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Short communication

Prediction of fixed-bed breakthrough curves for H_2S adsorption from biogas: Importance of axial dispersion for design

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HIGHLIGHTS

- Modeling of the fixed-bed adsorption with axial dispersion for H_2S removal was simulated by Comsol.
- The effects of mass transfer resistance and axial dispersion on breakthrough curves were obtained.
- Axial dispersion is key to avoid underestimating the overall mass transfer coefficient for design.

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ABSTRACT

An axially dispersed plug flow model with non-linear isotherm based on the linear driving force (LDF) approximation was used to predict the fixed-bed breakthrough curves for H_2S adsorption from biogas on sewage sludge thermally treated. The model was implemented and solved numerically by Comsol Multiphysics software. The predicted breakthrough curves matched very well the experimental data and were clearly better than those predictions obtained in our previous work by Aspen Adsorption assuming ideal plug flow. The comparison between the present and previous models, as well as a sensitivity analysis of the model and operational parameters, revealed that the overall mass transfer coefficient is usually underestimated when axial dispersion is neglected in a scale-up from lab scale, and hence, the importance of axial dispersion for design purposes of H_2S fixed-bed adsorption.

1. Introduction

Biogas, produced from anaerobic digestion of organic matter, is an attractive alternative energy source as it is methane-rich. However, it is necessary to remove hydrogen sulfide from the biogas before it can be used in engines for electricity generation to prevent the equipment corrosion and the formation of sulfur oxides when combusted. Desulfurization of biogas can be successfully performed through fixed bed adsorption. Low-cost sewage sludge-based adsorbents have been produced by several thermal treatments, and then tested, characterized and modeled [1,2].

Two modeling approaches of the dynamic behavior of a fixed bed were used to predict the breakthrough curves, neglecting the effects of axial dispersion, in our previous paper [2]. The first approach was based on the Bohart–Adams model, and the second one used the linear driving force model (LDF), solving it analytically by Klinkenberg equation, with a linear isotherm, and numerically, with a non-linear isotherm, by Aspen Adsorption.

Some mathematical models to predict the adsorption fixed bed dynamics have been published over the years [3]. Their development has been described in terms of partial differential equations, with a very difficult numerical solution and extensive experimentation to determine the model parameters. Thus, the accurate and efficient simplification of the model and the use of empirical correlations are continuously searched to reduce the required computational time.

Comsol Multiphysics software has been recently used to compute different fixed-bed adsorption models for both simple models [4,5] and more complex models [6,7], focusing on a few applications such as adsorption of CO_2, biosorption of metals, adsorption of volatile organic compound, or ethanol. However, to our knowledge, there are no previous studies of modeling and simulation of breakthrough curves for hydrogen sulfide adsorption from biogas using Comsol Multiphysics software.

Thus, with the aim of revealing the relative importance of axial dispersion for design purposes, an axially dispersed plug flow model with non-linear isotherm based on the linear driving force (LDF) approximation was implemented and solved numerically by Comsol Multiphysics software to predict the fixed-bed

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Nomenclature

C	H_2S concentration in gas phase (mol/m^3)
C_0	H_2S feed concentration (mol/m^3)
d_p	mean particle diameter (m)
D_e	effective diffusivity (m^2/s)
D_g	distribution coefficient (−)
D_m	molecular diffusivity (m^2/s)
D_z	axial dispersion coefficient (m^2/s)
k_g	external mass transfer coefficient (m/s)
K_G	overall mass transfer coefficient (s^{-1})
K_L	equilibrium constant of Langmuir (m^3/mol)
K_F	coefficient of the Freundlich isotherm (mol/kg) (m^3/mol)^n
l	dimensionless distance from the bed entrance (−)
L	total bed depth (m)
n	exponent in the Freundlich isotherm (−)
Pe	Peclet number (−)
q	concentration of H_2S (or H_2S loading) in solid phase (mol/kg)
q^*	value of q in equilibrium with C (mol/kg)
q_s	saturation value of q in the Langmuir isotherm (mol/kg)
Re	Reynolds number (−)
R_p	particle radius (m)
S	dimensionless mass transfer coefficient (−)
Sc	Schmidt number (−)
Sh	Sherwood number (−)
t	time (s)
v	gas interstitial velocity (m/s)
x	dimensionless concentration of H_2S in gas phase (−)
y	dimensionless concentration of H_2S in solid phase (−)
z	distance from the bed entrance (m)
Greek	
ε	bed void fraction (−)
ρ_b	adsorbent bulk density (kg/m^3)
τ	dimensionless time (−)

breakthrough curves for H_2S adsorption. Simulation results were validated, by contrasting them to experimental results, and compared with previous simulations results, where ideal plug flow was assumed [2]. In addition, the influence of the variables involved in the process, based on the mathematical model developed, was studied by performing a sensitivity analysis.

2. Mathematical model

The mathematical model used to describe the dynamics of adsorption for this system considers nonlinear adsorption isotherm. The axially dispersed plug flow model describes the flow pattern, and the overall mass transfer rate is represented by the linear driving force (LDF) approach.

Isothermal conditions, negligible radial dispersion and changes in fluid velocity, and spherical and homogeneous in size and density adsorbent particles (bed void fraction constant) are assumed. All the symbols and units used are detailed in the Nomenclature.

The differential mass balance for the adsorbate in the fixed-bed includes axial dispersion, convection flow, accumulation in the fluid phase, and adsorption rate on the particles (Eq. (1)):

$$-D_z\frac{\partial^2 C}{\partial z^2}+v\frac{\partial C}{\partial z}+\frac{\partial C}{\partial t}+\frac{(1-\varepsilon)}{\varepsilon}\frac{\partial q}{\partial t}=0 \tag{1}$$

where C represents the H_2S concentration in gas phase, v is the gas interstitial velocity, ε is the bed void fraction, q is the concentration of H_2S in solid phase, t is the time, z is the distance from the bed entrance, and D_z is the axial dispersion coefficient, calculated from the correlation of Wakao and Funazkri [8] (Eq. (2)).

$$\frac{\varepsilon D_z}{D_m}=20+0.5ScRe \tag{2}$$

where Sc and Re are the Schmidt and Reynolds numbers, respectively, and D_m is the molecular diffusivity of H_2S, calculated by the Fuller–Schettler–Gridding correlation.

In the adsorption process, the transport rate of the adsorbate from the fluid phase to the inner surface of the solid is the controlling step when the adsorption rate is fast enough. The LDF model (Eq. (3)) represents this mass transfer rate by the overall mass transfer coefficient (K_G) determined by the Eq. (4):

$$\frac{\partial q}{\partial t}=K_G(q^*-q) \tag{3}$$

$$\frac{1}{K_G}=\frac{R_p q_0^* \rho_b}{3k_g C_0 \varepsilon}+\frac{R_p^2 q_0^* \rho_b}{15 D_e C_0 \varepsilon} \tag{4}$$

where q^* is the value of q in equilibrium with C (H_2S concentration in gas phase) and R_p, q_0^*, D_e and k_g are the particle radius, the equilibrium adsorbate loading for the H_2S initial concentration (C_0), the effective diffusivity (by the Bosanquet equation [2]), and the external film mass transfer coefficient, respectively. The latter was estimated by an empirical correlation (Eq. (5)), recommended when the axial dispersion term is included.

$$Sh=\frac{k_g d}{D_m}=2.0+1.1Sc^{1/3}Re^{0.6} \tag{5}$$

The isotherm parameters were estimated by fitting the experimental data of the H_2S adsorption of previous works [1,2] into the isotherm models (Eqs. (6) and (7)):

$$\text{Freundlich equation}: \quad q^*=K_F\cdot C^n \tag{6}$$

$$\text{Langmuir equation}: \quad q^*=\frac{q_s\cdot K_L\cdot C}{1+K_L\cdot C} \tag{7}$$

where K_L is the equilibrium constant of Langmuir and q_s is the saturation value in the Langmuir isotherm; likewise, K_F and n are the parameters in the Freundlich isotherm.

The initial and boundary conditions are the following:

$$t=0: C=0, q=0 \quad (0\leqslant z\leqslant L);$$
$$z=0: C=C_0 \quad (t>0);$$
$$z=L: \tfrac{\partial C}{\partial z}=0 \quad (t>0).$$

Table 1 shows the dimensionless variables by which Eqs. (1)–(7) were converted into dimensionless Eqs. (8)–(11), presented in Table 2.

Table 1
Dimensionless variables used for Comsol implementation.

Concentration of H_2S in gas phase	$x=\frac{C}{C_0}$
Concentration of H_2S in solid phase	$y=\frac{q}{q_0^*}$
Distance from the bed entrance	$l=\frac{z}{L}$
Time	$\tau=\frac{t\cdot v}{L}$
Distribution coefficient	$D_g=\frac{\rho_b\cdot q_0^*}{\varepsilon\cdot C_0}$
Mass transfer coefficient	$S=\frac{K_G\cdot L}{v}$
Peclet number	$Pe=\frac{L\cdot v}{D_z}$