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Porosity and pore size distribution influence on thermal conductivity of yttria-stabilized zirconia: Experimental findings and model predictions

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Abstract

Porous yittria-stabilized zirconia is an important advanced ceramic material for technological applications. One of the most important characteristics of this material is low thermal conductivity, which is greatly influenced by the presence of pores into the microstructure. In fact, air trapped in the pores represents a better thermal insulator. The role of the pore volume fraction on porous material characteristics has been extensively studied. On the other hand, the influence of the structure disorder, the pore size range and pore size distribution have been studied much less. In this study, an intermingled fractal model capable of relating thermal properties of ceramic materials and their pore microstructure has been proposed. Model predictions are found confirming the experimental data fairly well, even better than the others models available in the literature.

Keywords: Advanced porous ceramics; Fractal model; Thermal conductivity predictions; Pore size distribution; Yttria-stabilized zirconia

1. Introduction

Advanced porous ceramics have been widely studied for their various technological applications; filters and membranes [1], fuel cell electrodes [2], catalyst support for biomaterials [3], piezo-electric materials [4], acoustically and thermally insulating bulk media [5,6].

Various fabrication methods of porous ceramics have been performed, including the pore-forming agent technique [7], infiltration of ceramic sol into template structures [8], gel-casting process [9], slip-casting [10], starch consolidation [11], microwave processing [12], electrophoretic deposition [13], and freeze casting [14].

Recently, research has been targeted to Yittria-stabilized zirconia (YSZ). It reveals good thermo-mechanical stability, high ionic conductivity and compatibility with electrode materials, hardness, chemical inertness, corrosion and wear resistance, low density, thermal shock resistance and unique thermal insulation properties [15–17]. Owing to these peculiarities, it is becoming essential for several applications, wherein YSZ offers several distinct advantages over other materials such as polymers or metals [18].

Fully stabilized zirconia as well as partially stabilized zirconia offers low thermal conductivity, but the later has higher mechanical properties that make it appropriate for various engineering applications [19].

Moreover, thermal conductivity of YSZ further decreases introducing pores into the microstructure, because air trapped in the pores represents a better thermal insulator. Owing to this reason, fabrication of porous zirconia ceramics with a high porosity is very essential for its engineering applications in bulk thermal isolators as well as thermal barrier coatings [20,21]. Since 1960s, the protection of gas turbines against degradation by high temperature oxidation processes and hot corrosion has being studied [22].

For these materials, thermal conductivity is a basic characteristic for classifying them into different branches of

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applications. In this regard, very important factor requiring explanation is that the effective thermal conductivity (k_{eff}) of porous materials depends on following factors:

(a) the solid phase characteristics and its morphology (k_s, different solid phases could be present); (b) the fluid phase characteristics (k_f, different fluid phases could be present in the pore structure); (c) volume pores fraction, ε; (d) pore size distribution; (e) pore diameter range; (f) ordered or random microstructure; (g) environmental conditions [20,21,23–26].

The role of the volume fraction on the influence of porous material properties has been extensively studied [27,28]. On the other hand, the influence of the structure disorder, the pore size range and pore size distribution have been studied much less [29,24]. This last aspect is associated to the complexity to control the formation of porous structure during production process as well as to the difficulty of describing and characterising it from a geometrical point of view.

During recent years, much attention has been paid to the prediction of the thermal conductivity of heterogeneous or composite materials. Various effective thermal conductivity models have been proposed [1–4]. These models, even analytical, exhibit several parameters, whose values must be empirically determined. Even though the study of the literature reveals that the number of heat transfer modelling works is growing fast, still no single model or prediction procedure has been discovered with universal applicability [30].

Owing to this reason, the application of the Fractal Geometry is of great interest. Fractal Geometry has been formalised and developed during 1970s by Mandelbrot. He recognised various fractal forms in organic as well as inorganic systems of nature [31]. Fractals are geometric figures characterised by non-integer dimension (D_f), intricate as well as complex structure, geometric construction based on iteration procedure and geometric arrangements that are repeated with different scales (self-similarity) [32].

Fractal characteristics have been recognised in several aspects of the microstructure of the materials, and they are found relevant for describing and predicting different aspects of their macroscopic behaviour.

Wang et al. applied fractal geometry to study fluid flow in porous media composed of a number of pores with arbitrary shapes and tortuous paths [33]. They proposed a fractal analytical expression to predict the flow rate, the average flow velocity, and the effective permeability for the power-law. Xu et al. studied the effect of pore fraction, fractal dimension of pore size distribution and tortuosity fractal dimension on the multiphase flow through unsaturated porous media [34].

A step forward in fractal modelling fluid flow was presented by Cai et al. [35]. They elaborated a generalised fractal model to study spontaneous imbibition process in different rock types, fibrous materials, and silica glass. Atzeni et al. determined fractal dimension from pore size distribution data in order to correlate it with mechanical behaviour of vesicular basalt used in prehistoric buildings and earth based materials, permeability of cement pastes and advanced ceramics [36–39]. Gao et al. [40] discussed the hydration factors of cement pastes in terms of the fractal dimension and the multi-fractal spectrum. This procedure was found valuable in characterising the heterogeneity of pore structure.

Moreover, fractal modelling has been used to study heat transfer on porous media. Indeed, Huai et al. developed several types of fractals to reproduce porous microstructures and to elucidate heat conduction by finite volume method [41] as well as lattice Boltzmann method [42,43]. Based on statistical self-similar, Huang et al. proposed a theoretical model in order to predict thermal conductivity of salt/expanded graphite composite material [44].

Xiao et al. [45] proposed a model to study the effect of Brownian motion of nanoparticles on the thermal conductivity. The analytical expression takes into account the fractal distribution of nanoparticles. A good agreement was found between the proposed model predictions and experimental data.

Recently, it has been presented that by applying a fractal procedure and analysis, known as Intermingled Fractal Units model (IFU), both fractal and non-fractal types of pore size distributions can be reproduced. This model is particularly suitable for simulating every type of pore size distribution. This versatility is guaranteed by the use of different fractal figures combined (intermingled, mixed) together [46,47,52]. This geometric representation of the porous microstructure is converted into electrical patterns. Consequently, it is possible to calculate thermal conductivity for every porous material.

In this paper, an IFU procedure will be used to predict thermal conductivity values of YSZ. The results will be compared with experimental and derived from Maxwell–Eucken [48] model as well as the Effective Medium Theory (EMT) data [30]. This approach, in addition to forecasting thermal conductivity, could be very useful tool to design porous ceramic microstructure controlling other properties such as fluid flow and mechanical behaviour [49].

2. Materials and methods

IFU model procedure aimed at predicting thermal conductivity values is applied to porous ceramic materials by Hu et al. [14] and Lang et al. [50] named A and B respectively.

The system A is obtained from commercially available YSZ powder (ZrO_2–8 mol% Y_2O_3, AR grade, Fanmeiya Powders Co. Ltd., Jiangxi, China). The median dimension of this powder is 1.26 μm and the value of specific surfaces is equal to 6.49 m^2/g. Freeze casting process is used, wherein the shaping vehicle and the binder are Tert-butyl alcohol (TBA) and polyvinyl butyral (PVB). Three different temperatures are used −30 °C (A1, A2, A3), −78 °C (A4, A5, A6) and −196 °C (A7, A8, A9).

In order to obtain a premixed solution, 0.5 wt% of PVB is mixed into TBA. This premixed solution is blended with YSZ powder, in the proportion of 85 vol% and 15 vol% respectively. The ball-milling process, conducted for 4 h, produced a homogeneous suspension [14].

The mixture was moulded and subjected to unidirectional freezing using different freezing agents. After freezing, a cylindrical sample (height of 18 mm and a diameter of

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