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Pore-level engineering of macroporous media for increased performance of solar-driven thermochemical fuel processing

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ABSTRACT

The performance of high-temperature solar reactors incorporating porous ceramic materials that serve as radiative absorbers and chemical reaction sites can be improved significantly by tailoring their pore structure. We investigated the changes in their effective heat and mass transport properties with increasing mass loading of porous ceramics fabricated by the replica method. We applied a methodology consisting of the experimental characterization of the structure via 3D tomographic techniques coupled to pore-level direct numerical simulations for the determination of the effective transport properties. This approach was extended by using digital image processing on the structure data to allow for artificial changes in the morphological characteristics – corresponding to actual variations in the fabrication process. We derived transport correlations of porous ceria foam with varying mass loading, i.e. reticulate to dense foams with porosity from 0.85 to 0.45. We observed that the correlations proposed in literature do not accurately describe the behavior of low-porosity foams. The numerical findings of this study provide guidance for pore-level engineering of materials used in solar reactors and other high-temperature heat and mass transfer applications.

1. Introduction

Porous ceramic materials exhibit favorable morphological, mechanical, and transport properties when applied as absorbers [1], heat exchangers [2], insulators [3], chemical reaction site, and reactants [4], in a wide variety of high-temperature applications ranging from chemical processing, combustion, and filtering, to solar reactor technology. The effective heat and mass transport properties of these porous materials largely depend on their morphology [5,6]. For example, solar reactors designed for thermochemical water and CO_2-splitting using porous, ceria-based redox materials have shown an increase in the efficiency by a factor of four when changing the material's morphology from monolithic-type geometry with μm-range pore size to a foam-type geometry with mm-range pore size [1,4]. Thus, pore-level engineering of materials can significantly improve the performance of solar reactors.

Frequently, the effective transport properties of macroporous media are approximated by empirical correlations or semi-empirical and analytical models derived for simplified morphologies or unit-cell structures. To predict the permeability of a porous medium, approximations based on the semi-heuristic packed-bed model of Carman and Kozeny [7] are used with a modified shape factor for e.g. assemblies of parallel cylinders [8] and fibrous beds [9]. Another drag flow approach was analytically derived by Ergun [10] for packed columns. An alternative flow analysis considers the Hagen–Poiseuille relation in a stack of tubes with diameters equal to the pore size [5]. To predict the Dupuit–Forchheimer coefficient, semi-empirical models based on Ergun's equation (Ergun [10] and Macdonald et al. [11]) and phenomenological correlations proposed by Ward [12] and Cooke [13] are developed for packed beds. These models exhibit an inverse proportionality to the permeability. The tortuosity has mainly been investigated in porous sediment layers by Archie [14], and Iversen and Jorgensen [15], who found porosity-dependent empirical correlations, and by Weissberg [16], and Boudreau and Meysman [17], who derived geometrical models based on stacked spheres and disks, respectively. The volume averaged interfacial heat transfer coefficient is commonly expressed by correlations $\mathrm{Nu} = f(\mathrm{Re}, \mathrm{Pr})$, as suggested by Wakao et al. [18]. Kuwahara et al. [19] modeled an uniform, 2D flow through a periodic arrangement of isothermal square rods and fitted the ztheoretically derived correlation to the heuristic model of Wakao

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Nomenclature

Symbols

A	empirical fitting parameter in correlation of Calmidi and Mahajan
A_0	specific surface area ($\mathrm{m^2\,m^{-3}}$)
a	free fitting parameter in Archie's correlation
a_0, a_1, a_2	constants in Eq. (1) ($\mathrm{m^2\,m^{-3}}$)
b	geometrical model parameter in Weissberg correlation
b_0, b_1	constants in Eq. (2) (m)
c	empirical fitting parameter of Iversen and Jorgensen
c_p	specific heat capacity at constant pressure ($\mathrm{J\,kg^{-1}\,K^{-1}}$)
d	pore diameter (m)
d	geometrical model parameter in Eq. (10)
e_0, e_1, e_2, e_3, e_4, e_5	constants in Eq. (11)
F	Dupuit–Forchheimer coefficient ($\mathrm{m^{-1}}$)
f	three-resistor model parameter
g_0, g_1	constants in Eq. (12)
h	heat transfer coefficient ($\mathrm{W\,m^{-1}\,K^{-1}}$)
i	phase indices
l	length (m)
K	permeability ($\mathrm{m^2}$)
k	thermal conductivity ($\mathrm{W\,m^{-1}\,K^{-1}}$)
k_K	shape factor
m	fitting parameter in Cooke's correlation
$\mathbf{n}$	unit normal vector
n	Fitting parameter in Cooke's correlation Eq. (8) and empirical fitting parameter in correlation of Calmidi and Mahajan in Table 2
n_Ω	number of discretized angles
Nu	Nusselt number
p	pressure (Pa)
Pr	Prandtl number
q	heat rate (W)
r	reflectivity
Re	Reynolds number
T	temperature (K)
$\mathbf{u}$	velocity vector ($\mathrm{m\,s^{-1}}$)
u_D	Darcean velocity ($\mathrm{m\,s^{-1}}$)
V	volume ($\mathrm{m^3}$)
z	Cartesian axis in main flow direction

Greek symbols

β	extinction coefficient ($\mathrm{m^{-1}}$)
ε	Porosity
ζ	dimensionless effective thermal conductivity
η	dimensionless fluid thermal conductivity
κ	absorption coefficient ($\mathrm{m^{-1}}$)
λ	wavelength (m)
μ	dynamic viscosity ($\mathrm{kg\,m^{-1}\,s^{-1}}$)
μ_s	cosine of reflection angle
ρ	density ($\mathrm{kg\,m^{-3}}$)
σ	scattering coefficient ($\mathrm{m^{-1}}$)
τ	tortuosity
Φ	scattering phase function
ψ	scalar property (–)
Ψ	empirical fitting parameter for the extinction coefficient models

Subscripts

b	blackbody
e	effective
f	fluid
j	counter
l	local
lm	logarithmic mean
mf	mean fluid
r	radiation
s	solid
sf	solid–fluid interface

Abbreviations

CT	computed tomography
MC	Monte Carlo
Nu	Nusselt
Pr	Prandtl
Re	Reynold
REV	representative elementary volume
RMS	root mean square
RPC	reticulate porous ceramic
RTE	radiative transfer equation

et al. Gunn [20] studied convective heat transfer in packed beds with a stochastic model. Artificial unit-cell structures were mainly used to study thermal conduction in porous materials. Bhattacharya et al. [21] modeled a 2D structure as a field of hexagons with hexagonal nodes. Boomsma and Poulikakos [2] used 3D tetrakaidecahedron cell elements with cubes at the intersection to derive an analytical heat conduction correlation. Russell [22] and Loeb [23] published theoretical models for the simple porous structure consisting of equally sized void cubes distributed in the solid matrix. Maxwell [24] derived a porosity dependent upper bound for the effective thermal conductivity of a two-phase medium. A phenomenological correlation is provided by the three-resistor model of Wyllie and Southwick [25]. Calmidi and Mahajan [26] determined the effective thermal conductivity of aluminum foams empirically. Two models used to predict the effective extinction coefficient are based on geometrical optics for porous media consisting of a suspension of mono-dispersed and independently scattering particles. Hsu and Howell [27] considered spherical particles whereas Loretz et al. [28] investigated multi-faced particles.

It is evident that these effective properties strongly depend on the morphology. This is supported by theoretical derivation of the effective transport properties by the volume averaging theory [29], which indicates that the effective properties are a function of the morphology of the porous medium, the bulk properties of its phases, and the phase boundary conditions only. However, the aforementioned empirical, semi-empirical, and analytical correlations do not consider the exact morphology of complex and stochastic porous materials and, consequently, provide less accurate transport characteristics. Furthermore, they can only provide trends in the same class of materials when applied to pore-level engineering of media. The incorporation of the exact structure is therefore crucial for the accurate heat and mass transport characterization and subsequent pore-level engineering for enhanced transport.

Recently, coupled experimental–numerical techniques have been proposed for accomplishing that. They use 3D imaging-based techniques such as computed tomography, magnetic resonance imaging, or focused ion beam, to obtain the exact structural information of the porous media, which in turn is used in pore-level numerical simulations to solve the governing conservation equations in the various phases of the porous media. In conjunction with the volume averaging theory, the effective transport properties are then derived. Computed tomography (CT) has been one of the preferred methods as it can offer non-destructive and non-invasive techniques with relatively high resolution. Examples of the CT-based methodology are the determination of the effective