



Contents lists available at ScienceDirect

Energy Conversion and Management

journal homepage: www.elsevier.com/locate/enconman

Theoretical and experimental studies of the recovery of volatile organic compounds from waste air streams in the thermal swing adsorption system with closed-loop regeneration of adsorbent

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ARTICLE INFO

Article history:
Available online xxx

Keywords:
Thermal swing adsorption
Volatile organic compounds
Activated carbon
Closed-loop regeneration
Mathematical modelling of TSA processes

ABSTRACT

The cyclic thermal swing adsorption (TSA) process for volatile organic compounds (VOCs) recovery from the waste air is studied theoretically and experimentally. Toluene is chosen as the volatile organic compound. Activated carbon Sorbonorit 4 is used as an adsorbent. The TSA cycle is operated in three steps: an adsorption step with cold feed, a desorption step with hot purge gas and a cooling step with cold inert gas. The desorption and cooling are affected by nitrogen circulated through a heater, an adsorber and a condenser. A nonequilibrium, nonisothermal mathematical model is developed to simulate temperature and concentration breakthrough curves for both adsorption and desorption steps. The computer simulation results are compared with the experimental data. A bench scale fixed bed adsorption unit was used for the experimental study. It is shown that the theoretical model predicts the experimental results well. The computer simulation results are used to study the effect of the purge gas and condensation temperature on the process efficiency.

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1. Introduction

Volatile organic compounds (VOCs) are the most common pollutants emitted from chemical, petrochemical, pharmaceutical and allied industries [1,2]. Furthermore, emission of these compounds originates from venting of the process vessels, storage tanks, and leaks from piping and equipment. Common examples of VOCs emitted from a variety of industrial sources are methanol, isopropyl alcohol, toluene, and xylene [3]. Volatile organic compounds are hazardous to the environment and human health. For this reason, their removal from flue gas streams became an important issue for researchers and engineers.

A number of treatment techniques are available for handling VOCs laden air streams, such as thermal oxidation, catalytic oxidation, adsorption, absorption, condensation, biofiltration, and membrane technology [4]. These techniques are classified into two major groups: destruction based and recovery based. Oxidation and biofiltration are considered as destructive techniques, while adsorption, absorption, condensation, and membrane technology are classified as recovery based ones [1].

The most currently applicable technology for volatile organic compounds control is adsorption. This method is commonly used to recover VOCs and to increase their concentration to a level at which it is more feasible to clean up the flue gas streams using a reasonable sized recovery plant [1]. Adsorption processes are also widely used for separation of gases. Recently, adsorption phenomenon is exploited to heat-pumping and refrigeration [5,6].

There are a number of commercially available adsorbents for controlling VOCs, including granular activated carbon (GAC), activated carbon fiber (ACF), zeolites, and polymers (styrene/divinylbenzene macroporous resins). Admittedly the proper adsorbent choice depends upon the application, but currently activated carbon is most often used as an adsorbent of VOCs [7]. This adsorbent possesses a high surface area, an intricate pore structure, and hydrophobic surface. Most commonly, the adsorption system consists of a fixed bed of adsorbent in a adsorption column. Some adsorption processes use moving or fluidized beds [3,7].

An adsorption process is conducted in a cyclic manner. The VOCs laden gas stream passes through a bed of adsorbent, whereas clean gas is exhausted to atmosphere. When the capacity of the bed is exhausted, the gas flow is stopped to terminate the adsorption step, then the bed is treated to remove the adsorbed molecules in a regeneration step, and the cycle is subsequently repeated [7]. The purification process can be made continuous by the use of two

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Nomenclature

a_u	useful capacity of the adsorbent bed, mol/kg	Re	Reynolds particle number
a_{uo}	useful capacity of the adsorbent bed in first adsorption cycle, mol/kg	R_p	adsorbent particle radius, m
C_{pc}	heat capacity of column, J/(mol K)	Sc	Schmidt number
C_{pg}	heat capacity of gas, J/(mol K)	Sh	Sherwood number
C_{ps}	heat capacity of adsorbent, J/(kg K)	t	time, s
D	internal diameter of column, m	T	temperature, K
D_{ax}	axial diffusion coefficient, m^2/s	T_c	column wall temperature, K
D_K	Knudsen diffusion coefficient, m^2/s	T_{cr}	critical temperature, K
D_M	molecular diffusion coefficient, m^2/s	T_{cond}	condensation temperature, K
D_{ps}	effective particle diffusion coefficient, m^2/s	T_g	gas temperature within the bed, K
D_S	surface diffusion coefficient, m^2/s	T_{in}	gas temperature at feed conditions, K
D_{so}	preexponential factor in (10), m^2/s	T_s	solid phase temperature, K
D_V	diffusion volume of adsorbate	T_{amb}	ambient temperature, K
D_{Vg}	diffusion volume of inert gas	U	overall heat transfer coefficient for column insulation, $W/(m^2 K)$
E	surface diffusion energy of activation, J/mol	V	volume adsorbed, m^3/kg
E_o	constant in (22), J/mol	V_o	constant in (22), m^3/kg
G	superficial molar gas flow rate, $mol/(m^2 s)$	V^{calc}	calculated value of V , m^3/kg
G_c	superficial molar carrier gas flow rate, $mol/(m^2 s)$	V^{exp}	experimental value of V , m^3/kg
h_f	heat transfer coefficient from the bulk gas phase to the particle, $W/(m^2 K)$	y	mole fraction of adsorbate in the gas phase (mole ratio), mol/mol
h_w	heat transfer coefficient from the bulk gas phase to the column wall, $W/(m^2 K)$	y^*	mole fraction of adsorbate in the gas phase in equilibrium with q , mol/mol
ΔH_a	heat of adsorption of adsorbate, J/mol	y_{cond}	saturation concentration of adsorbate at T_{cond} , mol/mol
k	overall mass transfer coefficient, 1/s	y_{in}	concentration of adsorbate at feed conditions, mol/mol
k_{ax}	axial thermal conductivity, $W/(m K)$	y_{out}	concentration of adsorbate at outlet from adsorption column, mol/mol
k_f	film mass transfer coefficient, m/s	z	axial distance, m
k_g	thermal conductivity of gas, $W/(m K)$		
L	bed length, m		
m_{ads}	mass of adsorbent, kg	Greek symbols	
m_{cond}	mass of condensed adsorbate, mol	α_{air}	ratio of the log mean surface area of the insulation to the volume of the column wall, 1/m
M	molecular weight of adsorbate, kg/kmol, kg/mol (24)	α_c	ratio of the internal surface area to the volume of the column wall, 1/m
M_g	molecular weight of inert gas, kg/kmol	α_p	particle external surface area to volume ratio, 1/m
n	constant in (22)	β	constant in (22)
N	number of experimental points	ε	bed voidage
Nu	Nusselt number	ε_p	particle porosity
P	partial pressure of adsorbate, Pa	ρ_c	column wall density, kg/m^3
p_s	saturation pressure, Pa, mmHg (25)	ρ_g	gas density, mol/m^3
P	total pressure, Pa	ρ	density of liquid adsorbate, kg/m^3
Pr	Prandtl number	ρ_p	particle density, kg/m^3
q	adsorbate concentration in solid phase, mol/kg	τ_p	pore tortuosity factor
q^*	equilibrium value of q , mol/kg		
r_e	mean pore radius, Å		
R	gas constant, J/(mol K)		

(or more) beds in parallel: one is off-line for desorption while the other is on adsorption.

Adsorption processes are identified by their method of regeneration. Two basic process cycles are most commonly used for controlling VOCs: temperature swing adsorption (TSA) and pressure swing adsorption (PSA) [1–3,7–9]. In the TSA cycle regeneration takes place at a temperature much higher than adsorption, whereas the PSA cycle uses changes in pressure to release adsorbed component. Sometimes hybrid regeneration cycle, known as temperature–pressure swing adsorption (TPSA), is used to increase the regeneration efficiency. In TPSA cycle, desorption is achieved both by increasing the bed temperature and decreasing the column pressure [8].

The recovery of volatile organic compounds from gas streams is most often accomplished by TSA cycle with activated carbon as an adsorbent [1–3,7,8,10]. In this cycle, activated carbon is commonly regenerated using superheated steam [7,10]. Regeneration process consists of three steps: desorption, drying and cooling. During desorption steam flows through the adsorber, transferring heat to

the activated carbon bed, and desorbs the VOCs. The steam–VOCs vapor mixture is then condensed and sent to appropriate separation devices to divide the water phase from the organic one. After desorption, the carbon becomes wet and hot, which is not suitable for the next adsorption step. Thus, the drying and cooling steps processes are required, in order to retrieve the adsorption ability of the activated carbon.

An alternative method of regeneration involves the use of hot nitrogen as purge gas, instead of steam [2,10]. In this method, regeneration consists of two steps: desorption and cooling. For strongly adsorbed species cooling step can be omitted [9]. In this case the bed will be cooled early during the adsorption step, because the thermal wave should pull ahead the solute wave [9]. Desorption is accomplished by passing a hot purge gas through the bed, most commonly in the direction countercurrent to the flow direction for the adsorption step [7,10]. The hot gas adds heat necessary for desorption to the bed, and sweeps the desorbed VOCs out of the adsorption column. The desorbed VOCs are then condensed. Because the purge gas is the primary heat source, high gas

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