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Radiative characterization of ceramic foams with microporosity

S Haussener[1], J. Randrianalisoa[2]
[1] Ecole Polytechnique Fédérale de Lausanne, Institute of Mechanical Engineering, Lausanne, Switzerland
[2] Groupe de Recherche en Sciences Pour l'Ingénieur (GRESPI), EA 4694, Université de Reims Champagne-Ardenne, Campus du Moulin de la Housse, F-51687, Reims, France

E-mail: sophia.haussener@epfl.ch, jaona.randrianalisoa@univ-reims.fr

Abstract. The porous morphology of ceramic foams can significantly influence its heat and mass transport phenomena. Ceramic foams with dual-scale porosity provide flexibility for tailoring the coupled transport characteristics for enhanced performance. We numerically characterized the radiative transport in porous ceria foams with dual-scale porosity, i.e. exhibiting pores in the millimeter range in the micrometer range. Ceria can act as a catalyst-equivalent in high temperature thermochemical reactions for the direct synthesis of solar fuels and its bulk material properties vary significantly with wavelength. The methodology used is based on Monte Carlo methods for the solution of the volume-averaged radiative transfer equations for the determination of macroscopic optical properties such as reflectance or transmittance of a 1D slab. The exact millimeter-scale structure was incorporated by effective transport properties obtained through collision-based Monte Carlo methods. The micrometer-range strut porosity was incorporated using Mie theory and assuming independent scattering. The results allow for guiding the synthesis of ceramic foams with dual-scale porosity for enhanced radiative transport characteristics.

1. Introduction

Ceramic foams play an important role in high temperature applications as radiative burners or absorber and reactant for solar thermochemical fuel processing. The latter provide a direct pathway for solar fuel processing using ceramics such as ceria as a catalyst for thermochemical water splitting [2]. The ceramic foam acts simultaneously as direct solar absorber, heat exchanger, reactant and reaction site. The accurate characterization of the radiative transfer in these complex structures (shown in figure 1.a) is required for an in-depth understanding of the coupled heat and mass transfer, and chemical reaction. Tomography-based approaches have been used to accurately determine the heat and mass transport in complex structures at scales above 1μm [5], and to tailor the foam morphology for increased performance [10]. A further performance improvement by increasing the specific surface through the introduction of dual-scale porosity, i.e. small scale pores within the struts of the porous foam [4] (shown in figure 1.b), has been experimentally demonstrated. The performance increase results from enhanced radiation absorption and increased specific surface for the chemical reaction. We aim at providing a methodology which allows for the characterization of ceramic foams exhibiting dual-scale porosity, i.e. macroscopic pores in the range of mm, and microscopic pores in the range of μm.

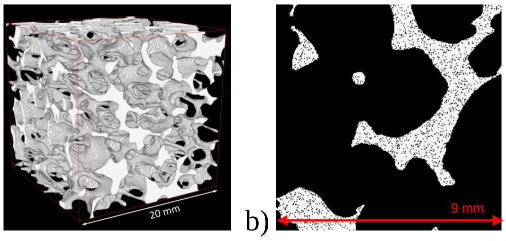
a) b)


Figure 1. a) 3D rendering of a ceria ceramic foam with porosity of 82%, obtained by X-ray tomography with resolution of 35.7 μm [10], and b) cut through a quarter of the sample indicating the 10% strut-porosity with micro-pore radius of 18 μm.

2. Problem statement

The media considered were statistically isotropic and homogeneous porous media obtained by the replica method [9], and composed of an isotropic, percolating, opaque or semitransparent solid phase, and a radiatively non-participating, stagnant fluid phase. The radiative bulk properties of ceria, i.e. its spectral complex refractive index, are given in literature [8]. The investigations were based on the digitalized morphological data of a sample with porosity of 82%. The porosity of this sample was then artificially decreased by increasing the strut diameter by dilation operations [10]. We considered 2 samples of 82% and 60% porosities. The microscopic pores in the struts were artificially added to the foam struts and spanned pore sizes of 10 to 40 μm at a volume fraction in the solid phase of 0 to 40%.
Geometrical optics was assumed for the calculation of the macroscopic properties as the smallest dimensions of the solid structure (about 0.4 mm) was significantly larger than the wavelength range considered. Diffraction was neglected as for large particles it is predominantly in forward direction within a small solid angle. The radiative characteristics of struts containing microscopic pores was obtained by including the contribution of ceria absorption and the scattering by pores in the framework of independent scattering theory [1]. Possible dependent scattering effects for highly porous struts were neglected. Scattering efficiency factors and phase functions by small pores were calculated using generalized Mie theory [11], accounting for the possibly absorbing ceria surrounding the pores.

3. Methodology

The methodology is based on volume-averaging approaches used for the characterization of the radiative transfer in multi-phase media. Two coupled radiative transfer equations (RTEs) are solved [7, 6, 3] for an infinite 1D slab. The boundary conditions for the slab are reflecting at the lateral walls, absorbing environment at the outlet, and collimated irradiation at the inlet. A path-length based Monte Carlo method is used for the solution of the volume-averaged RTEs. The macroscopic radiative properties, reflectance and transmittance of the 1D slab, are calculated based on the volume-averaged intensity vector field in the two phases and its integration over all solid angles of the positive and negative half-sphere at the inlet and outlet. The effective properties required for the volume-averaged RTEs, namely absorption coefficients for the fluid and solid phases, four scattering coefficient accounting for refraction and reflection at both sides of the interface boundaries, and extinction coefficients for the fluid and solid phase, and four scattering phase functions accounting for the angular distribution of the refraction and reflection at both sides of the interface boundaries, are determined using a collision-based Monte Carlo method in the 3D numerical sample [7, 6, 3]. The bulk properties accounting for the small-scale porosity within the struts are calculated using the BHMIE algorithm [1].

4. Results

The effective radiative properties for the void and solid phases (subscripts 1 and 2, respectively), namely extinction coefficients (β_1, β_2), scattering albedos (ω_{11}, ω_{12}, ω_{21}, ω_{22}), and scattering function asymmetry factors (g_{21}, g_{22}), are depicted in figure 2. The macroscopic optical properties, namely transmittance and reflectance of a 1D slab, are depicted in figure 3. The introduction of micro pores in the struts (strut porosity) has no effect on the radiative behaviour for smaller wavelengths as the ceria is opaque given the large imaginary part of the refractive index. For larger wavelengths and smaller imaginary part of the refractive index, an increase in the strut porosity at a constant micropore size or an increase in the micropore sizes at a constant strut porosity, respectively, increases the transmittance and reduces the reflectance due to increased void space and reduced averaged solid extinction coefficient. This effect is most significant at small pore sizes or volume fractions and vanishes at larger pore sizes and volume fractions as the strut becomes optically thick due to the strongly increased scattering.

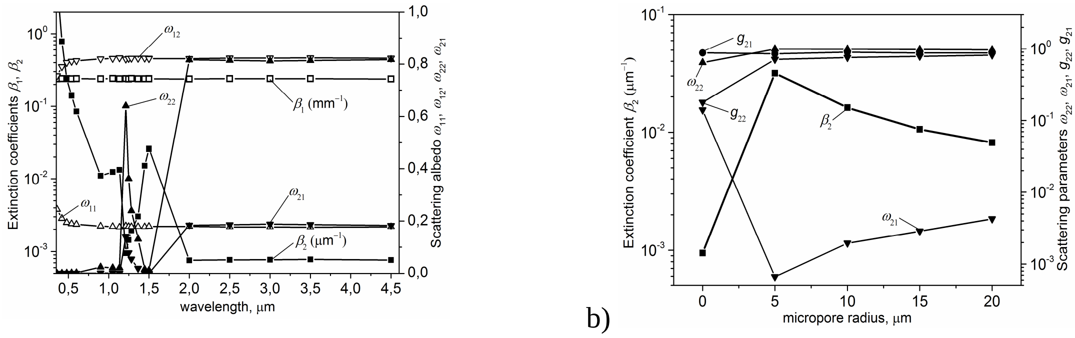


Figure 2. a) Effective radiative properties of the ceria foam with 82% porosity and no microscopic strut porosity, and b) effective radiative properties for the solid phase only for the ceria foam of 82% porosity with additional 10% microporosity in the strut for varying micropore radii.

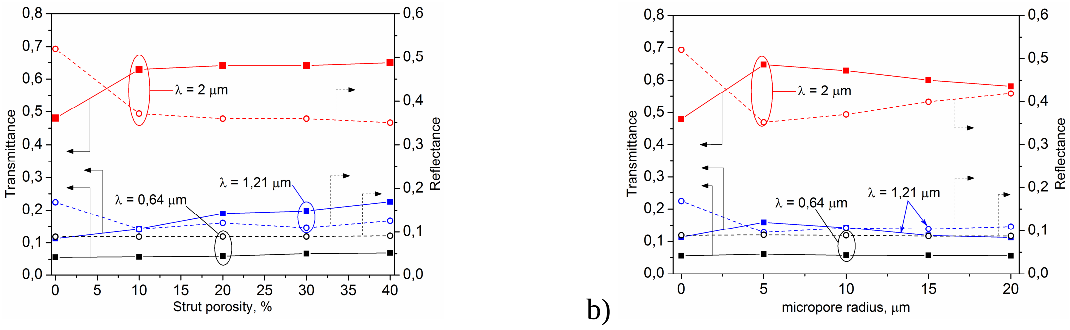


Figure 3. Transmittance and reflectance of a 9.5 mm-thick sample of ceria foam of 82% porosity (a) at constant strut pore size of 20 µm and with increasing strut porosity, and (b) at strut porosity of 10% and with increasing strut pore diameter.

5. Conclusions

The proposed methodology based on the volume-averaged RTEs and Mie theory for the small-scale scatterers allows for the radiative characterization of porous ceramic foams with dual-scale porosity.

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