



Column dynamics of an adsorption–drying–desorption process for butanol recovery from aqueous solutions with silicalite pellets

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ABSTRACT

The objective of this work is to study the column dynamics of an adsorption–drying–desorption (ADD) process for recovering butanol from diluted aqueous solutions with silicalite pellets. In the adsorption step, the butanol from the aqueous solution is adsorbed onto the column up to saturation. In the drying step, the column previously saturated with the butanol aqueous mixture is purged with air at low temperature (323–343 K). During this step the unbound liquid (liquid not adsorbed filling pipes and interstices with the same concentration as the feed mixture) is removed. In the desorption step, the column is heated at high temperature (403–423 K) and purged with air, for recovering the butanol adsorbed in the silicalite crystals by condensation. The experimental results show that the butanol can be recovered with 98% (w/w) purity from dilute aqueous solutions (0.5–2% w/w). A theoretical model has been proposed to describe the adsorption step. Another theoretical model has been developed to describe the concentration and temperature profiles of the drying and desorption steps. Both models have been validated with experimental data. The last model has been used to estimate the energy consumption of the drying and desorption steps of an ADD process considering compression, heating and refrigeration requirements. Simulations results show that it is possible to recover butanol from diluted aqueous mixtures using an ADD process with high recovery (95%), and with an energy requirement significantly lower (about 3.4 MJ kg^{-1}) than the energy content of butanol (36 MJ kg^{-1}).

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1. Introduction

The production technology for bio-fuels by fermentation such as bio-ethanol and bio-butanol is well-known from a long time ago [1], but it has attracted renewed interest lately because of the increase of oil prices and the depletion of fossil fuels. Also, the concern about the carbon dioxide emissions from fossil fuels has prompted this interest, as the net carbon balance for bio-fuels is theoretically neutral (ignoring the carbon generated in their production). Bio-butanol offers several advantages over bio-ethanol: it has higher energy density and less volatility, it is non-corrosive, less hygroscopic and it has better miscibility with gasoline [2]. However, the concentration of *n*-butanol typically reached in the fermenter is rather low (10–20 g/L) because this compound is toxic to the butanol-producing microorganisms [3]. Furthermore, acetone, ethanol, acetic and butyric acid are also produced in the fermentation broth. Due to the low concentration of butanol, the recovery cost of butanol by traditional distillation is high, as water is the majority component, with a boiling point below the one of butanol (373 K vs. 390 K). The expensive product recovery and

purification from the dilute fermentation broth still hinders the industrial production of bio-butanol [4]. Various separation techniques have been studied to replace the distillation method for butanol recovery, including steam stripping, gas stripping, pervaporation and adsorption. According to Qureshi et al. [5], the energy requirement for butanol separation by an adsorption–desorption process is lower than by any other technique. The usage of an adsorption method for recovering butanol from dilute butanol/water mixtures was first proposed by Milestone and Bibby [6]. An aqueous 0.5 w/v butanol mixture was passed through a column of silicalite crystals in powder form. Afterwards, the crystals were took out from the column and dried at 313 K to remove water, and then, heating at 423 K the butanol was desorbed and recovered by condensation. This method will be called adsorption–drying–desorption process (ADD) from now on. They reported that practically pure butanol was obtained by this method. The key features making this process possible are the very high selectivity of silicalite towards butanol in butanol/water mixtures, and the reversible character of the butanol adsorption. As it was noted by Qureshi et al. [5], this result shows that it is possible to remove most of the water from a diluted butanol/water mixture in one step with this method. The energy required to remove the major component (water) may come from a waste heat source, reducing substantially the energy requirement of the process. Despite this encouraging

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Nomenclature

C	total gas concentration, mol m ⁻³	r_w	bed radius, m
$C_{p,g}$	gas heat capacity at constant pressure, J mol ⁻¹ K ⁻¹	S_{bed}	bed cross-section, m ²
$C_{p,Li}$	heat capacity of liquid of <i>i</i> th component, J kg ⁻¹ K ⁻¹	Sc	Schmidt number, $\mu/(\rho \cdot D_m)$
$C_{p,s}$	solid heat capacity, J kg ⁻¹ K ⁻¹	T	temperature, K
$C_{v,g}$	gas heat capacity at constant volume, J mol ⁻¹ K ⁻¹	t	time, s
D_c	diffusivity of butanol in silicalite crystals, m ² s ⁻¹	t_∞	time required to reach the butanol inlet concentration at column outlet, s
D_L	axial dispersion coefficient, m ² s ⁻¹	t_D	delay time, s
D_{mg}	molecular diffusivity in a gas mixture, m ² s ⁻¹	$t_{des+cond}$	time period in where condensate was recovered, s
D_{mL}	molecular diffusivity in a liquid mixture, m ² s ⁻¹	$t_{desorption}$	duration of the desorption step, s
d_p	particle diameter, m	t_{drying}	duration of the drying step, s
D_p^*	effective macropore diffusivity, m ² s ⁻¹	\bar{t}	stoichiometric time, s
F_i	mole flow rate of <i>i</i> th component, mol s ⁻¹	T_g	temperature in the gas phase, K
h_{sg}	gas to solid heat transfer coefficient, W m ⁻² K ⁻¹	T_s	solid temperature, K
h_w	wall to gas heat transfer coefficient, W m ⁻² K ⁻¹	T_w	column wall temperature, K
k_f	external mass transfer coefficient, m s ⁻¹	u	superficial velocity, m s ⁻¹
k_g	gas thermal conductivity, W m ⁻¹ K ⁻¹	u_0	interstitial velocity, m s ⁻¹
$k_{s,i}$	mass transfer coefficient from gas to adsorbed phase, s ⁻¹	V_D	dead volume, m ³
$k_{s0,i}$	preexponential constant of $k_{s,i}$, s ⁻¹	V_p	specific pore volume of the adsorbent, m ³ kg ⁻¹
L	bed length, m	W	mass of adsorbent in column, kg
m	mass of unbound + bound liquid in the column, kg	x	butanol mass fraction in the bulk liquid
m_0	initial mass of unbound + bound liquid in the column after drainage, kg	x_m	mass fraction of butanol in macropores
$M_{w,i}$	molecular weight of <i>i</i> th component, kg mol ⁻¹	y_i	mole fraction of <i>i</i> th component in gas phase
n^*	adsorbed concentration of butanol from the vapour phase in equilibrium with p , mol kg ⁻¹	z	axial coordinate, m
$N_{bound,i}$	mass transfer rate of <i>i</i> th bound component, mol m ⁻³ -bed s ⁻¹	$\Delta H_{bound,i}$	adsorption enthalpy of bound <i>i</i> th component, J mol ⁻¹
$N_{unbound,i}$	mass transfer rate of <i>i</i> th unbound component, mol m ⁻³ -bed s ⁻¹	$\Delta H_{unbound,i}$	condensation enthalpy of unbound <i>i</i> th component, J mol ⁻¹
p	partial pressure, Pa	Greek symbols	
P	pressure, Pa	ε	bed porosity m ³ _{void} m ⁻³ _{bed}
P_{atm}	atmospheric pressure, Pa	ε_m	macropore volumetric fraction, m ³ _{macropore} m ⁻³ _{pellet}
P_r	Prandtl number, $C_{p,g} \mu_g / k_g$	η_c	compressor efficiency
$p_{v,i}$	vapour pressure of <i>i</i> th component, Pa	λ	thermal axial dispersion coefficient, W m ⁻¹ K ⁻¹
q	adsorbed concentration of butanol from the liquid phase in micropores, kg kg _{pellet} ⁻¹	μ_L	liquid viscosity, Pa s
\bar{q}	average adsorbed butanol concentration from the liquid phase in micropores, kg kg _{pellet} ⁻¹	μ_g	gas viscosity, Pa s
Q	volumetric flow rate, m ³ s ⁻¹	ρ_g	gas density, kg m ⁻³
$q^*(x)$	adsorbed butanol concentration in equilibrium with x , kg kg _{pellet} ⁻¹	ρ_L	liquid density, kg m ⁻³
$q_{bound,i}$	adsorbed concentration of bound <i>i</i> th component, mol kg ⁻¹	ρ_p	particle density, kg _{pellet} m ⁻³ _{pellet}
$q_{unbound,i}$	concentration of unbound <i>i</i> th component, mol kg ⁻¹	Δ	increment
R	gas constant, 8.31 J mol ⁻¹ K ⁻¹	Subscripts	
r	radial coordinate in the adsorbent particle, m	bound	liquid adsorbed; affected by the attracting field of the adsorbent
R_b	rate of butanol transfer between macropore and micropore regions, kg kg _{pellet} s ⁻¹	unbound	liquid nonadsorbed; not affected by the attracting field of the adsorbent
r_c	crystal radius, m	F	feed condition
Re	Reynolds number, $d_p u \rho / \mu$	g	gas
R_p	particle radius, m	ini	initial value
		fin	final value
		L	liquid
		s	solid

result, little information can be found in the literature about the dynamics of an ADD or similar processes, particularly for the drying–desorption step [1]. Lin et al. [7] have recently studied the adsorption of butanol from aqueous solution onto a new type of macroporous resin (KA-I resin). Li and Ma [8] studied the adsorption of butanol from aqueous mixtures on a column loaded with silicalite crystals. In order to avoid an excessive pressure drop, the usage of columns loaded with silicalite crystals (with sizes typically about 3 μm) is not advisable. Thus, agglomerated silicalite

particles must be employed for this process. Saravanan et al. [9] have recently studied the recovery of butanol with an ADD process with ZSM-5 extrudates with a high Si/Al ratio from aqueous mixtures containing acetone, butanol and ethanol. However, the estimated purity of recovered butanol is only about 84% (recovery of butanol by condensation is not studied). Furthermore, neither the modelling of the drying–desorption step nor the estimation of the energy consumption is done. The general objective of this work is to study the column dynamics of an improved ADD process for

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