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## Regular Article

# Thermal shock resistance of alumina ceramics enhanced by nanostructured conformal coatings using metal–organic frameworks



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#### A R T I C L E I N F O

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#### 1. Introduction

Ceramics are widely used due to their excellent chemical stability and high melting point, but their lower resistance to thermal shock (a sudden increase of temperature) has been a long-standing problem [1–3]. In the past few decades, great efforts have been made to enhance the thermal stress resistance of ceramics. Nieto et al. improved the thermal shock resistance of alumina through the addition of submicronsized aluminum nitride particles into the substrate [4]. Wang et al. studied the thermal shock behavior of alumina-base ceramic substrate composite containing 5 vol.% of copper particles [5]. The improvement of the thermal shock resistance of these ceramic composites is due to the higher conductivity and toughening effect of the second phase particles. Another kind of method to enhance thermal shock resistance is by coating. Zhang et al. presented a sol-gel method for the fabrication of a hydrophobic YSZ (yttrium stabilized zirconia) coating with a mesoporous structure on ZrB<sub>2</sub> composite substrate [6]. However, the mismatch due to the thermal expansion coefficients between the coating materials and substrate severely induces cracking and spalling. Song et al. developed a method to increase the surface thermal resistance using biomimetically inspired nanofins, which were made by high temperature plasma etching and chemical corrosion. Although this method

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#### ABSTRACT

In this article we provide a large-scale and low-cost method, based on metal–organic frameworks, to fabricate nanostructured conformal coatings with the same material as the substrate. Such design can reduce the thermal mismatch between coatings and substrate and massively change the local thermal conductivity and interfacial heat transfer coefficient. Therefore, the thermal shock resistance can be enhanced to about 75%. We describe all aspects of such nanostructured conformal coatings from fabrication to characterization. Moreover, the mechanism of this enhancement due to high porosity and low pore size of nanoprous coating is discussed.

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significantly enhances the resistance of ceramics in water quenching conditions, the preparation process is, however, very complex and expensive, making it difficult for large-quantity fabrication and application. Thus, a simple and easy way to fabricate coating with the same thermal expansion coefficient (or simply, the same material) as the ceramic substrate is desired.

Metal–organic frameworks (MOFs) are materials constructed from organic ligands and metal cations [7–8], providing the low-cost and efficient way to fabricate the nanostructured coatings with the same materials as substrate [9]. The variety of metal ions and organic linkers affords an infinite possibility of MOFs structures [10] and shows the excellent ability to tune the nanoscale structures and properties [11]. Considering the crystalline porous structures and high surface areas of MOFs, they have been excellent candidates for many application such as gas storage [12–13], separation [14–15], drug delivery [16–17], catal-ysis [18–19] and sensors [20–22].

As the excellent porous materials, MIL-101 (MIL, Matérial Institut Lavoisier) consist of trimeric inorganic building blocks, formed by the assembly of three octahedra sharing a  $\mu_3$ -O common vertex. Also, they owe the unique characteristic to define the pore size, cell dimensions and surface area [23]. For instance, Liu et al. used MOF-5(Zn) as template for the synthesis of porous carbon, which exhibits a high specific surface area and excellent electrochemical property as an electrode material for electrochemical double-layered capacitor (EDLC) [24]. Xu et al. prepared spindle-like mesoporous  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> anode material prepared based on MOF template for high-rate lithium batteries [25]. Here we present a large-scale and low-cost method, based on MOFs, to fabricate



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the nanostructured conformal coatings with the same material as the substrate. Such design and fabrication can reduce the thermal mismatch between coatings and substrate and massively change the local thermal conductivity and interfacial heat transfer coefficient.

Alumina is one of the most widely used ceramics, and its crack behavior under thermal shock is always the hotspot of ceramics until now [26–28]. We adopt the aforementioned MIL-101(Al) as precursor to synthesize alumina coatings on  $Al_2O_3$  ceramic substrate to enhance the thermal shock resistance. The substrate is alumina ceramic (96%) with sizes of 50 mm × 20 mm × 1 mm, annealed at 1273 K. The properties of such material at room temperature are listed in Table 1.

#### 2. Fabrication of nanoporous coatings

(a) Preparation of solvothermal mother solution: terephthalic acid (TPA), anhydrous aluminum chloride (AlCl<sub>3</sub>) and N,Ndimethylformamide (DMF) were provided by Sinopharm Group Co., Ltd., China, DMF was used as solvent in precursor solution. Transparent solution was obtained by dissolving 1.2 g AlCl<sub>3</sub> and 2.04 g TPA in 120 mL DMF and by stirring for 10 min. All the regents used were in analytical degree without any further purification. (b) Synthesis of microporous γ-Al<sub>2</sub>O<sub>3</sub> membranes: the MOF-101(Al) films were synthesized through a solvothermal treatment. Firstly, Al<sub>2</sub>O<sub>3</sub> ceramic samples were immersed in 20% HCl solution for 30 min at room temperature. Then they were washed by high purity water and dried with hot air. Afterward, the ceramic samples were dispersed into a Teflon-lined autoclave with the resulting precursor solution. MOF particles grow on the surface of ceramic conformally after the solvothermal reaction at 333 K for 10 h. Calcination was conducted by slowly heating the ceramic samples to 1273 K (in 2 h) and cool them down spontaneously. The end products were obtained with 4 times repeat of solvothermal treatments and calcinations.

#### 3. Characterization

The morphologies and microstructure of as-prepare membranes were analyzed using scanning electron microscope (SEM, Quanta FEG 450) and high resolution transmission electron microscopy (HRTEM, Tecnai G2). The information about the crystalline phase of samples was obtained by X-ray powder diffraction (Bruker D8 ADVANCE X-ray diffractometer). Nitrogen adsorption and desorption measurements were performed to evaluate the Brunauer–Emmett–Teller (BET) surface area and pore size distribution of the porous  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>. MOF powder in the Teflon-lined autoclave was collected and heated with the ceramic samples to 1273 K for X-ray powder diffraction, high resolution transmission electron microscopy (HRTEM) test and nitrogen adsorption/desorption measurements.

The nanoporous coatings are conformally formed and tightly attached to the substrate with any geometric shape after solvothermal treatment and calcination. The nanostructured coatings on the substrate are investigated by scanning electronic microscopy (SEM) and transmission electronic microscopy (TEM), which primarily endows the specimens with excellent thermal shock resistance. Then the specimens are subjected to thermal shock and mechanical test, and results demonstrate the great enhancement of thermal shock resistance. Moreover, finite element simulation is conducted to investigate the mechanism of such nanostructures.

Usually, growing nanostructured MOF coatings from solvothermal mother solutions is based on the conventional solvothermal MOF synthesis process. The substrate is immersed into the mother solution during the MOF formation to get rigid deposition of the MOF particles on a substrate [29]. Fig. 1a shows the flowchart of fabrication of nanoporous  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> coatings on alumina ceramics. Nanostructured coating was synthesized by combination of a solvothermal reaction and calcination. HCl immersion was first carried out for the original ceramics before the solvothermal reaction, and the right-up inset figure shows the SEM image of such original surface. The pretreatment of the ceramic substrate before the MOF coatings growth plays an important role in the self-assembling process of MOF particles. In the solvent, MOF particles grow on the surface of ceramic and form highly ordered coating. The right-bottom inset shows the SEM image of such surface, which indicates MOF particles covered with the substrate. After calcination the MOF coating turned into nanoporous Al<sub>2</sub>O<sub>3</sub> coating with the same material as Al<sub>2</sub>O<sub>3</sub> substrate. The homogeneity of coating and substrate can eliminate the mismatching during thermal shock to provide high efficient thermal resistance without spalling of coating. Repeating the facile solvothermal and calcination process can control the thickness of Al<sub>2</sub>O<sub>3</sub> coating. Fig. 1b shows the SEM image of specimen surface with nanostructured coating after this procedure, which obviously indicates the nanopores distributing in coating from the MOF template. The nanopores are further investigated by TEM in latter. Fig. 1c shows the cross-sectional image and demonstrates the nanoporous  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> coating tightly bonding to the Al<sub>2</sub>O<sub>3</sub> substrate. Moreover, there are no micro-voids or cracks observed on interface, which implies the strong interface strength and may reduce the possibility of interfacial failure during thermal shock. After 4 times solvothermal treatment and calcination for each specimen, the average thickness of the  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> coating is about 4 µm. There are a few advantages of the proposed process method. The specimen was immersed into solution and then coatings fully covered all surfaces of specimen with regular or irregular geometry. Also, such method can naturally deal with the large-scale process with low cost to tune the nanostructures, which provides a powerful tool to control the performance of coatings. Moreover, the proposed method demonstrates the unique way to fabricating the coatings with the same material as substrate. According to the pore volume and density of  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> (3.9 g/cm<sup>3</sup>) we can easily get porosity  $\phi$  = 0.70.

The specimens were subjected to thermal shock test by water quenching. Thermal shock resistance can be characterized by the temperature and surface crack of specimen after quenching. Fig. 2 shows the water quenching results at different  $\Delta T$  (i.e. temperature difference of quenching) from 200 K to 800 K. "U" and "C" represents the uncoated and coated specimens in sub-figures, respectively. In order to verify the performance of coating, we compare the uncoated specimen (i.e. "U") to coated specimen (i.e. "C") at the same thermal shock temperature. Fig. 2a to h shows the serial of specimen surfaces after thermal shock with  $\Delta T$  from 200 K to 800 K, respectively. When  $\Delta T$  is 200 K, the surface cracks firstly occur for uncoated specimen but coated specimen has the smooth surface without any crack, as shown in Fig. 2a. Until  $\Delta T$  increases to 350 K, surface cracks firstly occur for coated specimens but the surface cracks of uncoated specimen become dendritic, as shown in Fig. 2c. In other words, R, the parameter of thermal shock resistance, is improved by 75% via the nanostructured coating. Obviously, surface cracks become denser and denser and their topography geometry changes from dendrite to network for both uncoated and coated specimens when  $\Delta T$  increases from 350 K to 600 K. When  $\Delta T$  reaches 700 K, the surface cracks evolve into tunnel cracks for uncoated specimen and such specimen breaks. Comparatively, the coated specimen keeps the surface cracks without fracture, shown in Fig. 2g. Even if  $\Delta T$  increases to 800 K, the coated specimen still keep the surface cracks and coatings are tightly bonded to substrate. However, the uncoated specimen spalls into pieces at such temperature. Such comparison indicates clearly that

Table 1

| Properties | Density                | Young's modulus | Thermal conductivity | Thermal expansion | Heat capacity |
|------------|------------------------|-----------------|----------------------|-------------------|---------------|
| Value      | 3.80 g/cm <sup>3</sup> | 315 GPa         | 20 W/m-K             | 6.7 μ/K           | 890 J/kg-K    |

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