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Prospects & potential of biobutanol production integrated with organophilic pervaporation – A techno-economic assessment

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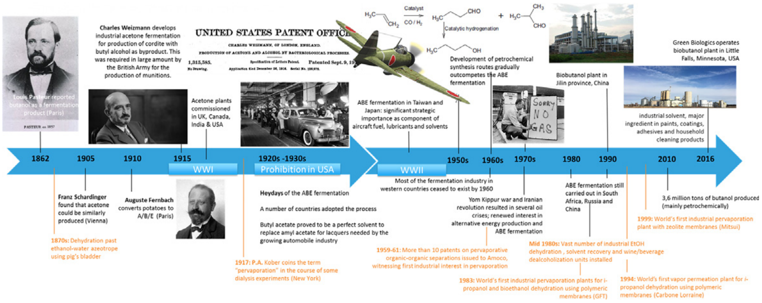
HIGHLIGHTS

- Engineering guidelines for pervaporation units are provided for ABE production.
- The effects of permeate pressure and cooling were taken into account.
- a pervaporation module purchase price of 50–100 € m^{-2} should be targeted.

GRAPHICAL ABSTRACT



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ABSTRACT

The technical feasibility of integrating ABE fermentations with organophilic pervaporation has been described and demonstrated numerous times. However, engineering guidelines for integration of pervaporation with fermentation are currently not available. A novel calculation procedure to size pervaporation units in function of carbohydrate concentration in the feed is elaborated in detail. The overall energetic and economic outlook are less investigated and remain unclear. Furthermore, the effect of permeate pressure and cooling are frequently ignored. Therefore, the advantages and economic outlook of such an integration are estimated and calculated for fermentative *n*-butanol production at a capacity of 100 ktonnes per year. Biobutanol production costs for two cases were calculated. The base-case consists of a multi-stage acetone-butanol-ethanol fermentation with default downstream processing. The alternative is a continuous hybrid process where default downstream processing is complemented with organophilic pervaporation for recovery of solvents during the fermentation. Bare pervaporation module costs were estimated to ensure improved economics in comparison to the base-case. Equal installed costs for both cases are reached at a pervaporation module purchase price of 176 € m^{-2} for a composite POMS membrane. To derisk this potential large scale organophilic pervaporation application, a module purchase price of 50–100 € m^{-2} should be targeted.

1. Introduction

N-butanol is a bulk chemical amongst others used as an industrial solvent and as an ingredient in paint, coatings and adhesives. Currently, its main production route is the petrochemical oxo synthesis, starting from propene. For a number of reasons, the older established acetone-butanol-ethanol (ABE) fermentation enjoyed and still enjoys academic, industrial, and military interest in waves throughout the last decades [1,2]. Typically, *C. acetobutylicum*, *C. beijerinckii*, *C. saccharobutylicum* and *C. saccharoperbutylacetonicum* are used in industrial ABE

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Abbreviations

A	pervaporation surface [m^2]
ABE	Acetone-Butanol-Ethanol
B	butanol
BkW	brake power [kW]
C	concentration [$g\ L^{-1}$]
$C_{B,3}$	butanol concentration in the third fermentor [$g\ L^{-1}$]
$C_{B,permeate}$	butanol concentration in the permeate [$g\ L^{-1}$]
$C_{B,total}$	average butanol concentration from effluent from third fermentor and permeate flow [$g\ L^{-1}$]
$C_{carb,F}$	carbohydrate concentration in the feed [$g\ L^{-1}$]
$C_{carb,3}$	carbohydrate concentration in the third fermentor [$g\ L^{-1}$]
$C_{i,fermentor}$	concentration of compound i in the fermentor [$g\ L^{-1}$]
C_P	f.o.b. purchase cost
$CEPCI$	chemical engineering plant cost index
CFM	cubic feet per minute
COP	coefficient of performance
D	dilution rate [h^{-1}]
D_{ov}	overall dilution rate [h^{-1}]
$DCFROR$	discounted cash flow rate of return
E	ethanol
$EROI$	energy return on investment
F	feed flow [$m^3\ h^{-1}$]
$f.o.b.$	free on board
FS	flow at suction [$m^3\ h^{-1}$]
G	flow rate of component i [$kg\ h^{-1}$]
HHV	higher heating value [$MJ\ L^{-1}$ or $MJ\ kg^{-1}$]
HP	horsepower
HTA	heat transfer area [m^2]
IF	installation Factor
IRR	Internal Rate of Return
$ISPR$	*in situ* product recovery
J	flux [$g\ m^{-2}\ h^{-1}$]
J_{ABE}	ABE flux [$g\ m^{-2}\ h^{-1}$]
J_B	butanol flux [$g\ m^{-2}\ h^{-1}$]
J_{water}	water flux [$g\ m^{-2}\ h^{-1}$]
J_i	flux for component i [$g\ m^{-2}\ h^{-1}$]
MEE	Multiple Effect Evaporator
MM	Million (mille mille)
$M.W.$	molecular weight
NPV	Net Present Value
NRG	energy
$NRTL$	non-random two-liquid model thermodynamic model
P	productivity [$g\ L^{-1}\ h^{-1}$]
$P_{B,ov}$	overall butanol productivity [$kg\ m^{-3}\ h^{-1}$]
$PDMS$	polydimethylsiloxane
$PEBA$	polyether block amide
$POMS$	poly(octyl methyl siloxane)
$PTMSP$	poly[1-(trimethylsilyl)-1-propyne]
R	ratio of effluent flow to feed flow
ROI	return on investment
S_{ov}	overall carbohydrate consumption [$g\ L^{-1}\ h^{-1}$]
$SCDS$	Simultaneous Correction Distillation System
$S.F.$	size factor
SE	steam economy
SF	split factor; ratio of solvents recovered by pervaporation to total solvents produced
T	temperature (°C)
V	net reactor volume [m^3]
$Y_{B/Carb}$	butanol yield [$g_{butanol}\ g_{carb}{}^{-1}$]

fermentations [3]. While product ratios can be different from strain to strain, the solvent concentration at the end of a fermentation barely exceeds ~2 wt%, leading to high distillation costs, high waste water volumes coming from the effluent of the beer stripper and low productivities due to product inhibition [4,5]. To illustrate this, Mariano et al. [6] calculated the production of 43–86.7 L stillage per L butanol and the consumption of 29.8–47.5 MJ per kg butanol (excluding stillage treatment) in a conventional plant consisting of mainly fermentation and distillation. The quest for increased solvent productivities, decreased process flows and especially decreased energy consumption explain the interest in *in situ* product recovery (ISPR) technologies [7]. A plethora of research results are available on a multitude of ISPR technologies for fermentative *n*-butanol recovery [8].

To the best of our knowledge, introduction of ISPR technologies for fermentative *n*-butanol production on an industrial scale has not materialized yet [other than the Green Biologics plant in Minnesota, USA (personal communication)] due to the financial risks associated with introducing novel technology on large scale, the required in-depth knowledge of fermentation technology and (bio)chemical engineering to size, design and operate an integrated production plant, and last but not least, the petrochemical production route towards *n*-butanol as a competing process [9].

The term pervaporation was first coined in 1917 by Kober [10]. Independently from evolutions in ABE production, pervaporation evolved from a mere scientific observation to full scale industrial processes. In contrast to the more mature and developed hydrophilic pervaporation membranes, organophilic pervaporation selectively removes organics from dilute aqueous streams. The potential of several organophilic pervaporation membranes coupled to solventogenic fermentations has already been thoroughly investigated [11–19]. The flux of a pervaporation module is a determining factor for the capital investment while the separation factor of the used membrane has implications towards energy consumption and hence operational costs in the production plant. Initially, dense polydimethylsiloxane (PDMS) membranes were used by early pioneers resulting in low (solvent) fluxes. To illustrate this, a total flux of 10.40 $g\ m^{-2}\ h^{-1}$ was reported by Groot et al. in 1987 [20] and 25.1–34.8 $g\ m^{-2}\ h^{-1}$ by Qureshi et al. in 1999 [21]. In the last decade, thin film composite membranes consisting of a selective top layer on a porous support resulted in significantly higher (solvent) fluxes. PDMS can be considered as a reference material for the top layer, but other silicone based polymers (PTMSP [22] and POMS [23]), polyether block amide (PEBA) polymers [24] and liquid membranes [25] were investigated as well. PDMS based membranes are of particular interest as they do not suffer from fouling during more than 27 days of operation, a prerequisite for long-term operations [13,26]. More recently, POMS based membranes proved their superior performance in terms of (solvent) flux and separation factor in comparison to PDMS based membranes during long-term continuous integrated cultivations lasting more than 25 days [27].

A decreased energy consumption is often claimed as an advantage for introducing ISPR, but rarely calculated for the overall process taking into account all process streams [8]. Rigorously calculating the energy consumption for production of bioproducts, particularly of potential biofuels, is an arduous effort, but a necessity to advance emerging technologies that can impact society. Significant savings in steam consumption of 51.7% from fermentation till purified products (from 69.5 $MJ\ kg^{-1}$ to 33.52 $MJ\ kg^{-1}$ butanol using ChemCAD simulations, excluding pretreatment) in the distillation and evaporator sections of the plant were calculated for a multistage continuous fermentation integrated with organophilic pervaporation (as described in WO2018/015415 A1) in comparison with a conventional continuous fermentation [11,28].

Often neglected aspects in pervaporation are costs associated with maintaining the vacuum and costs associated with cooling the permeate