



Hybrid membrane and conventional processes comparison for isoamyl acetate production



Wilmar Osorio-Viana^a, Harold Norbey Ibarra-Taquez^a, Izabela Dobrosz-Gómez^b, Miguel Ángel Gómez-García^{a,*}

^a Departamento de Ingeniería Química, Facultad de Ingeniería y Arquitectura, Universidad Nacional de Colombia, Sede Manizales, Campus Palogrande, Cra 27 64-60, Apartado Aéreo 127, Manizales, Caldas, Colombia

^b Departamento de Física y Química, Facultad de Ciencias Exactas y Naturales, Universidad Nacional de Colombia, Sede Manizales, Campus Palogrande, Cra 27 64-60, Apartado Aéreo 127, Manizales, Caldas, Colombia

ARTICLE INFO

Article history:

Received 3 April 2013

Received in revised form 4 December 2013

Accepted 12 December 2013

Available online 19 December 2013

Keywords:

Hybrid process

Energy integration

Economic evaluation

Isoamyl acetate

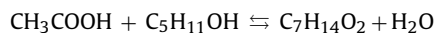
ABSTRACT

Four process alternatives for the production of isoamyl acetate, by the liquid phase esterification of acetic acid with isoamyl alcohol, were evaluated by simulation in terms of product purity, energy integration and economics. The analysis involves a transition from conventional (two structures that use acetic acid or alcohol in excess) to hybrid membrane process (two distillation–pervaporation hybrid systems). Acetate recovery is identified as a crucial factor to minimize energy costs in all considered processes. For conventional processes, the amount of energy required for separation, at low acetate recovery levels, is considerably lower if acetic acid is used in excess. For the hybrid processes, there is an optimum value of acetate recovery that minimizes the total required heat duty and membrane area. Hybrid distillation–pervaporation process allows obtaining the specified product purity with lower energy requirements and more economical tradeoffs than the considered conventional processes. The economic optimum design maximizes energy savings and minimizes total annualized costs. After optimization and energy integration, the best process alternative includes, in a hybrid system, one packed bed reactor, two pervaporation units and a distillation column.

© 2013 Elsevier B.V. All rights reserved.

1. Introduction

In some South American countries, biofuel ethanol is obtained from sugar cane fermentation [1]. This process generates considerable amounts of wastes, among them the so-called *fusel oil*: a complex mixture of higher alcohols and water which has received increasing attention for both its separation using membranes and its transformation to more valuable products [2–5]. Fusel oil consists of mainly isoamyl alcohol (>60 wt./wt.%) [6,7]. Liquid phase esterification of acetic acid (*HAc*) with isoamyl alcohol (*ROH*) could be a possible source of isoamyl acetate (*E*), a higher added value product, which is widely used in the pharmaceutical, food, green solvent and perfumery industry. This reaction, in which water (*W*) is obtained as a byproduct, is as follows:



Isoamyl acetate production presents serious difficulties including phase separation, limited chemical equilibrium and challenging

product purification. Conventionally, high conversion is achieved using reactant in excess and/or by selective recovery of one of the reaction products [8,9]. Process intensification by membrane technologies has been successfully used for esterification reactions [10]; specifically pervaporation is a very selective separation technique and is less energyvore than conventional or reactive distillation. In fact, pervaporation membrane reactors and pervaporation process have received increased attention as a potentially competitive process alternative for different esterification reactions [11,12].

Conventional processes for esters production consist of a reaction step followed by several separation and purification stages. Traditionally, esterification reactions are industrially performed under batch conditions (with a coupled column–reactor process) in homogeneous liquid phase, using mineral acids as homogeneous catalyst [13] or using heterogeneous catalyst like ion exchange resins [14,15]. In the separation/purification step, molecular sieves are often applied for water recovery in the esterification reactions. However, their drawbacks include water adsorption (only in limited amounts) and their reuse and scale-up [16,17].

Very few works related to isoamyl acetate process analysis and design are reported in the literature. For a similar ester (which could also be present in some *fusel oils*), Chiang et al. [18] compared, by simulation, two design alternatives for n-amyl acetate

* Corresponding author. Tel.: +57 6 8879300x50210; fax: +57 6 8879300x50129.
E-mail address: magomez@unal.edu.co (M.Á. Gómez-García).

Nomenclature

<i>a</i>	NRTL parameter
<i>b</i>	NRTL parameter
<i>c</i>	NRTL parameter
<i>x</i>	molar fraction
K_{eq}	chemical equilibrium constant
<i>r</i>	reaction rate
<i>k</i>	rate constant
<i>T</i>	temperature
<i>C</i>	concentration
<i>N</i>	membrane flux
<i>a</i>	liquid phase activity
<i>D</i>	diffusion coefficient
D_0	pre-exponential factor for diffusion coefficient
<i>z</i>	membrane thickness
<i>B</i>	exponential interaction parameter
<i>X</i>	fractional conversion
<i>R</i>	product recovery fraction
<i>F</i>	molar flow
<i>i</i>	fractional interest rate
<i>n</i>	project number of years

Subscripts

<i>i</i>	component
<i>j</i>	component
<i>m</i>	membrane
<i>p</i>	product

Abbreviations

<i>HAc</i>	acetic acid
<i>ROH</i>	isoamyl alcohol
<i>E</i>	isoamyl acetate
<i>W</i>	water
<i>NRTL</i>	non random two liquid
<i>RCM</i>	residue curve map
<i>TAC</i>	total annualized cost
<i>ACC</i>	annualized capital cost
<i>HETP</i>	height equivalent to a theoretical plate
<i>hom</i>	homogeneous
<i>het</i>	heterogeneous
<i>gas</i>	gaseous
<i>liq</i>	liquid

production: a reactor coupled to a distillation column and reactive distillation. In both cases, separation train of the conventional process is avoided. They concluded that reactive distillation process is at least four times cheaper than coupled column-reactor process. Saha et al. [19,20] developed an experimental set-up for the iso-amyl acetate production by reactive distillation, defining operational conditions. As far as we know, there are not comparative studies reported in the literature for different process schemes for isoamyl acetate production, including conventional and membrane processes.

In this work, four process schemes are studied by simulation using ASPEN Plus® together with a homemade MS Excel®–MatLab® interface for membrane modules simulation (membrane modules are not included in the equipment model library incorporated in ASPEN Plus®). Alternatives include: two conventional processes using reagent in excess (with reaction followed by a distillation train) and two hybrid pervaporation-distillation processes with different number of units. Conceptual design of each process alternative is accomplished using residue curve maps analysis [21]. A systematic design procedure was planned to optimize distillation

columns performance, based on Turton et al. [22], and energy integration (through Pinch point methodology). Total annualized cost (TAC) and main design variables are used for comparison among process alternatives.

2. Mathematical models for simulation

Several models are required to perform the process simulation under study: (i) a thermodynamic model, for stream properties and general simulation procedures; (ii) a kinetic one, for the reactor simulation; and (iii) a membrane mass transfer model, for the pervaporation unit evaluation. The following subsections present their detailed description.

2.1. Thermodynamic model

To account for nonideal phase behavior for the quaternary system, the Non-Random-Two-Liquid (NRTL) model [23] was used to compute activity coefficients. Table 1 lists the NRTL model parameters, as proposed by Osorio et al. [24]. Vapor phase nonideality is considered as a consequence of acetic acid dimerization and described by the Hayden-O'Connell model [25]. The ASPEN Plus® built-in association parameters were used to calculate fugacity coefficients.

2.2. Kinetic model for Amberlite IR-120 catalyst

The reaction rate at which the catalyzed reaction proceeds is adequately described in the terms of mole fractions and is given by Eq. (1) [26]:

$$r_{HAc} = \frac{-k_{1,het}(x_{HAc}x_{ROH} - (x_E x_W / K_{eq}))}{(1 + 0.0133x_{HAc} + 0.0444x_{ROH} + 0.028x_W)^2} \quad (1)$$

where the equilibrium constant value is equal to 5.0, and it can be treated as independent of temperature, and the kinetic constant can be calculated by the following equation:

$$\ln k_{1,het} \left\langle \frac{\text{mol}}{\text{h} \times \text{g}} \right\rangle = 0.034 - \frac{52.2}{0.008314} \times \left(\frac{1}{T} - \frac{1}{363.15} \right) \quad (2)$$

Additionally, it is necessary to consider the intrinsic homogeneous reaction rate. It takes into account the autocatalytic effect of acetic acid [27]. This is given by Eq. (3):

$$r_{HAc,hom} = -k_{1,hom} x_{HAc}^{1.21} \left(x_{HAc} x_{ROH} - \frac{x_E x_W}{K_{eq}} \right) \quad (3)$$

where the rate constant is evaluated by Eq. (4):

$$\ln k_{1,hom} \left\langle \frac{\text{mol}}{\text{h} \times \text{L}} \right\rangle = 1.417 - \frac{62.336}{0.008314} \times \left(\frac{1}{T} - \frac{1}{363.15} \right) \quad (4)$$

2.3. Membrane permeation model

A hydrophilic silica membrane is considered in this study. The flux through the membrane is expressed by Eq. (5), based on a Maxwell–Stefan approach [28]:

$$N_i = -c_i \frac{D_{im}}{z} \ln \left(\frac{a_{i,gas}}{a_{i,liq}} \right) \quad (5)$$

where, for each substance, c_i is the concentration in kg/m^3 and the parameters D_{im}/z are calculated according to Eq. (6) and the corresponding parameters presented in Table 2.

$$\frac{D_{im}}{z} = D'_{ij0} \exp \left(\sum B_i a_i \right) \quad (6)$$

Download English Version:

<https://daneshyari.com/en/article/687026>

Download Persian Version:

<https://daneshyari.com/article/687026>

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