Ceramics International xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Ceramics International

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

Heat transfer in high porous alumina: Experimental data interpretation by different modelling approaches

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ARTICLE INFO

Keywords:
Alumina
Modelling
Pore size distribution
Porous materials
Thermal conductivity

ABSTRACT

Advanced porous ceramics are a remarkable class of materials with important applications in engineering fields. Porosity features have received wide attention for their capability to influence all properties. In this paper, the correlation between pore structure and heat transfer has been studied. Different analytical procedures found in literature as well as an Intermingled Fractal Units' model are proposed. Models predictions are compared with experimental data. It has been observed that IFU is particularly suitable to predict thermal conductivity values very close to experimental ones. This fact is related to its capability to replicate porous microstructures in terms of pore volume fraction, pore size range and pore size distribution.

1. Introduction

Advanced ceramics characterised by high porosity have received wide attention for their unique combination of favourable properties such as low density, high elastic properties, high surface area, very good fluids filtering control, optimum chemical stability, excellent oxidation resistance, low dielectric constant and low thermal conductivity. These features candidate high porous ceramics for both structural and functional applications related to environment, energy and mass transport, health, etc. [1]. In this panorama, particular attention is drawn to heat transfer properties. Indeed, in spite of the presence of a porous structure, mechanical resistance decreases which remarkably increases thermal insulation properties [2–5]. The best compromise between mechanical and thermal properties represents the starting point for numerous engineering applications of porous alumina ceramics as thermal barrier coatings. It consists of an insulating layer applied on metallic surface. This thermal protection is fundamental for components subjected to extreme thermal conditions, which can influence their mechanical resistance (for example: gas turbine and aero engine parts) [6]. Although numerous studies are carried out since 1960s, still their efficiency improvements remain an important issue of material science and technology applications.

Different materials are studied for thermal barrier applications; YSZ, LZ, LZ_7C_3 [4,7–10]. One of the most promising is represented by high porous alumina, which can be obtained by different techniques such as (*a*) direct foaming method, (*b*) the sacrificial template method, (*c*) the gel-casting method and (*d*) pore forming agent method. (*a*) Direct foaming method starts incorporating air into a suspension or liquid media, which is subsequently set in order to keep the structure of air bubbles created. In most cases, the consolidated foams are afterwards sintered at high temperatures to obtain high-strength porous ceramics. (*b*) The sacrificial template method consists of fabrication of a biphasic composite based on ceramic or ceramic precursor and a sacrificial material, which is ultimately extracted (during drying, pyrolysis, evaporation or sintering) giving rise to conversion of a porous microstructure into the materials matrix. (*c*) The gel-casting method is based on mixing ceramic power with a monomer solution. Polymerisation generates a chemically cross linked gel, which fixes ceramic particles. The gel-cast green body is then removed from the mould followed by drying, thermal removal of the organic matrix and finally sintered. (*d*) Pore forming agent method is based on the mixture of ceramics and combustible substances, which are burnt out originating a porous microstructure [1,11,12].

Porous microstructures, obtained by described techniques, remarkably influence different properties of advanced ceramics. For thermal barrier coatings, particularly interesting is the influence of porosity on heat transfer. Obviously, bulk materials have a superior ability to transfer heat. For this reason, the introduction of air (low heat transmission degree) trapped into microstructure allows modifying thermal conductivity values. On one hand thermal conductivity (k_{eff}) is influenced by solid (k_s, different solid phases could be present) and fluid phase of material's microstructures (k_f, different fluid phases could be present in the pore structure); but on the other hand, in order to adequately understand this phenomena, it is important not to neglect pores fraction of the voids (ε), pore size distribution and random level of microstructure [16,17].

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http://dx.doi.org/10.1016/j.ceramint.2017.04.071
Received 8 March 2017; Received in revised form 9 April 2017; Accepted 10 April 2017

Moreover, aspects which regard morphological features (pore size distribution and random level of microstructure) [20,21] still are debated and represent an important research field. The difficulties in adequately defining their influence on thermal properties are related to the complexity of describing them in modelling terms. Moreover, the final results of obtained ceramics' microstructures are often very hard to control because of high number of variable during production process.

Different models have been proposed in order to describe porous microstructures and to calculate porosity influence on thermal conductivity. However, these approaches based on solid and fluid phases' proportions or standard distributions are validated only for specific materials and their general values must be still demonstrated.

In this last decade, it has been noted that a promising road in microstructure modelling could be achieved by applying Fractal Geometry.

This fact is probably due to the complexity of managing it during the production process, but also to the difficulty of describing and characterising it from a geometrical point of view. Indeed, Euclidean geometry is not often suitable to reproduce complex shapes existing in nature. For this reason, the study and application of the basis related to Fractal Geometry are of great interest.

Fractal Geometry has been formalised and developed all over the 1970s by Benoit Mandelbrot. The starting point is represented by the observation that every natural shape, if carefully studied, exhibits very complex characteristics. Contrary to Euclidean descriptions, mountains are not cones, clouds are not spheres and surfaces are not smooth [13]. Moreover, this complexity characterised the high number of distinct inspection scales.

The base concept of Fractal Geometry is the existence of figures with non-integer dimension (fractal dimension, D_f). A typical example is represented by a mono-dimensional element as a thread (in geometry: line), which rolling up generates a ball. Is this a three-dimensional or mono-dimensional new figure? Fractal Geometry responds that this generated figure has a proper dimension with non-integer values. The geometrical figures related to this innovative geometry are called fractals, from *Latin* word, which means irregular, indented, etc. Fractals are constructed by an iteration process, which consists of reproducing the same configuration at different scales. This property is known as *self-similarity* and is fundamental aspect in modelling approaches particularly for microstructures reproduction.

For these reasons, fractal analytical procedures have been proposed for describing different properties of materials [14–16]. In this sense, Cai et al. carried out significant scientific works in which they presented an innovative analytical fractal model constructed taking into account pore sizes, shapes and tortuosity. Their calculations show that this model may describe the imbibition process in different, natural or artificial, porous materials [17–20]. Recently, a special attention has been reserved to nanoparticles properties, aggregation and convective heat transfer of nanofluids [21]. It has been shown that fractal geometry is capable to describe nanoparticle size distribution which influenced thermal conductivity values [21].

Yu et al. proposed a model based on tortuosity and pore-area fractal dimension, phase fractal dimension, saturation and micro-structural parameters in order to predict fluid flow into porous media [22]. Fractal models have been also proposed to study elastic properties of porous ceramics materials [23] as well as heat transfer. For example, Fan et al. [24] proposed a fractal model for describing thermal conductivity values of wood fibred based materials. The predictions calculated are in agreement with experimental data. A finite volume method has been applied by Huai et al. [25] on fractal figures. Their elaborations show that pore fraction is the most important factor capable of conditioning effective thermal conductivity. However, they indicated that the spatial distribution of pores, particularly big pores, could influence substantially the final results. Huang et al. carried out a research on the thermal conductivity of salt/expanded graphite composite material [26]. They formalised a theoretical model to calculate thermal conductivity values of this three-phase medium. The predictions indicated that these data better fit experimental results than traditional model used for these materials [26].

In spite of these efforts, modelling approaches are often not generalisable for disordered materials. Indeed, random structure, owed to a relevant pore size distribution, influences thermal conductivity property. Recently, an intermingled fractal units' model has been proposed in order to reproduce porous materials' microstructures and to predict their different properties as mechanical, fluid circulation and thermal. The obtained results are in good agreement with experimental ones [27,28].

In this work, an application of IFU procedure to high porous alumina ceramics is shown. The results are compared with experimental data and calculations from different models proposed in literature for porous materials.

2. Materials and methods

Two different porous alumina systems have been considered for this work. The first group of samples, as indicated in [29], has been fabricated starting from tabular alumina (≤0.074 mm and ≤0.045 mm, 98% Al_2O_3); α-Al_2O_3 (2 μm, 99% Al_2O_3); carbon black (CB, N774, 50–100 nm), ρ-Al_2O_3 (5 μm, 85% Al_2O_3) and aluminium powder (≤0.045 mm, 98% Al,) were used as starting materials, with commercially-available poly vinyl alcohol solution (PVA, liquid, the specific gravity is $1.02 g/cm^3$) as a binder.

Production steps can be summarised as follows: a) wet-milling for 3 h at rotating rate of 300 rpm; b), the obtained mixtures were mixed with 8 wt% PVA solution for 30 min; c), the results were dried at 110 °C for 24 h; d), the mixtures were divided in specimens, dimensions of 25×25×140 mm^3, and pressed at 5 MPa. Synthetisation has been carried out at 1550 °C for 3 h.

The second group of ceramics' materials, as indicated in [30], has been obtained from Al_2O_3 powder with average diameter equal to 1 μm. The fabrication method was gel-casting process and Tert-butyl alcohol (TBA) and wheat granular starch (particle size 10 μm) were used as pore forming agents and binders. The summarised gel-casting technique used consists of preparing a liquid suspension, moulding, drying, binder removal and sintering at 1450 °C for 2 h.

Pore size distribution was analysed by mercury intrusion porosimetry (AutoPore-IV9510). Thermal conductivity at room temperature was measured on 5×5×3 mm^3 machined specimens, using Thermal Transport Option (TTO, PPMS, Model 6000).

3. Intermingled Fractal Units' Model to predict thermal conductivity

Geometrical and morphological reconstruction of porous microstructures represents an important tool for understanding physical properties of specific materials. The Intermingled Fractal Units' model (IFU) is demonstrated to be capable of replicating experimental structures. The base units of this model are the well-known Sierpinski carpet constructed following different and possible schemes. In general, this fractal is obtained starting from a square subdivided into nine sub-squares. Some of them are subtracted creating a bi-modal frame, solid and voids. Gradually, iteration (i) process repeats the same configuration on remained sub-squares. At the end, it is possible to observe a geometrical figure, which has a peculiar voids distribution. Fig. 1a and b reports a representation of Sieprinski (i=0, 1, 2, 3) carpet with two and three voids at first iteration. Consequent voids' distribution is typical of fractal figures, with the maximum value of frequency in association to maximum size of void (the bigger sub-square subtracted). Experimental pore size distributions are often very different by fractal distribution and might present one or more peaks along voids size range. This fact could represent serious limitation,