

Adsorption of phenanthrene on activated carbons: Breakthrough curve modeling

R. Murillo, T. García, E. Aylón, M.S. Callén, M.V. Navarro, J.M. López, A.M. Mastral *

Department of Energy and Environment, Instituto de Carboquímica, CSIC, María de Luna, M Luesma Castan 4, 50015 Zaragoza, Spain

Received 26 January 2004; accepted 5 April 2004

Available online 4 May 2004

Abstract

The modeling of breakthrough curves obtained in the adsorption of phenanthrene as model compound on different activated carbons is described. All the runs were performed in a fixed bed reactor at atmospheric pressure, with a process temperature of 150 °C and low contaminant concentrations. Therefore, experimental conditions were similar to the observed in the flue gases of energy generation systems. This work is mainly focused on the study of how adsorbent characteristics (surface area and micropore size distribution) influence the kinetics of the adsorption process. First, equilibrium values are found from the breakthrough curves and they are satisfactorily fitted to the Freundlich isotherm. Using the obtained equilibrium values together with the linear driving force model as kinetic expression, the breakthrough curve modeling is achieved. It was found that this model fits all the breakthrough curves and it is a useful tool for modeling purposes. Values for the phenanthrene surface and effective diffusion coefficients are calculated and reported, and a relationship with the microporosity is found. As it was expected, it is observed that the phenanthrene molecule finds kinetic restrictions for the diffusion in those carbons with narrow microporosity, especially in those with a mean pore diameter close to the molecular size.

© 2004 Elsevier Ltd. All rights reserved.

Keywords: A. Activated carbon; B. Coke; C. Adsorption properties; D. Modeling

1. Introduction

Polycyclic aromatic hydrocarbons (PAH) are an important family of environmental pollutants that are mainly generated during combustion processes [1–3]. PAH emissions, due to their high volatility and reactivity, can be released, not only supported onto particulate matter (PM) but also in the gas phase [4] downstream the dust collection systems. PAH supported on the PM can be easily trapped using the proper systems such as cyclones, electrostatic precipitators, fabric filters, etc. However, the atmospheric PAH emission control becomes more difficult.

Different technologies for gas-phase emissions control have been developed during the last years [5,6]. The two technologies that are the most promising alternatives to reduce gaseous PAH emissions are: catalytic PAH destruction [7] and PAH adsorption/concentration

on carbonaceous materials [8]. Activated carbons are extensively used to remove contaminants from exhaust gases [9–11]. The gas is generally passed through a bed of activated carbon where the contaminant molecules are transferred to the solid phase (adsorption process). Information about the adsorption of pure organic vapors [12–16] for environmental purposes has been recently published. In these previous works, it was shown that the adsorption of PAH by sorbents from waste hot gas emissions is inversely proportional to their volatility. These studies also indicated that other chemical species in the gas stream like moisture [14] and CO₂ [15] have a negative influence on phenanthrene (Phe) adsorption capacity. First, the determined parameters showed that, for a carbonaceous material, the higher the steam percentage (in volume) in the gas stream, the lower its Phe adsorption capacity; and second, it was also observed the existence of a negative influence of CO₂ in the adsorption capacity of Phe. That is probably due to a competitive effect between CO₂, H₂O and Phe molecules for the primary adsorption sites within the carbonaceous material and also the increase of the surface hydrophilicity.

* Corresponding author. Tel.: +34-976-733-977; fax: +34-976-733-318.

E-mail address: amastral@carbon.icb.csic.es (A.M. Mastral).

Nomenclature

C_0	gas inlet concentration (mol/m ³)	V_{CO_2}	narrow micropore volume (cm ³ /g)
$C(t)$	gas outlet concentration (mol/m ³)	V_{N_2}	total micropore volume (cm ³ /g)
D_e	effective diffusivity (m ² /s)	V_T	total pore volume (cm ³ /g)
D_k	Knudsen diffusivity (m ² /s)	w	carbonaceous material introduced into the reactor (kg)
D_m	molecular diffusivity (m ² /s)	W	equilibrium adsorption capacity (mol/kg)
D_s	surface diffusivity (m ² /s)	W^*	concentration adsorbed on the surface of adsorbent (mol/kg)
K	equilibrium constant	W_0	adsorbate concentration in the solid in equilibrium with the gas inlet concentration (mol/kg)
K_f	Freundlich equation constant	W_{tr}	adsorption capacity at the breakthrough (mol/kg)
k_f	fluid mass-transfer coefficient (m/s)		
k_p	intrapellet mass-transfer coefficient (m/s)	<i>Dimensionless numbers</i>	
L	bed length (cm)	Re	Reynolds number = $\frac{u \cdot \rho_g \cdot R_p}{\mu}$
L_T	height of the mass-transfer zone (m)	Sc	Schmidt number = $\frac{\mu}{\rho_g \cdot D_m}$
M	molecular weight (g/mol)	Sh	Sherwood number = $\frac{R_p \cdot k_f}{D_m}$
n_f	Freundlich equation exponent		<i>Greek symbols</i>
p	nitrogen partial pressure (Pa)	ε	porosity of bed
p^0	nitrogen saturation pressure (Pa)	ε_p	porosity of particle
q	nitrogen adsorption capacity (cm ³ (STP)/g)	ρ_g	gas density (kg/m ³)
Q	total inlet flow (m ³ /s)	ρ_1	bed density (kg/m ³)
r_0	mean pore radius (m)	τ_p	pore tortuosity
R_p	equivalent radius of pellet (m)	τ_s	surface tortuosity
S_{BET}	surface area calculated according to BET isotherm (m ² /g)	μ	gas viscosity (Pa s)
t_f	bed saturation time (s)		
t_r	breakthrough time (s)		
T	temperature (K)		
u	superficial velocity (m/s)		

Up to now, the quantitative studies of PAH adsorption on activated carbons are limited to apply different adsorption isotherm models in a fixed packed bed [17]. However, to improve the industrial design of these adsorbents performance and to find the best operating conditions, the adsorption processes need to be modeled. Numerous models have been published in recent years to describe the phenomena of fixed bed adsorption [18–20]. Model equations are conservation equations (in this case only mass conservation applies), kinetic law at the fluid/solid interface (adsorption equilibrium isotherm), kinetic law of mass transfer, boundary conditions and initial conditions.

Several publications on the adsorption modeling of volatile organic compounds (VOC) on activated carbons and zeolites can be found in the literature [21–24]. In these references, the transport properties of different small organic molecules like acetone, toluene, methyl ethyl ketone, heptane and octane are evaluated fitting the experimental results with the linear driving force (LDF) model. Hence, Brosillon et al. [21] investigated the kinetic performance of zeolites for VOC abatement observing that a constant value of effective diffusivity is found independent of the nature and the amount of

VOC adsorbed. Malek and Farooq [24] studied the kinetic behavior of different small VOC molecules in their adsorption on activated carbon and silica gel finding that the adsorbate transport inside the adsorbent is essentially by Knudsen and surface flow. However, none of these authors have focused on the PAH adsorption modeling and the relationship between kinetic and equilibrium parameters with the structural characteristics of the adsorbent. In this work, the successful application of the LDF model to VOC adsorption is used to predict the breakthrough curves of PAH on activated carbons at low concentrations. In addition, the relationship between kinetic parameters and structural properties is also assessed.

2. Theory

The mathematical model of the isothermal, dynamic adsorption breakthrough process in a fixed bed is based on transient material balance, gas phase and intraparticle mass transfer, the adsorption equilibrium relationship, boundary conditions and initial conditions. In this model, both the adsorption equilibrium and kinetic

Download English Version:

<https://daneshyari.com/en/article/1419357>

Download Persian Version:

<https://daneshyari.com/article/1419357>

[Daneshyari.com](https://daneshyari.com)