Pressure infiltration of molten aluminum for densification of environmental barrier coatings

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Abstract: Environmental barrier coatings (EBCs) effectively protect the ceramic matrix composites (CMCs) from harsh engine environments, especially steam and molten salts. However, open pores inevitably formed during the deposition process provide the transport channels for oxidants and corrosives, and lead to premature failure of EBCs. This research work proposed a method of pressure infiltration densification which blocked these open pores in the coatings. These results showed that it was difficult for aluminum to infiltrate spontaneously, but with the increase of external gas pressure and internal vacuum simultaneously, the molten aluminum obviously moved forward, and finally stopped infiltrating at a depth of a specific geometry. Based on the wrinkled zigzag pore model, a mathematical relationship between the critical pressure with the infiltration depth and the pore intrinsic geometry was established. The infiltration results confirmed this relationship, indicating that for a given coating, a dense thick film can be obtained by adjusting the internal and external gas pressures to drive a melt infiltration.

Keywords: infiltration process; channel pore; densification; pressure assistant; environmental barrier coating (EBC)

1 Introduction

With an increasing demand for high operating temperature of new gas-turbine engines, SiC-based ceramic matrix composites (SiC-CMC) have attracted great interest for their high-temperature tolerance, superior mechanical properties, and low density [1–4]. In the engine combustion

environment containing hot steam or corrosives (such as sand and volcanic ash, generally known as molten salts, calcium–magnesium–alumino–silicate (CMAS)), SiC-CMC materials will undergo a severe degradation, especially accelerated oxidation caused by steam [5–11]. To prevent the deleterious environmental attack of SiC components, environmental barrier coatings (EBCs) were developed to cover the SiC surface. Candidate materials for EBCs should meet a set of requirements [4, 12–14], such as high-temperature phase stability, similar thermal expansion coefficient (CTE) to the matrix, chemical compatibility,

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steam stability, and CMAS corrosion resistance.

Ytterbium monosilicate (Yb_2SiO_5 , YbMS) was approved as a promising candidate for EBC due to its non-phase transition and low silica activity during high temperature service [15–18]. Nevertheless, a CTE mismatch between the top layer and the substrate is presented, which induced thermal stresses and the appearance of vertical pores or cracks [17–20]. These pores cross-linked together, forming complex networks [8,21,22]. Although these pores endow the coatings excellent stress tolerance, steam and other corrosives infiltrate from the openings and pass through these networks to reach the sub-layers and even the substrate, and react with Si bond layer, resulting in severe EBC oxidization and degradation. Over the past years, researchers have studied the accelerated oxidation of SiC in detail when being exposed to high-temperature steam, and found that the oxidation occurred at a parabolic rate while volatilization by a linear rate [23]. Under the protection of EBC, based on engine tests, the life of SiC components was improved by at least 3 times [24]. Nevertheless, for higher operating temperatures, the rate of oxidation and corrosion is further accelerated, especially when there are open penetration channels for steam and corrosives in the coating. Therefore, to obtain long operating life, a completely dense film without open-pores is of paramount importance.

The considerable efforts have been put into developing dense EBCs mainly from two aspects. One is concerning the optimization of suitable deposition techniques and deposition parameters. A lot of studies have tried various preparing techniques, such as sol–gel [25], slurry [26–28], chemical vapor deposition (CVD) [29], thermal spray techniques including air plasma spray (APS) [6,30–32], electron-beam physical vapor deposition (EB-PVD) [33], and plasma spray physical vapor deposition (PS-PVD) [34–37]. Among them, thermal spray technique is primarily used, which has a high deposition temperature and can completely melt the powder materials and form a dense coating. Modification of deposition parameters reduced the loss of Si and phase decomposition during the powder spraying [38,39], as well as was conducive to evaporate the species with high vapor pressure and form stoichiometric coatings [18]. The increase of substrate temperature could facilitate the crystallization of each layer [2,31,40]. The other is the post-densification treatment of porous EBC. Some researchers tried to eliminate vertical cracks through post-annealing treatment [32,40,41]. However, due to

the significant CTE mismatch of YbMS and SiC substrate, mud cracks with a wide average spacing appeared and penetrated the top and middle layers vertically [18]. Our group covered the surface of EBCs with an aluminum layer of several microns, and then reacted *in situ* to form a dense aluminum oxide layer of several hundred nanometers, which effectively improved the corrosion resistance of coatings [42]. Nevertheless, the dense film is too thin to protect the coating for a long time, and it is easily damaged by foreign objects in an engine environment.

This work proposed a densification method that blocked these open pores in the coating through pressure infiltration of molten aluminum. Tri-layer EBC of YbMS/mullite/Si system and the aluminum film were firstly produced by PS-PVD and magnetron sputtering techniques, respectively. Then the infiltration process is mainly investigated, especially the influence of gas pressure and intrinsic pore structure on the infiltration behavior. Four different pressure conditions were performed, including spontaneous and pressure-assistant infiltration. The results show that the infiltration results are determined by the pressure conditions and the pore geometry. According to the actual pore geometry, a zigzag pore model with multiple converging and diverging segments is established [21], and there is a mathematical relationship between the known geometry and the critical infiltration pressure. Once the critical pressure is exceeded, infiltration will occur successfully. The experimental results also confirmed this mathematic relationship, indicating that for a given system, only adjusting the gas pressures can achieve the deep infiltration of the melt, thereby forming a thick dense film.

2 Experimental

2.1 Coating preparation

Tri-layer system EBCs comprised of top layer YbMS, middle layer mullite ($3\text{Al}_2\text{O}_3\text{--}2\text{SiO}_2$), and bond layer Si, were deposited on SiC_f/SiC substrates, using PS-PVD facility (Sulzer-Metco Multicoat, Switzerland) assembled with an O3CP gun. The detailed deposition parameters are listed in Table 1. The thickness of each layer of YbMS, mullite, and Si is about 100, 50, and 50 μm , respectively. Afterward, the EBC coated CMC substrate was placed in a magnetron sputtering facility (J-1250, Jingzhou Industrial Coating, China) to deposit a metal aluminum

Table 1 Deposition parameter of tri-layer system EBCs prepared by PS-PVD technology

Material	Current (A)	Plasma gas (slpm)	Feed rate (g/min)	Stand-off distance (mm)	Pre-heating (°C)
Si	1650	Ar/He = 110/6	8.2	450	~500
Mullite	2600	Ar/He = 70/50	10	1000	~1000
Yb ₂ SiO ₅	2400	Ar/He = 100/20	20	1000	~800

film of ~5 μm onto the surface of EBCs. Pure Al target (99.99%) was used for the deposition, as well as the deposition parameters with a direct current of 3 A, a voltage of 150 V, and a vacuum of 5×10^{-3} Pa.

2.2 Infiltration process

Before vacuum infiltration, the Al-coated sample was sealed in a quartz tube with a vacuum as low as 10^{-3} Pa. The film was heat-treated in a high-temperature furnace at 750 °C for 2 h, and the temperature is slightly higher than the melting point of aluminum (about 660 °C) to fully melt and infiltrate the metal aluminum.

The pressure assistant infiltration was carried out using a two-step method. First, the film was heated to 750 °C at a rate of 4 °C/min in a vacuum furnace to fully melt the metal aluminum. In the second step, ultra-high purity argon ($\text{Ar} \geq 99.999\%$, $\text{O}_2 < 0.001$ ppm) at a certain pressure (2, 20, and 100 kPa) was backfilled into the furnace and held for 1 h to obtain a stable infiltration depth. For each condition, the assistant pressure was constant during the infiltration process.

2.3 Microstructural characterization

The surface of coatings and polished cross-section was characterized using the scanning electron microscope (SEM; SU6600, Hitachi, Japan) by secondary electrons. Elements maps were identified by energy-dispersive X-ray spectroscopy (EDS, Japan).

3 Results

3.1 Microstructure of coating system before melt infiltration

Before infiltration, an aluminum film was first prepared on the as-sprayed EBC surface. Figure 1 shows the microstructure of the multi-layer coating system before and after magnetron sputtering aluminum films. The obvious micro-cracks appear on the surface of as-sprayed tri-layer EBC in Fig. 1(a), which is caused by the rapid cooling of the molten particles in the PS-PVD facility. These cracks are harmful to the internal coating

because they provide infiltration channels for corrosives, such as water vapor, oxygen, and molten salts. Figure 1(b) indicates a crack-free surface covered by the aluminum film. The cross-section morphology of the hybrid coating is exhibited in Fig. 1(c). A dense film composed of closely arranged columns, with a thickness of about 5 μm , was evenly covered on the topcoat. It should be noted that this film also blocks the entrance of EBCs openings, which explains the difference between the two surface morphologies. Elemental mapping confirmed that the dense layer was aluminum. Therefore, the porous three-layer EBC surface, especially the exposed openings, was completely covered by the additional dense aluminum film.

3.2 Vacuum spontaneous infiltration results of aluminum into open pores

The vacuum infiltration is generally considered as one of the conditions for spontaneous infiltration. The results are shown in Fig. 2. The polished cross-section of the coating, as shown in Fig. 2(a), indicates that the top film remained at the pore entrance with little infiltration. The elemental maps around the pore entrance is marked with a blue rectangle, as shown in Figs. 2(b)–2(d). The aluminum distribution also confirmed its non-infiltration. During the infiltration process, both the external and internal pores are vacuum. Consequently, no external gas exerts pressure on the melt infiltration, and no internal gas hinders its infiltration. The infiltration is mainly driven by the surface tension of melt in the capillary channel. Due to the poor wettability of melt to the EBC material, the driving force is too small to press the melt into the pores. In addition, the complex pores or networks may be one of the powerful barrier to infiltration. Therefore, only driven by the surface tension of the melt, it is difficult to press the molten aluminum into the actual pores of the coating under vacuum condition.

3.3 Low-pressure infiltration results of aluminum into open pores

After the pore is vacuumed, a low-pressure protective

Fig. 1 Microstructure of multi-layer coatings before and after magnetron sputtering aluminum film: (a) as-prayed surface of EBC, (b, c) surface and cross-section microstructure after magnetron sputtering aluminum films, respectively, and (d) element mapping of aluminum of (c).

Fig. 2 Uninfiltrated pores in EBC under vacuum infiltration (also known as spontaneous infiltration) of molten aluminum: (a) cross-section view of the pore; (b, c, d) Al, O, and Si element maps of the blue rectangle area in (a), respectively.

atmosphere of 2 kPa was filled in the furnace, and the infiltration result was shown in Fig. 3. Figures 3(a)–3(c) display the different infiltration depths of molten

aluminum under the same pressure condition. A small amount of melt is filled near the pore entrance. Component analysis at points I and II exhibited the

Fig. 3 Near-entrance infiltrated pores under pressure-assisted infiltration at 2 kPa: (a, b, c) cross-section view of the infiltrated channel pores; (d) composition analysis of the points I and II in (a).

presence of aluminum, which confirmed that aluminum infiltrated and further oxidized to form alumina. In this case, the infiltration driving force not only includes the surface tension of the melt but also the external gas pressure, which results in a greater driving force and a deeper depth compared to that under vacuum conditions. It should be noted that the pore morphologies were diverse and extremely irregular, including size and shape, so the result of each pore was slightly different, but there was obvious infiltration. Consequently, the external gas is beneficial for pushing the melt to infiltrate; however, the depth is shallow as a result of the low assistant pressure.

To confirm the influence of external gas pressure on the infiltration depth, the external pressure was further increased to 20 kPa, as well as the same vacuum inside, and the infiltration results are shown in Fig. 4. Interestingly, despite being driven by the same pressure, completely different infiltration results were obtained in each channel pore stopped at different positions. We have listed three typical depths here, reaching the top layer (Fig. 4(a)), the middle layer (Fig. 4(b)), and the bottom layer (Fig. 4(c)). This work further investigated other seven pores, and the statistical results of the infiltration depths were shown in Fig. 4(d). For a better comparison, the infiltration depth was converted into dimensionless indexes based on the total depth of each

pore. It can be seen that the higher external gas pressure significantly increases the infiltration depth. The external and internal conditions of these pores are identical, but their intrinsic geometric characteristics are significantly different. It can be seen from each cross-section image of the pore that there are large geometric mutations, including expansion or contraction. This may be one of the main reasons for the extensive distribution range of infiltration depth, similar to Fig. 3. Therefore, the higher external gas pressure generally increases the infiltration depth significantly. But for some special pores, this kind of increase failed. The reason for this phenomenon will be discussed in detail below.

3.4 High-pressure infiltration results of aluminum into open pores

To weaken the influence of the special pore, a high assistant pressure of 100 kPa, which is close to atmospheric pressure and easily reached, was applied to the melt. The infiltration results of two different channel pores are shown in Fig. 5. One is regular pores with relatively parallel walls, and the other one is irregular pores with a series of converging and diverging geometries. The expansion angle of the pore wall is similar to that of Fig. 4. With the assistant of this high pressure, the melt undoubtedly filled every pore. The

Fig. 4 Partially infiltrated pores under pressure-assisted infiltration at 20 kPa: (a) infiltration stopped inside the top Yb_2SiO_5 layer, (b) infiltration stopped inside the middle mullite layer, (c) infiltration stopped inside the bottom Si layer, and (d) statistical infiltration depths of ten randomly selected pores.

Fig. 5 Fully infiltrated pores under pressure-assisted infiltration at 100 kPa: (a) regular pore with smooth and relatively parallel walls and (b) irregular pore with rough and zigzag walls.

result was completely different from the condition of 20 kPa, indicating that the surface tension induced by capillary pores no longer dominated, but was replaced by external pressure. Therefore, driven by the high external assistant pressure, the melt successfully passed through the structural barrier and reached the bottom of the pore.

4 Discussion

4.1 Determinant factors of infiltration depth in pores

According to the infiltration results under four different

conditions, the factors that determine the infiltration depth must include external pressure, as well as pore geometry [43–45]. In order to figure out how these parameters work, a mathematical model needs to be established. First of all, considering the irregular outline of these pores in actual environmental barrier coatings [18,21,22], this work established a wrinkle geometry that contains a series of converging and diverging segments, as shown in Fig. 6(a). Denote the pore length by H , the pore radius by r , and the pore wall inclining angle by α (half of the open angles marked in Fig. 6). Define the inclining angle α to be positive ($\alpha > 0$) in converging

Fig. 6 Infiltration model in zigzag channel pore: (a) geometry model of channel pore with series converging and diverging structures and (b) physical model of melt infiltration, including four pressures that external gas pressure (P_{ext}), internal gas pressure (P_{int}), capillary pressure (P_{cap}) induced by the curved surface of melt in microscale channels, and mass pressure (P_m) here is gravity.

segments and negative ($\alpha < 0$) in diverging segments. For a given structure, their geometric relationship between the pore outline and the depth is known, assuming the functional relationships are $r = S_r(h)$ and $\alpha = S_\alpha(h)$.

On the basis of the geometry model, this work selected an infiltration unit to analyze the pressures of the melt and to obtain the infiltration physical model, as shown in Fig. 6(b). Due to the poor wettability of molten aluminum to EBC materials, the direction of curvature of the melt surface in the diverging segment is opposite to the direction of the melt movement [43], which has also been explained in previous research [21]. Denote the infiltration depth by h and take the curved liquid surface in the diverging segment as the research object. There are mainly four kinds of pressures, which have been marked in Fig. 6(b): (1) P_{ext} : external gas pressure, whose direction is consistent with that of melt movement; (2) P_{cap} : capillary pressure, i.e., additional pressure below the bending liquid surface, whose direction is identical with the curvature center of the bending liquid surface; (3) P_{int} : internal gas pressure induced by the entrapped gas, whose direction is opposite with that of melt movement; (4) P_m : mass pressure, namely, gravity here. It should be emphasized that gravity in lateral pores was offset by the rigid pore walls, and therefore, this research mainly focuses on the vertical channel pore for its complexity in the physical force analysis of melt during infiltration. The branches of the channel pore can be considered as a combination of vertical and lateral pores, and the intersections can be regarded as a huge expansion and contraction.

Assume that:

(1) Ignore the chemical reaction between molten aluminum and coating materials, and thus the pores are of an invariantly stable shape.

(2) Treat the molten aluminum as a continuous, homogeneous, and incompressible Newtonian fluid.

(3) Treat the flow of the melt as a thermodynamic equilibrium.

As indicated by the experimental results, the maximum infiltration depth (h) depends on wetting properties of melt, intrinsic characteristics of pores, applied external pressure, and internal pressure, which can be described by the function that

$$h = f(\theta, S, P_{\text{ext}}, P_0) \quad (1)$$

where θ represents the contact angle of molten aluminum on the pore wall, which characterizes the wettability of melt, and S represents the functions of the geometrical parameters of pores, including pore wall inclining angle α and pore radius r , and P_0 represents the initial gas pressure induced by the gas trapped in pores by the melt before the infiltrating process.

The driving pressure during the infiltrating process is expressed as follows:

$$P = \frac{2\sigma \cos(\theta - S_\alpha(h))}{S_r(h)} + P_{\text{ext}} - P_0 \frac{\int_0^H S_r^2(h_3) dh_3}{\int_h^H S_r^2(h_2) dh_2} + f\rho h \quad (2)$$

where P represents the driving pressure, f represents the unit mass force, and ρ represents the melt density. Among them, the first term represents the capillary pressure, the second and the third terms represent the external gas pressure and the internal gas pressure, respectively, and the fourth term represents the mass pressure, which is gravity in this study.

After a sufficiently long infiltrating process, the melt

ceases and the infiltration depth reaches a maximum value ($h_{\max}$), which means that the system has reached an equilibrium state. Thus, the driving pressure on the melt surface is zero, and we have the equation:

$$\frac{2\sigma \cos(\theta - S_\alpha(h_{\max}))}{S_r(h_{\max})} + P_{\text{ext}} - P_0 \frac{\int_0^H S_r^2(h_3) dh_3}{\int_{h_{\max}}^H S_r^2(h_2) dh_2} + f\rho h_{\max} = 0 \quad (3)$$

Among them, the mass pressure (here expressed as gravity) is so small that can be ignored. Without considering the mass pressure, the mathematic model of vertical pore infiltration is equivalent to that of lateral pore infiltration. Then the expression can be simplified to

$$\frac{2\sigma \cos(\theta - S_\alpha(h_{\max}))}{S_r(h_{\max})} + P_{\text{ext}} - P_0 \frac{\int_0^H S_r^2(h_3) dh_3}{\int_{h_{\max}}^H S_r^2(h_2) dh_2} = 0 \quad (4)$$

There are four types of variables in the equation, namely, melt wettability, pore geometric parameters, external pressure, and internal pressure. Due to the phase decomposition of the coating materials in extremely heated plasma flame during the deposition process, the phase composition of the pore wall is not uniform [26,29]. Therefore, even in the same pore, the wettability of the melt on the pore wall is not a constant. According to the literature, aluminum poorly wets EBC materials, with a contact angle of slightly less than 90° . In practice, it is difficult to design good melt wettability on the surface of multiple materials at the same time. To facilitate the calculation and analysis, this research fixed the aluminum contact angle at 80° , focusing on the interaction of gas pressure and geometrical characteristics on the infiltration results.

4.2 Influence of gas pressure on infiltration depth

For pores of known geometry, in order to reach a certain infiltration depth h , there must be a critical pressure ($P_{\text{t-ext}}$) to overcome all barrier pressures. Gas pressure is an important factor affecting the maximum infiltration depth. The external gas pressure drives the infiltration. In theory, raising the external pressure may increase the infiltration depth. According to Eq. (4), the

expression of the critical pressure can be written as

$$P_{\text{t-ext}, P_0} = -\frac{2\sigma \cos(\theta - S_\alpha(h))}{S_r(h)} + P_0 \frac{\int_0^H S_r^2(h_3) dh_3}{\int_h^H S_r^2(h_2) dh_2} \quad (5)$$

When the internal pressure is minimized to vacuum, that is, the initial internal pressure $P_0 = 0$, the critical expression can be greatly simplified as follows:

$$P_{\text{t-ext, vacuum}} = -\frac{2\sigma \cos(\theta - S_\alpha(h))}{S_r(h)} \quad (6)$$

For a better understanding of the relationship between critical pressure, pore geometries, and internal pressure, specific values were assigned according to the actual range of each parameter. Among them, the entrance radius is $R_0 = 2 \mu\text{m}$, the pore depth is $H = 20 \mu\text{m}$, and the inclining angle of pore wall is $\alpha = \pm 10^\circ$ (the converging segment is 10° and the diverging segment is -10°). Thus, two curves of $P_{\text{t-ext}, P_0}$ and $P_{\text{t-ext, vacuum}}$ can be obtained by integrating Eqs. (5) and (6), as shown in Fig. 7. The blue line demonstrates the critical pressure $P_{\text{t-ext}, 100 \text{ kPa}}$ with the initial pressure to be 100 kPa, which is close to atmospheric pressure. The green line indicates the internal pressure evolution along with the melt infiltration. The pink line reflects the critical pressure $P_{\text{t-ext, vacuum}}$ under the condition of internal vacuum initially. It should be noted that the negative critical pressure means that the melt driving pressure is positive; in other words, the melt can spontaneously infiltrate without additional assistant. Since the internal pressure hindered infiltration increases exponentially, an excessively large critical external pressure is required to obtain deep infiltration. There is no obvious obstruction under the internal vacuum

Fig. 7 Relationship between critical external pressures ($P_{\text{t-ext}}$) and infiltration depth (H) under different initial internal pressures: the blue line represents the value of 100 kPa, which is based on Eq. (5) and the pink line represents the value of 0, corresponding to Eq. (6).

condition, so the critical pressure drops. A larger external gas pressure facilitates infiltration under both vacuum and non-vacuum conditions. According to the established pore model, the external pressure of 100 kPa exceeds most of the critical pressures during the vacuum infiltration process to reach a deeper depth.

To verify the influence of external pressures on the infiltration depths in practice, this work randomly selected ten vertical pores under each external pressure condition, and obtained the statistical results shown in Fig. 8. The definition of infiltration depth is the same as in Fig. 4(d). Under vacuum conditions, that is, the external gas pressure is 0, almost all pores are not infiltrated, which is consistent with Fig. 2. This indicates the poor wettability of molten aluminum to the surface of top materials Yb_2SiO_5 . Raising the external pressure to 2 kPa increased the infiltration depth to approximately 10%. Further increase of the pressure to 20 kPa increased the average depth to more than 50%. It is worth noting that the depth distribution was wide, and individual results were similar to that under 2 kPa, and individual pores had been full filled. Under 100 kPa high pressure infiltration, melt successfully filled these ten pores. The pressure is close to atmospheric pressure, and thus it is easy to implement. Part of the results is shown in Fig. 5. Therefore, the pressure of 100 kPa has exceeded the maximum critical pressure determined by these zigzag structures.

4.3 Influence of pore geometry on infiltration depth

The formation mechanism of irregular pore geometry,

Fig. 8 Improved infiltration depths by increasing external pressures, which is statistically known from the infiltration results of selected 10 pores under each external pressure.

including random width and zigzag profile, as shown in the pore cross-section images in Section 3, is related to the stress release during coating deposition and cooling process [10,14,30]. The material powder is melted into droplets in the high-temperature plasma flame. Then they have an impact on the cold substrate under the high-speed flame and rapidly cooling down to form splats. During the rapid cooling stress, the large quenching stress as high as several thousands of megapascal [46] will be generated in the coating, which further induced vertical pores or microcracks. The pores in individual splat are independent and stacked successively to form a coating. They are intersected with each other to form a complex structure. As shown in the schematic diagram in Fig. 9, the final pores or cracks are always considered to be a zigzag morphology. For the type I, although there is no lateral delamination, the melt still encounters several diverging structures when flowing in the pore at the intersections. In addition, there is usually lateral delamination in the pores shown as type II, which significantly enlarged the diverging structure including length, width, and opening angles.

For other random pores with unknown geometry, the expression of critical pressure also can be expressed as follows [47]:

$$P_{\text{t-ext, vacuum}} = -\frac{2\sigma \cos(\theta - \alpha)}{r} \quad (7)$$

The relationship between the critical pressure and geometric parameters including pore wall inclinations and pore radius during infiltrating is shown in Fig. 10(a). The four external assistant pressures of 0, 2, 20, and 100 kPa are drawn on the 3D surface as isobars, and also on the bottom project image by black, orange, blue, and pink curves, respectively. The negative pressures value in the 3D surface means that the infiltration does not require external assistance. A positive inclining angle representing the converging structure results in the negative critical pressure. It means that in a tapered segment, no matter how wide the channel is, the melt can infiltrate spontaneously without the assistance of external pressure. Conversely, a negative inclining angle corresponding to the diverging geometry results in a positive pressure value. The larger the diverging angle, and the smaller the radius, the greater the critical value. In other words, the wider the pores, the more sensitive to diverging. Therefore, the diverging segment of the zigzag structure hinders the infiltration of the melt, so the external pressure should be applied to push the melt.

Fig. 9 Schematic diagram of the formation of zigzag morphology with converging and diverging structures.

Fig. 10 (a) Critical pressure (P_{t-ext}) as a function of pore wall inclining angle (α) and pore radius (r), as well as four isobars of experimental pressures at 0, 2, 20, and 100 kPa and (b) the corresponding projection of the four isobars in (a), which divides the geometric parameters into five areas.

The minimum value of the hindrance angle depends on the wettability of melt, which is consistent with literature reports. According to the four assistant pressures in the infiltrating experiment, the pore geometric parameters can be divided into five regions, as seen in Fig. 10(b) and the project view in Fig. 10(a). The area on the right of each curve represents the geometries that can successfully infiltrate under the corresponding pressure, and the area on the left represents the failure of the pressure.

To prove the proposed relationship between the external pressure and the pore geometric parameters, the local enlarged views of four infiltrated pores under three pressure conditions are listed in Fig. 11. The four geometric parameters in front of the terminal liquid surface, including the radius and the inclination angles, were measured and marked in Figs. 11(a)–11(d). Each actual pore parameter was divided into corresponding

regions according to Fig. 10(b). The parameters in Figs. 11(a)–11(c) were all located in area IV, which was on the left of the isobar of 20 kPa, indicating that this pressure is not high enough to push the melt. When encountering these structures in Figs. 11(a)–11(c), the results of infiltration failure confirmed this conclusion. Similarly, the parameters in Fig. 11(d) were also located in area IV, but on the right of its assistant pressure of 100 kPa, indicating that the pressure was sufficient to promote the infiltration in these pores. It should be noted that all of these parameters were on the right of the 100 kPa isobar, and thus the melt successfully infiltrated into similar pores under this condition, which was confirmed by the previous results in Figs. 5 and 8. Therefore, raising the assistant pressure above the critical value can overcome the resistance caused by pore geometries and achieve deep infiltration.

Fig. 11 Infiltration interface of partially infiltrated pores at pressure of (a–c) 20 kPa and fully infiltrated pore at pressure of (d) 100 kPa.

The deep infiltration of the porous coating was achieved by adjusting the external and internal gas pressure in practice. The experimental steps and mechanism can be described in the schematic diagram of Fig. 12. First, a dense aluminum film is prepared on the surface of EBC to close the opening. Atmospheric gas is entrapped in the pores by the dense film. The sample is then placed in a high-temperature furnace combined with a vacuum pump. The sample is heated up above the melting point of aluminum and pumped into a high vacuum. At the same time, the air inside the pore is drawn out through the melt, and there is no pressure difference between the external and internal of the pore. The initial internal pressure before infiltration is close to zero. The infiltration occurred in the first two

steps is known as spontaneous infiltration. Finally, a certain pressure of almost oxygen-free argon is backfilled into the high-temperature furnace, which is equivalent to applying external pressure on the melt to drive the infiltration. A significant difference of pressure herein is achieved. The external assistant pressure is determined by the pore geometry, including radius and pore wall inclination. This means that under a certain external gas pressure, the geometric structure can determine the depth of infiltration.

In this way, the theoretical vacuum inside and pressure outside can be realized in practice. Although the pore geometry is an important factor of infiltration results, the deep infiltration can be obtained by controlling the external assistant pressures.

Fig. 12 Schematic diagram illustrating the steps of deep infiltration.

5 Conclusions

To improve the corrosion resistance of EBCs, this research is devoted to achieving the thick dense film by filling open-pores in coating with molten aluminum. A series of infiltration experiments were conducted under four different pressure conditions, from which a mathematic relationship was established between assistant pressure and pore intrinsic geometry according to the wrinkle pore geometry model. The detailed conclusions are as follows:

(1) The molten aluminum was difficult to spontaneously infiltrate in pores of EBCs without an external assistant.

(2) The melt infiltration occurred successfully with the assistance of external gas pressures. The distinct terminal depths were appeared under different pressure conditions, even on the same pressure condition, which was connected with pore intrinsic geometries.

(3) The critical assistant pressure to reach each depth mainly depended on pore geometry and internal gas pressure. Under internal vacuum condition, the critical pressure would augment with the increase of the diverging angle and the decrease of the pore radius.

(4) The infiltration results were in good agreement with the established mathematic model of infiltration. The deep infiltration was successfully achieved by raising external pressure above the critical value and simultaneously forming a vacuum inside the pores, thereby obtaining a thick dense film.

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