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Heat and mass transfer model of multicomponent adsorption system for hydrogen purification☆

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ABSTRACT

Pressure swing adsorption (PSA) is an excellent way for separation and purification of gas mixtures. The mass and energy equations are derived from thermodynamics conservation laws. A multicomponent adsorption model is developed for simulating the breakthrough curves of the mixtures of H_2/CO/CO_2/CH_4 and of H_2/CO_2 in the activated carbon (AC) bed of hydrogen purification system. The one-dimensional model is developed and implemented on Comsol platform. The model simulates molar fraction evolutions of four components and temperature profiles of the bed and the wall at the axial locations of 20 cm, 70 cm and 110 cm. The model is validated by experimental data and improved by considering a piping system. Parametric studies are made to investigate the influence of the system pressure, gas filling velocity and gas mixture composition on the performance of hydrogen purification system, and to propose guidelines for optimal design of PSA system for hydrogen purification.

Introduction

Purification of hydrogen is closely related to its production, storage, transport and utilization, which is of important significances for safe, efficient, reliable and durable operation of hydrogen energy utilization systems. The thermal effects caused by exothermic and endothermic phenomena in adsorption and desorption processes have significant impacts on performances of charge–discharge (adsorption-desorption) cycle in hydrogen storage system.

Chang-Ha Lee believes that the thermal phenomena have significant effects on the adsorption kinetics of hydrogen purification [1]. Therefore, he uses heat exchange to solve the thermal effects in adsorption process [2]. The results show that the carbon bed temperature has significant impacts on the concentration curves of carbon monoxide and methane [1]. By designing a heat exchange PSA (HE-PSA: Heat Exchange Pressure Swing Adsorption) system, the outer bed is isothermal, and the inner bed can reduce the adverse thermal effects [2]. Young-Woo You established two beds for PSA purification to produce high purity hydrogen for fuel

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Nomenclature

A_w	cross-section area of column wall, m^2
c	molar concentration of mixture, mol/m^3
c_i	molar concentrations of component i, mol/m^3
c_{pg}	specific heat capacity of gas phase, J/kg/K
C_{pg}	specific heat capacity of gas phase, J/mol/K
c_{ps}	specific heat capacity of adsorbent, J/kg/K
c_{pw}	specific heat capacity of column wall, J/kg/K
h_{in}	heat transfer coefficient with inner wall of column, $W/m^2/K$
h_{out}	heat transfer coefficient with outer wall of column, $W/m^2/K$
Δh_i	heat of adsorption of component i, J/kg
ΔH_i	heat of adsorption of component i, J/mol
k_i	mass transfer coefficient of component i , 1/s
M	molar-averaged molecular weight of mixture, kg/mol
M_j	molecular weight of component i, kg/mol
n_i	dynamic adsorption of component i, mol/kg
n_i^*	equilibrium adsorption of component i, mol/kg
n_i^s	saturation adsorption of component i, mol/kg
p	pressure, mmHg, atm, Pa
q_i	dynamic adsorption mass of component i, kg/kg
R	universal gas constant, 8.314 J/mol/K
R_{in}	inner radius of column, m
R_{out}	outer radius of column, m
s_m, s	mass source term of mixture, $kg/s/m^3$
s_i	mass source term of component i, $kg/s/m^3$
S_i	molar source term of component i, $mol/s/m^3$
t	time, s
T	temperature of adsorption bed, K
T_f	ambient temperature, K
T_w	wall temperature, K
$\vec{u}$	mass-averaged physical velocity, m/s
$\vec{U}$	molar-averaged physical velocity, m/s
U_z	axial velocity in adsorption bed, m/s
$\vec{v}$	Darcy velocity, m/s
x_i	mass fraction of component i
y_i	molar fraction of component i
z	axial position in the bed, m

Greek symbols

ε_b	bed porosity
ρ, ρ_g	mass concentration (density) of mixture gas, kg/m^3
ρ_i	mass concentrations of component i, kg/m^3
ρ_b	bed density of adsorbent, kg/m^3
ρ_p	particle density of adsorbent, kg/m^3
ρ_s	skeletal density of adsorbent, kg/m^3
ρ_w	density of column wall, kg/m^3

cell applications, therefore the simulation results of temperature did not have good agreement with the experimental data [3].

Breakthrough curves are good reflections of thermal effects, so the study for the thermal effect of PSA should start from breakthrough curves. Lopes FVS has carried out the activated carbon bed in H_2/CO_2, $H_2/CO/CO_2$ and $H_2/CO/CO_2/CH_4/N_2$ multicomponent gases through experiments, which is applied to verify the mathematical model. Also he has carried out a 10-step VPSA (Vacuum Pressure Swing Adsorption) experiment to verify the simulation results of the mathematical model under the condition of the circulation work [4]. Casas N studied breakthrough curve experiment of two-component mixture H_2/CO_2 in fixed bed filled with activated carbon. A constant isosteric heat of adsorption is used in his mathematical model and leads to the peak of simulated temperature curve much lower than the experimental value, so it should be replaced by a variable heat of adsorption (especially for CO_2) [5].

In the numerical simulation of thermal effect of the breakthrough curve, the concentration curve is consistent with that obtained from the experiment, but the temperature curve has big difference with the experimental data [3–5]. Pressure swing adsorption (PSA) experiments are carried out in a 2-column laboratory setup using activated carbon, in which an equimolar CO_2/H_2 mixture is used as feed. It is shown that the temperatures measured inside the columns provide an excellent possibility for comparison of experiments and simulations [6]. Joss L shows the importance of an accurate characterization of the extra-column volume and presents an experimental and computational protocol based on the characterization of the extra-column volume in terms of step-response experiments performed under various flow rates and pressures [7]. East China University of Science and Technology measured the axial temperature distribution in the process of hydrogen purification adsorption and desorption by a thermal imager. In addition, the finite difference method is used for numerical simulation. The simulated temperature is consistent with the experiment, but there is still some space for improvement by optimizing the heat transfer boundary conditions [8].

In this work, we established the heat and mass transfer model for multicomponent adsorption of hydrogen purification system. After the model is validated and improved by breakthrough curve experiments [9], the model of multicomponent gas flow, heat and mass transfer and adsorption kinetics is applied for parametric study. The influence of bed pressure, gas velocity and mixture composition on hydrogen purification performance is studied.

Model development

Molar and mass concentrations based on ideal gas equation of state

Ideal gas equation of state is used to describe the gas phase behavior. Molar concentrations of components are (mol/m^3):