Morphology Engineering of Porous Media for Enhanced Solar Fuel and Power Production

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The favorable and adjustable transport properties of porous media make them suitable components in reactors used for solar energy conversion and storage processes. The directed engineering of the porous media’s morphology can significantly improve the performance of these reactors. We used a multiscale approach to characterize the changes in performance of exemplary solar fuel processing and solar power production reactors incorporating porous media as multifunctional components. The method applied uses imaging-based direct numerical simulations and digital image processing in combination with volume averaging theory to characterize the transport in porous media. Two samples with varying morphology (fibrous vs. foam) and varying size range (mm vs. μm scale), each with porosity between 0.46 and 0.84, were characterized. The obtained effective transport properties were used in continuum-scale models to quantify the performance of reactors incorporating multifunctional porous media for solar fuel processing by photoelectrochemical water splitting or power production by solar thermal processes.

INTRODUCTION

The direct conversion of solar energy into a storable, high-energy density fuel via solar thermochemistry or photoelectrochemistry and the solar production of power via solar thermal processes are promising renewable fuel processing and power production routes. The essential requirements for the processes’ impact on our fuel and power economy are their sustainable, efficient, stable, and economic implementation via solar reactors and their assembly into practical large-scale systems.1–4 The engineering of solar reactors needs to address various issues including optimal design and operational conditions for enhanced coupled multiphysics transport and the integration and optimization of multiscale components, e.g., porous media. The latter is of special interest as porous media exhibit favorable and tunable transport properties. Particularly interesting is the observed fourfold increase in efficiency of a solar reactor for the thermochemical splitting of water and CO₂ into synthesis gas when changing the porous absorber and reactant morphology from a sintered backed bed to a highly porous foam morphology.5,6 Similar influences on performance are expected for solar receivers used for thermal power production, which rely on porous absorber and heat exchangers of various morphologies,7,8 or for photoelectrochemical fuel production devices relying on microstructured to nanostructured photoelectrodes9–13 or separators.14

The systematic understanding of how the transport properties of porous media change for distinct base morphologies (foam or fibrous structure) but varying morphological characteristics, i.e., varying porosity, is of practical interest due to the straightforward synthesis of such porous media. Systematic direct numerical studies on how morphology and its tailoring can influence the transport properties have been conducted for micron-sized sintered packed beds,15 reticulate porous ceramics and packed beds,16,17 and foams.18

We used a multiscale approach to quantify the changes in the performance of solar reactors incorporating porous media with varying morphologies (foams and fibers) and morphological properties (characteristic size and porosity). This multiscale approach consisted of (1) the transport quantification in porous structures by applying tomography-based approaches that incorporate the exact morphology into pore-scale numerical simulations19–22 in combination with image processing techniques,18 and (2) the subsequent use of the transport characteristics in continuum-scale models of reactors.
processing fuel via photoelectrochemical processes or power via solar thermal processes.

**METHODOLOGY**

**Multiscale Approach**

*Continuum-Scale Models of Reactors Incorporating Multifunctional Porous Media*

Photoelectrochemistry is a promising direct solar-to-fuel processing route. It uses the photon energy to generate electron–hole pairs in a semiconductor absorber. The electron–hole pairs are separated via an electric field and used at liquid–solid interfaces to drive catalyzed electrochemical reactions such as water electrolysis described by

\[
2\text{H}_2\text{O} + 4\text{h}^+ \rightarrow 4\text{H}^+ + \text{O}_2, \quad (1)
\]

\[
4\text{H}^+ + 4\text{e}^- \rightarrow 2\text{H}_2, \quad (2)
\]

in an acidic environment. Ions are transported through an ion-conducting phase to conserve the charge. This is schematically shown in Fig. 1a for a solar electrochemical reactor incorporating a multifunctional porous slab.

Solar thermal processes for renewable power production use the solar energy as sensible heat to heat a (pressurized) heat transfer fluid, which is used in subsequent power cycles. This is schematically shown in Fig. 1b for a solar receiver incorporating a multifunctional porous slab.

**Radiative Heat Transfer** The radiative transport in porous media, composed of a transparent and an opaque phase, was given by the volume-averaged radiative transfer equation

\[
\textbf{s} \cdot \nabla I = -\beta_e I + \chi_e I_b + \frac{\sigma_{\text{scat},e}}{4\pi} \int_0^\infty \Phi_{\text{scat},e} I d\Omega \quad (3)
\]

\[
\text{Fig. 1. Schematics of the 1-D slabs of multifunctional porous media to be used for solar (a) electrochemical fuel and (b) thermal power processing, and the corresponding boundary conditions for the continuum-scale models.}
\]
porous medium. This approach neglected possible bulk and surface recombination mechanism in the absorber. The effective ionic and electronic conductivities, \(\kappa_\text{e} \) and \(\sigma_\text{e} \), were calculated via the direct numerical simulation technique, and the reaction overpotentials at the anode and cathode, \(\eta_\text{a} \) and \(\eta_\text{c} \), were approximated by Tafel expressions assuming that reaction occurred at the top and bottom solid phase boundary. The second fraction on the right side of Eq. 4 described the fraction of the current density that was not lost due to product crossover via diffusion through the membrane. The effective hydrogen diffusivity was calculated via direct numerical simulations and the worst-case scenario for crossover was assumed, i.e., the hydrogen evolution side completely saturated with hydrogen and the oxygen evolution side completely hydrogen starved.\(^{14}\)

**Solar Thermal Power Production** We used a quasi-one-dimensional (1-D) model to solve the steady-state mass, momentum, and energy conservation equations at local thermal nonequilibrium in the solar thermal power production reactor incorporating a multifunctional porous media. The volume-averaged energy conservation equations were given by

\[
0 = \nabla \cdot (k_{\text{e,s}} \nabla T_s) - h_{\text{sf}} A_0 (T_s - T_f) + q_r
\]

(7)

\[
\rho C_p \nabla \cdot (u T_f) = \nabla \cdot (k_{\text{e,f}} \nabla T_f) + h_{\text{sf}} A_0 T_f (T_s - T_f)
\]

(8)

where the temperature variables represented intrinsic averages. It was assumed that only the solid fraction of the inlet and outlet boundary \(A_s\) reradiated, describing the boundary condition for the inlet and outlet boundaries

\[
Q_{\text{rerad}} = (1 - r) A_s \sigma T_s^4
\]

(9)

where \( r \) was the directional-hemispherical reflectivity of the material. The lateral walls were symmetric boundaries. The carrier gas was modeled as an ideal gas with temperature-dependent properties. The solar-to-power efficiency of the slab of multifunctional porous medium was described by

\[
\eta = \frac{m(h_{\text{outlet}} - h_{\text{inlet}})}{Q_{\text{sol,in}} + W_{\text{pump}}} \eta_{\text{Carnot}} T_{f|z=0}
\]

(10)

The pumping work was given by the volume-averaged momentum conservation\(^{29}\)

\[
W_{\text{pump}} = \Delta p u_f A = \left(\frac{h_f}{K} u_f + \rho_f F u^2 \right) u_f A
\]

(11)

with \( u_f \) the superficial velocity average. The effective transport properties permeability \( K \) and the Forchheimer coefficient \( F \) were determined via the direct numerical simulations. An inlet mass flow and an outlet pressure were set at the corresponding boundaries.

**Pore-Scale Transport Characterization**

**Radiative Heat Transfer** \(\beta_\text{e}, \sigma_{\text{scat,e}}, \) and \(\Phi_{\text{scat,e}}\) were calculated by solving the pore-scale radiative heat transfer equations. Radiative distribution functions were used to determine the effective radiative properties\(^{30}\) using a collision-based Monte Carlo technique. An opaque solid phase and a transparent fluid phase were assumed. The laws of geometrical optics were applicable as \(\pi d/\lambda \gg 1\).

**Conductive Heat Transfer** \(k_{\text{e,s}}\) and \(k_{\text{e,f}}\) were calculated by solving the steady-state conduction equations within the void and solid phases\(^{20,22,31,32}\) in a representational cubical sample of porous medium applying predefined inlet and outlet temperatures, insulating lateral wall boundaries, and providing continuity in temperature and heat flux at the solid–fluid phase boundary. For the orthotropic fibrous sample, the temperature boundary conditions were applied along the \(z\)-direction. \(k_{\text{e,s}}\) and \(k_{\text{e,f}}\) were determined as the asymptotic cases where the heat conductivity of the fluid phase was significantly smaller than the heat conductivity of solid phase \((k_f \ll k_s)\) and vice versa \((k_s \ll k_f)\).

**Convective Heat Transfer** The Nu correlations were calculated by solving the heat, mass, and momentum (incompressible and laminar) conservation equations within the fluid phase of the porous media for an isothermal solid phase. The fluid phase of cubical porous samples within a square duct with inlet and outlet regions was used as computational domain\(^{21,22,31,32}\) Uniform inlet velocity and temperature, constant outlet pressure, symmetry at the lateral duct walls, and no-slip and constant temperature at the solid–fluid interface were assumed. Inlet velocities in the range of 0.0008–0.5 m/s (corresponding to \(D_{\text{void,foam}}\)-based Re between 0.1 and 100) and Pr numbers of 0.5, 1, and 10 were simulated. For the orthotropic fibrous sample, the inlet and outlet boundaries were applied along the \(z\)-direction.

**Mass Transfer** \(K\) and \(F\) were determined by using the previously calculated pressure and velocity vector fields\(^{22}\) For the orthotropic fibrous sample, the pressure boundary conditions were applied along the \(z\)-direction. \(D_{\text{H}_2,e}\) was calculated by

\[
D_{\text{H}_2,e} = D_{\text{H}_2} \frac{e}{\ell_{\text{mean}}}
\]

(12)

The mean tortuosity, \(\ell_{\text{mean}} = \ell_{\text{path,mean}}/\ell\), was numerically determined using the previously calculated velocity vector field and determining the lengths of 3600 streamlines with uniformly distributed starting points at the medium inlet for \(Re = 0.1\).\(^{22}\)

**Charge transfer** \(\kappa_\text{e} \) and \(\sigma_\text{e} \) were calculated by solving the charge transport equations, i.e., Ohm’s law, in the ionic and electronic phases. The methodology
was analogous to the methodology used for the
determination of the effective thermal conductivity
in porous media.\textsuperscript{20,22} Again, the asymptotic cases
were considered, i.e., $\kappa_s at \kappa_s \ll \kappa_f$ and $\sigma_s at \sigma_s \ll \sigma_f$,
respectively. For the orthotropic fibrous sample, the
potential boundary conditions were applied along
the $z$-direction.

**Morphology**

Exemplary, we used two distinct morphologies
with continuous, percolating solid phases: (1) foam-like
structures and (2) fibrous-like structures. The foam-like samples have been manufactured by a
template method using a ceria-based slurry and
10 ppi polyurethane sponges.\textsuperscript{6} The fibrous samples
were ceria felts manufactured by Zircar Zirconia,
Inc. (Florida, NY, USA). These structures were im-
aged via x-ray computed tomography. The initial
samples can be subjected to fabrication variations,
e.g., increasing strut and fiber sizes by additional
applications of slurry or coating. This variation in
the fabrication process and the corresponding
changes in the sample’s morphology were virtually
reproduced by digital image processing, i.e., dilation
operations with spherical structuring elements.

The three-dimensional (3-D) rendering of the two
base morphologies and varying porosities, i.e.,
increasing numbers of applied coating steps, i.e., dilation
operations with increasing size of the structural
element, are depicted in Fig. 2. The $z$-direction
described one of the two symmetry axis of the orthotropic
fibrous sample, perpendicular to the fiber alignment.

The characteristic sizes of the two original samples
differed by two orders of magnitude. To allow
for a direct comparison and a meaningful size range
for the photoelectrochemical application, the foam-
like sample was shrunk by a factor of 100 to
represent a third type of media exhibiting the same
morphology as the original foam but in the same
size range as the fibrous material. We thus investi-
gated two foam-like samples with two distinctly
different size ranges (foam original and foam
shrunk) and one fiber sample.

The mean pore diameters, represented as the largest
spherical pore within the pore space, were calcu-
lated via opening operations and were $d_{\text{void,foam,o}} = 2.6, 2.3, 2.0,$ and 1.7 mm for the original foam-like
samples for decreasing porosity. The shrunk foam-like
sample’s pore diameters were $d_{\text{void,foam,s}} = 26, 23, 20,$
and 17 $\mu$m for decreasing porosity. The mean fiber and
pore diameter were calculated via digital image pro-
cessing, i.e., connectivity calculations, and they were
$d_{\text{solid,foam}} = 24, 36, 48, and 59 \mu$m and $d_{\text{void,foam}} = 95, 83, 71, and 59 \mu$m for decreasing porosity, respectively.

The specific surface areas were calculated via two-point
correlations and fitted to a second-order polynomial
functions

\begin{equation}
A_0 = -117740 \varepsilon^2 + 147250 \varepsilon - 1116 \quad (13)
\end{equation}

\begin{equation}
A_0 = (-2277 \varepsilon^2 + 2533 \varepsilon + 262) d_{\text{void,foam,o}} \quad (14)
\end{equation}

for fibrous and foam-like (original and shrunk) samples, respectively. The anisotropy of the sam-
ples was characterized via mean intercept lengths\textsuperscript{35}
and showed a degree of anisotropy of 0.23 and 0.55
for the foam and the fibrous samples, respectively.

**RESULTS**

**Transport Characterization**

Normalized $\beta_e$ of the foam (original and shrunk)
and fibrous samples with different porosities is
depicted in Fig. 3. The increase in $\beta_e$ with decreasing

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Fig. 2. Foam-like morphology with sample edge length = 20 mm (a–d),\textsuperscript{18} fibrous morphology with sample edge length = 0.49 mm (e–h), for
porosities 0.84 (a, f), 0.72 (b, f), 0.59 (c, g), and 0.46 (d, h).
porosity was linear and is in accordance with the increasing characteristic pore size for decreasing porosity. The increase in \( \beta_{\text{void}}/\beta_{\text{void,0.84}} \) with decreasing porosity was more pronounced for the foam sample. \( \Phi_{\text{scat,e}} \) for diffusely reflecting phase boundaries showed little dependence on the morphology and porosity and were fitted to a second-order polynomial

\[
\Phi_{\text{scat,e}} = 0.58\mu_{\text{scat}}^2 - 1.40\mu_{\text{scat}} + 0.81
\]  

(15)

\[
\Phi_{\text{scat,e}} = 0.59\mu_{\text{scat}}^2 - 1.41\mu_{\text{scat}} + 0.80
\]  

(16)

for fibrous and foam-like (original and shrunken) samples, respectively.

The Nusselt numbers were given by

\[
\text{Nu} = 5.54 + (0.71\varepsilon^2 - 0.63\varepsilon + 0.30)Re^{0.70}\varepsilon^{0.1}Pr^{0.6}
\]  

(17)

\[
\text{Nu} = 6.55 + (1.3\varepsilon^2 - 1.29\varepsilon + 0.62)Re^{0.93}\varepsilon^{0.55}Pr^{0.6}
\]  

(18)

for fibrous and foam-like (original and shrunken) samples, respectively. The foam-like samples showed an almost constant Nu at low Re, independent of porosity, while for Re > 10, the Nu was highly porosity dependent. The Nu numbers of the fibrous sample showed almost no porosity dependence for the whole Re range investigated.

The normalized effective conductivities are depicted in Fig. 4. The effective thermal and ionic conductivity of the fluid phase of the fibrous and foam-like samples were similar, i.e., \( k_{e,\text{fib}}/k_{e,\text{foam}} = k_{e,fib}/k_{e,\text{foam}} = 0.95-1.05, \sigma_{e,\text{fib}}/\sigma_{e,\text{foam}} = \sigma_{e,fib}/\sigma_{e,\text{foam}} = 2-25 \), with the largest

differences for high porosities. The decreased effective electronic and thermal conductivities of the solid phase of the fibrous samples were three orders of magnitude larger than \( K \) of the fibrous samples. \( F \) differs by almost two orders of magnitude. The samples’ permeabilities can be accurately (RMS = 2%) described by exponential functions in porosity \( (K = a\exp(b\varepsilon)) \), i.e., lumping all the other morphological characteristics into two constants.

Table I. \( \tau_{\text{mean}} \) of the fiber and foam-like samples is given in Table I. \( \tau_{\text{mean,fiber}} > \tau_{\text{mean,foam}} \), especially at lower porousities as dead ends and recirculation zones were more frequent. Consequently, the effective (hydrogen) diffusivity of the foam-like samples was larger than the effective diffusivity of the fibrous sample at similar porosities.

**Application to Solar Fuel Processing and Solar Thermal Power Generation**

The pure porosity-dependent absorption behavior of the fibrous and foam-like samples (original and shrunken) with \( r = 0.1 \) is depicted in Fig. 6. The foam-like samples with largest porosity reached maximal absorptance \( (T < 0.005) \) within \( t/d_{\text{void,foam}} \geq 7 \), while the fibrous samples required \( t/d_{\text{void,fiber}} \geq 5 \). The maximal absorption achievable was only dependent on the sample porosity and not the sample morphology \((A_{\text{fiber}} - A_{\text{foam}})/A_{\text{fiber}} \leq 0.3\% \). The fibrous and the shrunken foam samples achieved the maximal achievable \( A \) within a sample thickness of only a few hundred microns; the original foam sample required thicknesses larger than 20 cm.
**Photoelectrochemical Fuel Processing**

We used Pt and RuO$_2$ catalysts for the hydrogen and oxygen evolution reactions with $i_{0,a} = 10^{-3} \text{A/cm}^2$, $i_{0,c} = 10^{-3} \text{A/cm}^2$, $A_a = 35 \text{mV/dec}$, and $A_c = 30 \text{mV/dec}$. The area of the porous slab modeled was $25 \text{cm}^2$. The solar irradiation was $1 \text{kW/m}^2$, collimated and 1.5 AM. The energy band gaps of the dual absorber were 1.6 eV and 1.0 eV, corresponding to a current matching absorber combination. The ambient temperature was assumed ($T = 300 \text{K}$) and the hydrogen saturation concentration in water was $0.78 \text{mol/m}^3$. We used $\kappa_L = 40 \text{S/m}$, which corresponds to the ionic conductivity of 1 M sulfuric acid. $\sigma_a$ varied significantly depending on the materials and their doping; we assumed $\sigma_a = 1 \text{S/m}$.

The efficiency of the multifunctional porous slab used for photoelectrochemical fuel production using the two different base morphologies (fiber and shrunken foam) and four different porosities is given in Fig. 7. The initial gain in efficiency resulted from the increase in absorbed solar radiation. The decrease in efficiency for larger thicknesses was a result of the dominance of the ohmic losses, especially the losses due to conduction in the solid absorber phase. The decrease in efficiency was observed at smaller sample thicknesses for the fibrous samples than the shrunken foam samples as $\sigma_{e,fiber} < \sigma_{e,foam}$. For samples with larger porosities, the thickness at which the ohmic losses started to dominate did not yet allow for complete radiation absorption, and consequently, the observed maximal efficiency was significantly lower than the maximal achievable efficiency for maximal absorption. The absorption-limited efficiency was only reached for the fibers with $e = 0.46$ and the shrunken foam samples with $e = 0.46–0.719$.

**Solar Thermal Power Production** A sample of area $0.503 \text{m}^2$ was irradiated by a collimated solar flux of $1000 \text{kW/m}^2$. Air was the heat transfer fluid assumed, modeled as an ideal gas with temperature-dependent properties and inlet mass flow of $\dot{m} = 0.4 \text{kg/s}$, initial temperature of 298 K, and an absolute pressure of 10 bars. These boundary conditions were an estimate of a 1 MW pressurized air

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**Table I. Mean tortuosity for the fiber and original and shrunken foam-like samples for various porosities**

| $e$  | 0.84 | 0.72 | 0.59 | 0.46 |
|------|------|------|------|------|
| $\tau_{\text{mean,fiber}}$ | 1.269 | 1.394 | 1.550 | 1.746 |
| $\tau_{\text{mean,foam}}$ | 1.150 | 1.234 | 1.342 | 1.482 |

---

**Fig. 5. Permeability and Forchheimer coefficient for fibrous (solid line) and original and shrunken foam-like (dotted, dashed) samples for varying porosities.**

**Fig. 6. (a) Absorptance and (b) reflectance as a function of layer thickness for fibrous (solid lines), original (dotted lines), and shrunk (dashed lines) foam-like samples with $r = 0.1$ and varying porosity.**
receiver for solar-driven gas turbines. Sintered silicon carbide was used as the solid absorber with $r = 0.1$. The efficiency of the multifunctional 1-D slab used for solar thermal power production for the three different morphologies and four different porosities is given in Fig. 8. The fibrous samples reached the maximal absorptance for slab thicknesses of several hundred microns, and the highest efficiency was achieved for samples with highest porosities as their reflectance were the smallest. The shrunken foam-like samples reached their maximum efficiencies within the first 100 μm due to the larger extinction coefficients; however, a higher reflectance for low porosity led to slightly lower maximum values. Increasing the sample thickness led to a decrease in efficiency because the pressure losses through the slab became significant. The influence of the pump work onto the efficiency of the fibrous samples was less significant but still apparent. In contrast, the original foam-like samples showed no decrease in efficiency with increasing thickness. The original foam samples at comparable porosity required larger sample thicknesses to reach the maximal efficiency due to significantly lower extinction coefficients. The maximum efficiency of a fibrous sample and the shrunk foam-like samples was always larger compared to the original foam-like sample with the same porosity mainly due to their larger specific surface area and enhanced convective heat transfer.

**CONCLUSIONS**

A multiscale experimental–numerical methodology has been used to quantify the gain in performance due to engineering of the morphology of porous media used as absorber, heat exchanger, charge conductor, and reaction site in solar reactors. Two base morphologies have been investigated, i.e., fibrous and foam-like samples. Their exact morphologies were experimentally obtained via x-ray computed tomography and subsequently manipulated by digital image processing to vary characteristic morphological sizes and porosity, both at a constant base morphology. The obtained morphologies (fibrous, original, and shrunk foam samples) were used in direct numerical simulations for the determination of their effective heat, mass, and charge transport properties, which were used in continuum-scale models of reactors incorporating 1-D multifunctional porous slabs performing photoelectrochemical hydrogen generation and solar thermal power production.

We showed that heat, mass, and charge transport in porous media cannot accurately be described based on only two morphological characteristics, e.g., porosity and a characteristic length. For the particular cases of fibrous, original foam, and shrunk foam-base morphologies, we determined the porosity-dependent effective extinction coefficients, effective scattering phase functions, permeabilities and Forchheimer coefficients, effective diffusivities and tortuosity distributions, Nu correlations, and effective ionic, electronic, and thermal conductivities.

The developed continuum-scale models of reactors incorporating 1-D slabs of porous media used in a photoelectrochemical hydrogen generation process and a solar thermal power production process were used to quantify the influence of morphology on process performance. We observed that the morphology leading to optimized efficiencies was
dependent on the application of the porous media. The efficiency of photoelectrochemical fuel processing could be increased by a factor of 23 when using a shrunken foam morphology instead of a fibrous morphology at high porosities. The efficiency of solar thermal power production increased by a factor of 1.2 and the required slab thickness was reduced by a factor of 12 when using a highly porous fibrous sample with fiber diameters in the \( \mu \text{m} \) range instead of a low porosity foam sample with pore diameters of a few mm. The study showed—in the limit of the validity of the continuum model’s assumptions—that the morphology, scale, and porosity of the multifunctional porous components incorporated in solar reactors significantly influence their performances.

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