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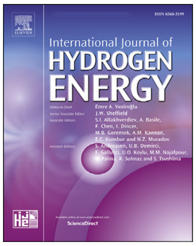

Hydroxyl aluminium silicate clay for biohydrogen purification by pressure swing adsorption: Physical properties, adsorption isotherm, multicomponent breakthrough curve modelling, and cycle simulation

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ABSTRACT

Hydroxyl aluminium silicate clay (HAS-Clay) is a novel adsorbent in pressure swing adsorption for CO_2 capture (CO_2-PSA) and can also adsorb H_2S. To investigate the performance of HAS-Clay as a CO_2-PSA adsorbent, multicomponent breakthrough curves were determined using experimental measurements and theoretical models, and, based on those results, CO_2-PSA simulations were conducted. The breakthrough curves produced from the theoretical models agreed well with those derived from experiment. CO_2-PSA with HAS-Clay could purify biomass-gasification-derived producer gas of contaminants (carbon dioxide, methane, carbon monoxide, and hydrogen sulfide) with high CO_2 recovery and low energy input. The CO_2 recovery rate of CO_2-PSA with HAS-Clay was 58.4%, and the CO_2 purity was 98.4%. The specific energy demand was 2.83 MJ/kg-CO_2. In addition, the H_2S regenerability of HAS-Clay was investigated. The results show that HAS-Clay retained the ability to adsorb H_2S at a steady-state value of 0.02 mol/kg for the regeneration cycles. Therefore, it is suggested that CO_2-PSA with HAS-Clay is suitable for CO_2 separation from multicomponent gas mixtures.

Introduction

Hydrogen (H_2) is attracting attention as a clean, abundant, and storable energy source. The combustion of hydrogen emits no air pollutant such as carbon dioxide (CO_2), sulfur dioxide (SO_2), and nitrogen oxide (NO_x), and hydrogen is generally stored in high-pressure gas vessels or solid metal hydrides. Because of its low molecular weight, hydrogen has a high energy density, making it suitable as an alternative transport fuel [1] and other

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Nomenclature

A_w	cross sectional area of the wall [m^2]
$a_{i,1}$	adsorption equilibrium constant of species *i* [mol/kg]
$a_{i,2}$	adsorption equilibrium constant of species *i* [mol/kg·K]
b_i	adsorption equilibrium constant of species *i* [kPa^{-1}]
$b_{\infty,i}$	adsorption constant of component *i* at infinite temperature [kPa^{-1}]
C_i	concentration of component *i* [$kmol/m^3$]
C_{H_2S}	H_2S concentration [ppm]
$C_{p,ads}$	specific heat capacity of adsorbent [J/kg·K]
$C_{p,w}$	specific heat capacity of wall substance [J/kg·K]
d_{bed}	vessel diameter [m]
d_p	pellet diameter [m]
D_{ax}	diffusion coefficient [m^2/s]
$D_{e,i}$	effective diffusion coefficient [m^2/s]
$D_{k,i}$	Knudsen diffusion coefficient [m^2/s]
$D_{m,i}$	molecular diffusion coefficient [m^2/s]
F	mass flow [kg/s]
h	fluid phase mass specific enthalpy [kJ/kg]
$-\Delta H_i$	heat capacity of compound *i* [J/mol]
k_i	mass transfer coefficient for LDF [1/s]
$k_{T,b-w}$	heat transfer coefficient [$W/m^2 \cdot K$]
$k_{T,w-a}$	heat transfer coefficient from bed wall to environment [$W/m^2 \cdot K$]
l_w	bed wall thickness [m]
M_{w_i}	molecular weight of species *i* [g/mol]
P	pressure [kPa]
q_i	particle average adsorbed concentration [mol/kg]
$q_{eq,i}$	adsorbed concentration as predicted by the isotherm [mol/kg]
$q_{m,i}$	specific saturation adsorption capacity of species *i* [mol/kg]
r_p	particle radius [m]
R	ideal gas constant [J/K mol]
Re	Reynolds number [–]
Sc_i	Schmidt number [–]
S_{cap}	saturation capacity [mol/kg]
t_{BT}	breakthrough time [min]
T	bed fluid temperature [K]
T_a	ambient temperature [K]
T_w	bed wall temperature [K]
v	fluid phase superficial velocity [m/s]
$\dot{V}$	flow rate of the gas containing H_2S [L/min]
V_m	molar volume (22.4 l/mol under standard conditions) [L/mol]
$W_{sorbent}$	adsorbent weight [g]
w_i	mass fraction for component *i* [–]
ε_{bed}	bed void fraction [–]
ε_p	pellet void fraction [–]
ε_{tot}	total void fraction [–]
μ	gas viscosity [Pa·S]
λ_{eff}	effective thermal conductivity [W/m·K]
λ_w	thermal conductivity of the wall [W/m·K]
ρ	fluid phase mass density [kg/m^3]
ρ_{bed}	bed bulk density [kg/m^3]
ρ_w	mass density of the wall material [kg/m^3]
τ	pore tortuosity [–]

uses requiring fuel portability [2]. Hydrogen is obtained from many resources via different conversion technologies (e.g., steam methane reforming [3], water electrolysis [4], and biomass gasification [5]). Among such conversion technologies, biomass gasification is one of the most promising technologies for hydrogen generation [6]. Biomass is a sustainable resource and is clean, renewable, and abundant [7].

From biomass gasification, the generated producer gas contains hydrogen and other impurities including carbon monoxide (CO), CO_2, methane (CH_4), and traces of hydrogen sulfide (H_2S, ca. 20–230 ppm [8]). In particular, H_2S removal is required to prevent catalyst poisoning, that often causes voltage reduction in fuel cells and shortens the catalyst lifetime. The criteria for hydrogen quality are standardised as $H_2 > 99.97\%$, $CO_2 < 2.0$ ppm, $CO < 0.4$ ppm, and $H_2S < 0.004$ ppm [9].

An effective method for removing the impurities is pressure swing adsorption (PSA) because of its high economic performance. PSA for H_2 purification usually uses multiple beds, and the adsorption and desorption operations are carried out simultaneously [10–13]. Each bed has a series of layers of different adsorbents. The first layer usually removes water vapour, commonly using activated alumina or silica gel, followed by a second layer of activated carbon, which adsorbs CO_2. The third layer removes the lighter impurities such as CO and CH_4. The adsorbent selectivity has a great impact on the purification efficiency, as well as the operating pressure and temperature. In particular, producer gas compression power during PSA accounts for a large portion of the auxiliary power consumption in biomass-to-hydrogen processes, suggesting that lower operating pressures are required for utility power reduction [14].

Low-pressure operation has been recently achieved using a new adsorbent, hydroxyl aluminium silicate clay (HAS-Clay) [15]. HAS-Clay is an amorphous aluminium hydroxide silicate ($SiO_2/Al_2O_3/H_2O$) that has excellent CO_2 adsorptivity and can possibly also be used for H_2S adsorption. It has been suggested that the operating pressure can be reduced from 700 to 400 kPaG if CO_2 is pre-separated by HAS-Clay [15]. Another strong point is that HAS-Clay could play a role as H_2S adsorbent during its use as a PSA adsorbent for bio-H_2 purification. Therefore, the H_2S adsorption performance of HAS-Clay for producer gas cleaning should be investigated.

In this study, the performance of HAS-Clay as an adsorbent for CO_2-PSA was investigated: (1) the physical properties and adsorption isotherm of HAS-Clay were experimentally determined, followed by theoretical modelling to obtain multicomponent breakthrough curves, (2) the multicomponent