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A multiscale model for carbon adsorption of BTX compounds: Comparison of volume averaging theory and experimental measurements

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HIGHLIGHTS

- Analysis of nonlinear sorption in a multiscale porous medium.
- Two levels of upscaling described.
- Model verification by comparison with experimental data from packed columns.

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ABSTRACT

In this work, the method of volume averaging is applied for the mathematical modeling of transport and adsorption of benzene, toluene, and xylene in a packed bed of activated particles. One benefit of this approach is that it allows one to directly incorporate measured microscale information into macroscale models for predicting the effective transport and adsorption process. This work is novel in that it combines two levels of upscaling and a nonlinear adsorption process. The first level of upscaling develops an effective model for the mass transport and reaction in an activated carbon particle; within the particle, only diffusion and reaction are considered because of the very small pore sizes. The second level of upscaling develops the effective model for a collection of carbon particles forming the porous medium contained in a fixed-bed reactor; here, convection, diffusion, and dispersion are considered. The resulting model resembles a classical *mobile-immobile* representation of the transport and adsorption process.

As part of the upscaling process, we develop the homogenized transport equations and their associated effective parameters using two different averaging volume support scales (i.e., at two disparate length scales). The effective parameters are all diffusion or dispersion tensors. These include (1) the effective diffusion tensor defining diffusion in the homogenized carbon particle, (2) the effective diffusion tensor for the immobile phase in the two-region representation of the medium, and (3) the effective hydrodynamic dispersion tensor (which included diffusion and dispersion) for the mobile region of the porous catalyst bed. Each of these effective parameters are determined by numerically solving closure problems over an idealized spatially periodic model of a porous medium. One novel feature of these particular closure problems is that they describe *nonlinear adsorption* at the microscale, which is a problem that is not currently represented in the literature.

Once derived, the two-scale, two-equation mobile-immobile model was applied to predict experimentally-measured concentration breakthrough curves from packed bed columns with activated carbon from coconut shell as the adsorbent to the removal of petrochemical contaminants (BTX) by adsorption. There were no adjustable parameters in this modeling effort; the only modeling choice

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Nomenclature

Roman letters

a_v	$=\frac{A_{\gamma\kappa}}{V_1}$ area per unit volume in the carbon particles (m^2/m^3)
$\mathscr{A}_{\gamma\kappa}$	the domain comprising the solid-fluid interface within an averaging volume $\mathscr{V}$ (m^2)
$A_{\gamma\kappa}$	the magnitude (area) of the solid-fluid interface within an averaging volume $\mathscr{V}$ (m^2)
$\mathscr{A}_{\gamma e}$	the domain comprising the area of intersection between the fluid and the boundaries of the averaging volume $\mathscr{V}_\omega$ (m^2)
$\mathscr{A}_{\beta\sigma}, \mathscr{A}_{\sigma\beta}$	the domain comprising the solid-fluid interfacial area within an averaging volume $\mathscr{V}$ (m^2)
$A_{\beta\sigma}, A_{\sigma\beta}$	the magnitude (area) of the solid-fluid interface within an averaging volume $\mathscr{V}$ (m^2)
$\mathscr{A}_{\beta e}, \mathscr{A}_{\sigma e}$	the domain comprising the area of intersection between the fluid and the boundaries of the volume $\mathscr{V}$ (m^2)
B_A	adsorption equilibrium constant (m^3/kg)
$\mathbf{b}_{\beta\beta}, \mathbf{b}_{\sigma\sigma}$	closure variable (field) (m)
$\mathbf{b}_\gamma$,	closure variable for the first level of upscaling (m)
c_{As}	surface concentration of species A in the κ-phase (internal surface area of carbon particles) (kg/m^2)
$c_{A\gamma}$	concentration of species A in the γ-phase (fluid phase) (kg/m^3)
$c_{A\beta}$	concentration of species A in the β-phase (fluid phase) (kg/m^3)
$c_{A\sigma}$	concentration of species A in the σ-region (carbon phase) (kg/m^3)
d_p	particle diameter (m)
d_c	column diameter (m)
$\mathscr{D}_{A\gamma}, \mathscr{D}_{A\beta}$	molecular diffusion coefficients for chemical species A in the fluid phases (m^2/s)
$\mathbf{D}_{A\sigma}$	Effective (macroscale) diffusion coefficient arising from the first level of upscaling for species A in the porous medium (m^2/s)
$\mathbf{D}^*_{A\beta\beta}$	Total dispersion tensor for species A arising from the second level of upscaling (m^2/s)
$\mathbf{D}^*_{A\beta\sigma}$	$=\mathbf{D}^*_{A\sigma\beta}$, coupling dispersion tensors for species A arising from the second level of upscaling (m^2/s)
$\mathbf{D}^*_{A\sigma\sigma}$	Effective diffusivity tensor for species A arising from the second level of upscaling (m^2/s)
$\mathscr{F}_{A\gamma}$	generic condition at the boundary $\mathscr{A}_{\gamma e}$ (kg/m^3)
$\mathscr{F}_{A\beta}$	generic condition at the boundary $\mathscr{A}_{\beta e}$ (kg/m^3)
$\mathscr{G}_{A\sigma}$	generic condition at the boundary $\mathscr{A}_{\sigma e}$ (kg/m^3)
h	mass transfer coefficient for the second level of upscaling (m/s)
h_{Wakao}	mass transfer coefficients in the liquid film (correlation Wilson and Geankoplis (1966)), second level of upscaling (m/s)
h_{Wilson}	mass transfer coefficients in the liquid film (correlation Wakao and Funazkri (1978), Wakao and Funazkri (1966)) (m/s), second level of upscaling (m/s)
$\mathscr{I}_{A\gamma}$	generic initial for the γ-phase (kg/m^3)
$\mathscr{I}_{A\beta}$	generic initial condition for the β-phase (kg/m^3)
$\mathscr{I}_{A\sigma}$	generic initial for the σ-phase (kg/m^3)
K_A	equilibrium adsorption coefficient (Linear isotherm) (m)
K^*_A	$=\frac{a_v\vert_{\gamma\kappa}K_A}{\varepsilon_\gamma}$ dimensionless adsorption equilibrium coefficient for the σ-region
ℓ_γ	characteristic length of the internal structure of the microporous carbon particles (m)
ℓ_β, ℓ_σ	characteristic lengths of the macroscale structure of the porous medium (composed of a packed bed of carbon particles) (m)
ℓ_j	lattice vector defining the structure of a periodic array (m)
L_p	characteristic length of the macroscale concentration filed (roughly, the carbon particle size) for the first level of upscaling (m)
L_ε	characteristic length of the macroscale porosity field (roughly, the carbon particle size) for the first level of upscaling (m)
L_c	characteristic length of the macroscale (roughly, the reactor dimension) for the second level of upscaling (m)
$\mathbf{n}_{\gamma\kappa}, \mathbf{n}_{\beta\sigma}$	unit normal vectors pointing from the fluid γ, β-phases toward the solid κ, σ-phases (dimensionless)
$\mathbf{O}$	symbol indicating an order-of-magnitude estimate
$q_{\max,A}$	maximum adsorption capacity (Langmuir isotherm) (kg/m^2)
Q	fluid flow rate (m^3/s)
$\mathbf{r}$	position vector (m)
r_1	radius of the averaging volume on the first level of averaging (m)
r_2	radius of the averaging volume on the second level of averaging (m)
Re_p	particle Reynolds number (dimensionless)
Sh	Sherwood number (dimensionless)
Sc	Schmidt number (dimensionless)
t_β, t_σ	closure variable (scalar) associated with the mass-transfer term in the second level of upscaling (–)
t^*	characteristic time scale for the microscale process (s)
T^*	characteristic time scale for the macroscale process (s)
T	temperature (°C)
$\mathbf{u}_{A\beta}$	dimensionless tensor convective transport in β-phase for species A (dimensionless)
$\mathbf{u}_{A\sigma}$	dimensionless tensor convective transport in σ-phase for species A (dimensionless)
$\mathbf{v}_\beta$	microscale fluid velocity (m/s)
$\langle\mathbf{v}_\beta\rangle^\beta$	intrinsic average velocity (m/s)
$\tilde{\mathbf{v}}_\beta$	$=\tilde{\mathbf{v}}_\beta+\langle\mathbf{v}_\beta\rangle^\beta$ deviation from the average velocity (m/s)
$\mathscr{V}_1$	The averaging domain for the first level of the averaging process. $\mathscr{V}_1=\mathscr{V}_\gamma\cup\mathscr{A}_{\gamma\kappa}\cup\mathscr{V}_\kappa$
V_1	The magnitude (volume) of $\mathscr{V}_1$ (m^3)
$\mathscr{V}_\gamma$	domain of the γ-phase in the averaging volume $\mathscr{V}_1$ (m^3)
V_γ	magnitude (volume) of $\mathscr{V}_\gamma$ (m^3)
$\mathscr{V}_\kappa$	domain of the κ-phase in the averaging volume $\mathscr{V}_1$ (m^3)
V_κ	magnitude (volume) of $\mathscr{V}_\kappa$ (m^3)
$\mathscr{V}_2$	The averaging domain for the second level of the averaging process. $\mathscr{V}_2=\mathscr{V}_\beta\cup\mathscr{A}_{\beta\sigma}\cup\mathscr{V}_\sigma$
V_2	magnitude (volume) of the averaging volume at the second level of upscaling (m^3)
$\mathscr{V}_\beta$	domain of the β-phase in the averaging volume $\mathscr{V}_2$ (m^3)
V_β	magnitude (volume) of $\mathscr{V}_\beta$ (m^3)
$\mathscr{V}_\sigma$	domain of the σ-phase in the averaging volume $\mathscr{V}_2$ (m^3)
V_σ	magnitude (volume) of $\mathscr{V}_\sigma$ (m^3)
(x_1, x_2, x_3)	Cartesian coordinate system
$\mathbf{x}$	position vector locating the centroid of the averaging volume, (m)
$\mathbf{y}_x$	position vector locating points relative to the centroid in the x phase or region, (m)