (a) morphology of the agglomerates; (b) surface morphology of the agglomerates with nanosized powders; (c) size distribution of the agglomerates and (d) size distribution of the nanosized primary particles.

Table 1
Plasma spray parameters for the deposition of individual splats and coatings.

Parameters	Individual splats	Coatings
Plasma arc voltage / V	60	60
Plasma arc current / A	650	650
Flow rate of primary gas (Ar) / L min^{-1}	80	80
Flow rate of secondary gas (H_2) / L min^{-1}	16	16
Flow rate of powder feeding gas (N_2)/ L min^{-1}	4.5	4.5
Spray distance / mm	110	110
Torch traverse speed / mm s^{-1}	1000	500
Preheating temperature (°C)	300	/

[24,27]. However, this fully molten zone is often deleterious to the original nanostructure of the feedstock. Thus, the presence of the partially molten feedstock is necessary to translate the excellent physical properties of the nanostructured feedstock to the coatings. The fully molten zone facilitates sufficient particle adhesion and cohesion, and thus acts as a kind of matrix to the coating, whereas the semi-molten zone preserves the nanostructure. As a result, the integrity of the coating is maintained due to the fact that the semi-molten zones are surrounded by the fully molten zones, which act as binders. This morphology has been commonly described as a bimodal microstructure in previous reports [28,29]. In brief, in nanostructured YSZ coatings, the typical lamellar structure and the embedded-nanostructure coexist; in such a structure, the performance and the sintering behavior of the coating are intimately related to the presence of the conventional YSZ coating as well as to the nanostructured particles.

It is clear that the properties of the nanostructured YSZ coating are intrinsically associated with the degree of melting of the feedstock. Therefore, the coating performance is highly sensitive to the spraying parameters, which determine the melting degree. A key issue is to control the powder temperature in the thermal spray jet, which should be only slightly higher than the melting point of the material. Lima et al. [22,29] investigated the effect of spraying parameters on the percentage of the semi-molten zones. Moreover, the obtained bimodal structure depended to a great extent on the microhardness. The relatively lower hardness of the semi-molten zones contributed considerably to the excellent global mechanical properties. In addition to the spraying parameters, the powder size distribution also affects the resulting microstructure [23]. A broad particle size distribution, where a certain proportion of nanozones can be maintained, is more advantageous when compared to a narrow particle size distribution.

This optimal bimodal structure contributes to the superior reliability and lifetime of nanostructured TBCs with respect to conventional TBCs. It has been widely reported that nanostructured TBCs are potentially promising to achieve higher thermal cycling lifetime both for isothermal cycling [30–32] and gradient thermal cycling [33]. Regarding the sintering behavior of nanostructured TBCs, the faster densification in nanozones induced by sintering partially counteracts the degradation effect caused by pore-healing in the matrix zones [34,35]. The reason for this is that the nanozones have a higher surface area with a loose structure when compared to the matrix. The faster shrinking rate of the nanozones results in the formation of coarse pores at the boundary between the nanozone and the matrix. This mechanism is responsible mainly for the durable thermal insulating property during thermal exposure. However, there has been very little work carried out on the sintering behavior of nanostructured TBCs with bimodal structure.

The features of the bimodal nanostructured YSZ coatings in the as-deposited state can be summarized as follows: (i) the matrix is formed from the initially molten but subsequently solidified particles. The

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