Doctoral Thesis

Tomography-based determination of effective heat and mass transport properties of complex multi-phase media

Author(s):
Haussener, Sophia

Publication Date:
2010

Permanent Link:
https://doi.org/10.3929/ethz-a-006334716

Rights / License:
In Copyright - Non-Commercial Use Permitted
TOMOGRAPHY-BASED DETERMINATION OF EFFECTIVE HEAT AND MASS TRANSPORT PROPERTIES OF COMPLEX MULTI-PHASE MEDIA

A dissertation submitted to
ETH ZURICH
for the degree of
Doctor of Sciences

presented by
SOPHIA HAUSSENER
MSc ETH ME
born March 22, 1983
citizen of Rüeggisberg (BE)

accepted on the recommendation of
Prof. Dr. Aldo Steinfeld, examiner
Prof. Dr. Jean Taine, co-examiner

2010
Abstract

Transport phenomena in multi-phase media are of interest in a wide range of areas in science and industry chemical processing, combustion, nuclear and civil engineering, environmental and medical engineering, filtering and automotive applications, atmospheric sciences and solar engineering. Of special interest are solar thermal and thermochemical processes to generate electricity and (storable) solar fuels, solar materials and chemical commodities. In these processes multi-phase media serve as insulators, radiant absorbers, heat exchanges, catalyst carriers, reactant and reaction sites. The analysis of the complex interactions between multi-mode heat transfer, multiphase flow, and chemical reaction – on multiple scales – is fundamental to understanding and optimizing these processes. Volume averaging models for multi-phase media are commonly applied for process simulations. However, these models rely to a great extend on the accurate knowledge of the multi-phase media’s effective transport properties, which in turn depend on the morphology and single phase properties.

A combined experimental-numerical procedure is adopted in this thesis: the exact 3D geometry of the complex multi-phase media are experimentally determined by computed tomography and used in direct discrete-scale simulations for morphological characterization and determination of the effective heat and mass transport properties. Two-point correlation functions and mathematical morphology operations are calculated for morphological characterization and validated by weight, BET surface area and laser scattering measurements. Collision-based Monte Carlo is utilized to calculate distribution functions of attenuation path length and direction of incidence at the phase boundary which are used to determine effective radiative properties. Spectroscopic measurements are conducted for validation of the calculated radiative properties. Finite volume techniques are used to solve for mass, momentum and energy conservation allowing for conductive/convective heat transfer and flow characterization. The methodology is then applied to four multi-phase media relevant in solar material and fuel processing: (i) reticulate porous ceramics, (ii) anisotropic ce-
Ceramic foams, (iii) reacting packed beds and (iv) semitransparent-particle packed beds. A fifth medium relevant in the area of environmental sciences and climate modeling is investigated to show the wide applicability of the methodology: (v) layers of characteristic snow types.

Porosity, specific surface area, and pore-size distribution of a non-hollow SiSiC reticulate porous ceramic with nominal pore diameter of 1.27 mm are calculated. The determined extinction coefficient of 431 m$^{-1}$ compares well to experimental estimates. The scattering phase function shows an enhanced fraction of backward scattering for assumed diffuse surface reflection. The ratio of effective conductivity to solid conductivity for small ratios of fluid to solid conductivities converges to 0.022. A Reynolds and Prandtl dependent Nusselt correlation is determined and converges to 6.8 for small Reynolds numbers. The numerically determined permeability and the Dupuit-Forchheimer coefficient compare well to values available in literature for materials with similar morphology. For neglected molecular dispersion, a Reynolds dependent dispersion tensor is calculated. Mean tortuosity is determined to be 1.07. The calculated effective properties are then incorporated in a continuum model of a solar evaporator/decomposer reactor. The reactor model is compared to experimentally measured temperatures at multiple locations within the reactor. Optimization of process parameters, reactor design and foam morphology is conducted; it shows a predominant influence of total acid solution flow rate and solar irradiation. Peak energetic and chemical efficiencies of 73% and 45%, respectively, are calculated for 2 ml/min acid inflow and 150 W solar power input.

Ceramic foams of ceria with structural anisotropy due to uniaxial pressing (along the z-direction) and anisotropic primary particles show enhanced extinction in z-direction because the pores are squeezed, which results in shorter attenuation paths. Effective conductivities in the x- and y-directions increase due to the more parallel alignment of the structures with the heat flux in these directions. Convective heat transfer in z-direction is larger because of the more tortuous path for fluid flow, increasing the accessible surface area for fluid-solid heat exchange. Reduced permeability and larger Dupuit-Forchheimer coefficient in z-direction are observed because of larger tortuosity along this direction. Preliminary studies on tailored foam designs, adjusted to the specific needs of the process in which the foam is applied, allow for foam engineering and consequently enhanced process performance.

A reacting packed bed is analyzed for heat and mass transfer. Gasification of waste tire shreds is chosen as model reaction. Porosity, specific surface and particle-size distribution are numerically calculated and compare to experimen-
tal data. Limitations in computerized tomography resolution shows to be the preliminary cause of discrepancies observed, especially at larger reaction extent where nanopores are formed. Larger extinction coefficients are calculated with increasing reaction extent due to particle shrinking and break-up, resulting in shorter attenuation path lengths. The scattering phase function shows to be independent of reaction extent for assumed diffusely reflecting particles. Effective conductivity, calculated by neglecting particle-particle contact resistances, decreases with reaction extent due to larger porosity and smaller particle size. The evolution of highly porous particles during pyrolysis and the subsequent shrinking and break-up of the particles cause a decrease and re-increase in convective heat exchange and Dupuit-Forchheimer coefficient. The largest permeability is calculated for the highly porous particles and decreases again for the final packed bed configuration. Comparison with heat and mass transfer properties available in the literature shows acceptable agreement for media with comparable morphology.

The derivation of the volume-averaged radiative transfer equations in multi-phase media builds the basis for the radiative characterization of a semitransparent-particle packed bed. Nonspherical calcium carbonate particles are chosen as model particles and properties in the spectral range of 0.1 to 100 μm are calculated. The extinction coefficient for the transparent void phase solely depends on morphology, while the extinction coefficient for the semitransparent particles increases with increasing wavelength. The scattering coefficients in each phase are strongly influenced by the surface reflectivity of the boundary and show complementary behavior. The spectral scattering phase functions for diffusely reflecting particles show minor dependence on wavelength for both phases while they strongly depend on wavelength for specularly reflecting particles. Validation of the methodology by the analytical solutions for a diluted particle cloud of large opaque particles shows good agreement.

The morphological and radiative properties of snow layers composed of five characteristic snow types are numerically determined. Calculated extinction coefficients, scattering coefficients and scattering phase functions in the spectral range of 0.3 to 3 μm are then incorporated in a continuum model of a layer of snow composed of the different snow types and irradiated by a diffuse or collimated radiation flux. Overall reflectance, transmittance and absorptance are determined and compared to transmittance measured with a spectroscopic setup. Comparison of the calculated radiative properties based on the exact snow morphology, obtained by computerized tomography, with the one calculated based on simplified morphologies (packed beds of spheres) shows deviations
up to 24% in reflectance, implying a significant influence of snow morphology on the radiative behavior. Additionally, soot impurities in snow are modeled and show a reduction in the calculated reflectivity by up to 83%.

The determined morphological properties can be used for the determination of structural parameters needed in kinetic models. The calculated effective heat and mass transport properties can be incorporated in volume-averaged (continuum) models of processes accounting for coupled heat/mass transfer (including chemical reactions) and fluid flow. The continuum models, in turn, are used for process design, modeling, optimization and scale-up. Accurate modeling and an in-depth understanding of the processes involving the multi-phase media is achieved. Additionally, the influence of multi-phase media’s morphology on heat and mass transfer and consequently process performance is understood. The tomography-based discrete-scale numerical simulations show to be widely applicable, also for nonsolar applications such as environmental science and medical engineering.
Zusammenfassung


Der Extinktionskoeffizient von keramischem Schaum aus Ceroxid, welches durch einachsiges Pressen (entlang der $z$-Richtung) und der Anisotropie der Primärpartikel strukturelle Anisotropie aufweist, ist entlang der $z$-Richtung erhöht. Dies weil Poren in dieser Richtung zusammengequetscht werden, was


Die Herleitung der volumengemittelten Strahlungsgleichungen in Mehrphasenmedien bilden die Basis für die Strahlungscharakterisierung einer Schüttung aus semitransparenten Partikeln. Nichtrunde Partikel aus Kalziumkarbonat werden als Modellpartikel gewählt. Die Eigenschaften werden in einem spek-
Zusammenfassung


Die morphologischen und die Strahlungseigenschaften von Schneeschichten, bestehend je aus fünf charakteristischen Schneetypen, werden numerisch bestimmt. Die in einem spektralen Bereich von 0.3 bis 3 μm berechneten Extinktionskoeffizienten, Streuungskoeffizienten und Phasenfunktionen werden in einem Kontinuummodell einer Schneeschicht, welche aus verschiedenen Schneetypen besteht und mit diffuser oder kollimierter Strahlung beschienen wird, verwendet. Reflektivität, Transmission und Absorption der Strahlung werden bestimmt und mit Transmissionswerten, gemessen mit einer spektroskopischen Einrichtung, verglichen. Die Strahlungseigenschaften, welche basierend auf der exakten Morphologie, die mit Computertomografie ermittelt wird, berechnet werden, werden mit Strahlungseigenschaften verglichen, die basierend auf vereinfachter Morphologie (Schüttsschicht von Kugeln) berechnet werden. Sie unterscheiden sich um bis zu 25% und zeigen auf, dass es einen signifikanten Einfluss der Schneemorphologie auf die Strahlungseigenschaften gibt. Zusätzlich werden Verunreinigungen aus Russ im Schnee modelliert und es wird gezeigt, dass diese Verunreinigungen die Reflektivität um bis zu 83% reduzieren können.

breite Anwendbarkeit auf. Dies ist der Fall sowohl für solare als auch für nichtsolare Anwendungen wie beispielsweise im Bereich der Umweltnaturwissenschaften und der Medizintechnik.