

# Simulation and energy analysis of the ABE fermentation integrated with gas stripping

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## **Abstract**

Acetone-Butanol-Ethanol (ABE) fermentation has recently gained renewed attention in the context of biorefinery approaches for the production of fuels and chemicals from renewable resources.

The main problem associated with the ABE fermentation by microorganisms is the self-inhibition of the process due to butanol toxicity to the culture, which limits the application on an industrial scale. The use of a separation method, which enables selective removal of toxic components from the fermentation broth, may contribute to improve the economics of such process. In this context, different recovery techniques have been investigated, including gas stripping. Several literature experimental studies on lab-scale integrated gas stripping units are available, but few deal with process simulation.

This work concerns the recovery of butanol from a batch fermenter, in which the ABE mixture is recovered from the broth by means of gas stripping and, subsequently, fractionated in a distillation train. The aim of this work is to compare the performances of the integrated fermentation-gas stripping process with those of a conventional batch fermenter. For this purpose, both processes have been modeled and their energy requirements have been determined by means of an energy analysis. Results show that product removal has a beneficial effect on sugar consumption and process productivity at the expense of an increased energy demand.

**Keywords:** bio-butanol, ABE fermentation, integrated product recovery, gas stripping, inhibitors removal, process intensification.

## **1. Introduction**

The increasing greenhouse gas emissions and the resulting global climate change, joined with a growing world population, are key challenges that are leading to an increased interest in processes able to produce fuels and chemicals from alternative, renewable resources.

Butanol is an important commodity chemical and an attractive biofuel, which shows several

39 advantages over ethanol [1]. It can be produced through the Acetone-Butanol-Ethanol (ABE)  
40 fermentation by clostridia. However, the fermentation is limited by product inhibition, especially due  
41 to butanol, and only 22 g/L of total solvents with a butanol concentration lower than 13 g/L are  
42 typically obtained in the bioreactor during a batch process [2].

43 Therefore, the ABE fermentation is characterized by high process costs, due to the large volumes  
44 required and the downstream product recovery from dilute broths, usually performed by distillation.

45 The application of integrated recovery techniques that remove butanol from the broth during  
46 fermentation represents a viable solution to solve the problems related to the ABE fermentation and  
47 to increase the profitability of the process [3]. In fact, product recovery reduces the effects of product  
48 inhibition and allows increasing the process productivity. The operating costs for product recovery  
49 are expected to decrease for higher concentrations of ABE in the mixture to be treated.

50 In an integrated process, products can be separated from the fermentation broth either within the  
51 reactor or by circulating the broth through an external separation unit. These two techniques are  
52 referred to as *in-situ* and *ex-situ*, respectively [4].

53 The most mature product removal techniques include gas stripping, solvent extraction, adsorption,  
54 vacuum fermentation, pervaporation and perstraction [5].

55 Gas stripping shows several advantages over other techniques. It is simple to operate and scale-up,  
56 it does not require expensive equipment, it does not remove nutrients and intermediates from the  
57 broth and it is not harmful to the fermentation microorganism [6]. Several literature works reported  
58 the effect of butanol titer in the broth on the stripping process. In this respect, Xue et al. [7] reported  
59 that it is necessary to conduct gas stripping at a butanol concentration higher than 8 g/L in the feed  
60 solution in order to obtain a condensate with a butanol titer higher than its solubility in water, which  
61 results in phase separation.

62 The effects of application of *in-situ* gas stripping on the ABE fermentation have been widely  
63 studied, demonstrating the reduction of product toxicity and the resulting improvements in  
64 productivity [8].

65 However, only a few papers appeared dealing with the energy demand associated with this  
66 technique. Oudshoorn et al. [9] reported an energy demand in the range of 14-31 MJ/kg of produced  
67 butanol considering a steady-state process for the separation of a water-butanol binary mixture. Groot  
68 et al. [10] considered a two-column system, neglecting the complete separation of acetone and  
69 ethanol, obtaining an energy requirement of 21 MJ/kg<sub>ABE</sub>. Outram et al. [11] compared different *in-*  
70 *situ* product recovery techniques for the separation of acetone, butanol and ethanol from a batch  
71 reactor. However, they adopted a simplified representation of the fermenter that was modeled as a  
72 stoichiometric, continuous, steady-state reactor. The continuous stream from the fermenter was

73 assumed to represent the desired conditions in the reactor and the fermentation broth was not recycled  
74 to the fermenter after the *in-situ* recovery had been applied.

75 In this work, the effect of the integrated recovery by gas stripping on the whole ABE production  
76 process has been studied.

77 Dynamic simulations of the batch fermentation have been performed in order to investigate the  
78 beneficial effect of product removal on the process productivity and to compute the actual  
79 concentration levels of the stream sent to the stripping column. Process simulations have allowed to  
80 assess the effectiveness of the integrated product recovery technique and the overall energy demand.  
81 The integrated process has been finally compared with a traditional batch fermentation, both followed  
82 by downstream fractionation of the solvents, in order to investigate the potential advantages of the  
83 process intensification.

84

## 85 **2. Thermodynamic Framework**

86 Thermodynamics plays an important role in process simulation, allowing to properly describe the  
87 phase equilibrium conditions involved in the process under study. The ABE mixture contains many  
88 polar compounds and, thus, shows a strong non-ideal behavior, with components pairs also forming  
89 azeotropic systems. In addition to this, butanol and water exhibit a miscibility gap, whose correct  
90 representation is fundamental for the proper description of the separation process. Therefore, the  
91 thermodynamic model must be able to properly describe both Vapor-Liquid Equilibrium (VLE) and  
92 Liquid-Liquid Equilibrium (LLE) conditions.

93 Considering the non-ideal behavior of the system under study, an indirect  $\gamma/\phi$  method (NRTL-RK)  
94 has been chosen in this work for its thermodynamic characterization. It is based on the Non-Random  
95 Two-Liquid (NRTL) model [12] and on the Redlich-Kwong (RK) Equation of State (EoS) [13] for  
96 the calculation, respectively, of the activity coefficient in the liquid phase and of the fugacity  
97 coefficient in the vapor phase. The considered components have been only those effectively removed  
98 from the broth, namely acetone, butanol, ethanol, water, and the stripping agent, nitrogen. Firstly, the  
99 capability of the model to reproduce phase equilibria using default parameters implemented in the  
100 Aspen Plus<sup>®</sup> process simulator has been checked. Then, the model parameters have been regressed  
101 to improve model predictions, making use of many binary and ternary VLE and LLE data available  
102 in the literature [14]. Nitrogen has been treated as a Henry component and its Henry's law constants  
103 have been kept equal to default values. The regressed parameters (reported in Table 1, where acetone,  
104 butanol, ethanol and water are denoted by A, B, E and W, respectively) have been implemented in  
105 Aspen Plus<sup>®</sup> [15], chosen for the simulation of the distillation process aimed at recovering acetone,  
106 ethanol and butanol downstream of the fermentation process. Those concerning the ethanol-butanol

107 and the ethanol-water pairs are not reported in Table 1 because the parameters available in the  
 108 simulator have been used for them, being already suitable for satisfactorily reproducing the  
 109 experimental data.

110  
 111 **Table 1.** Regressed parameters for the NRTL-RK thermodynamic model.

<b>i</b>	<b>A</b>	<b>A</b>	<b>A</b>	<b>B</b>
<b>j</b>	<b>W</b>	<b>B</b>	<b>E</b>	<b>W</b>
<b>a<sub>ij</sub></b>	-1.6803	3.9225	-2.2512	-2.0447
<b>a<sub>ji</sub></b>	4.9929	-3.2661	2.3301	4.2834
<b>b<sub>ij</sub></b>	860.74	-1273.33	879.45	980.98
<b>b<sub>ji</sub></b>	-1078.63	1196.83	-666.71	-239.15
<b>α</b>	0.5149	0.3000	0.8006	0.4454

112  
 113 Fig. 1 represents the temperature-composition diagram for the system water-butanol at 1 atm. The  
 114 comparison with the experimental data [16-23] shows a better agreement with the predictions by the  
 115 proposed model rather than with those by the default model, especially in the LLE region.

116  
 117 [FIGURE 1 HERE]

118  
 119 The model has been also applied to the ternary systems acetone-water-butanol and ethanol-water-  
 120 butanol and validated by comparison with the experimental data available in the literature. Results  
 121 are illustrated in the ternary diagrams shown in Fig. 2 for the system with acetone and in Fig. 3 for  
 122 the system with ethanol.

123  
 124 [FIGURE 2 HERE]

125  
 126 [FIGURE 3 HERE]

### 127 128 129 **3. Process Description**

130 In this work, the classical batch fermentation process has been compared with an alternative process  
 131 solution, which involves a gas stripping operation aimed at removing the fermentation products from  
 132 the broth in order to reduce inhibition. As reported in the literature, either nitrogen or the fermentation  
 133 gases (i.e., CO<sub>2</sub> and H<sub>2</sub>) can be used for this purpose. However, the use of CO<sub>2</sub> can alter the pH of the  
 134 fermentation broth, which is a parameter of paramount importance for the stability of the  
 135 fermentation. Therefore, nitrogen has been considered as stripping agent and an *ex-situ* removal of  
 136 the fermentation products has been preferred to an *in-situ* one, in order to reach higher performances  
 137 in a multi-stage rather than in a single-stage unit and without disturbing the fermentation.

138 Fig. 4 shows the scheme of the integrated process. The fermentation broth is withdrawn from the  
139 fermenter and fed to the stripping column at a time when the microorganism starts to be inhibited by  
140 the solvents. The fermentation broth depleted from the components stripped by nitrogen is then  
141 recycled back to the fermenter. As for the vapor stream leaving the stripping column, it is sent to a  
142 condenser, where it is cooled down to 3°C so that the stripped compounds are condensed and  
143 separated from nitrogen, which is recycled back to the stripping column after being heated-up to the  
144 fermentation temperature. A nitrogen make-up stream has been also considered to replace the losses  
145 occurring in the condensed stream. The composition of this stream is such that a phase separation  
146 occurs between a butanol-rich liquid phase (named ‘Organic phase’ in Fig. 4) and a water-rich liquid  
147 phase (named ‘Aqueous phase’ in Fig. 4).

148  
149 [FIGURE 4 HERE]

150  
151 The two liquid phases leaving the integrated fermenter-stripping process are collected in storage  
152 tanks and then sent to the distillation train for products fractionation, which is operated in continuous  
153 mode. The selected process scheme is the one described by Mariano et al. [26] and illustrated in Fig.  
154 5.

155 The aqueous phase collected in the storage tank is mixed with the fermentation broth remained in  
156 the reactor. The resulting stream (named ‘FEED’ in Fig. 5) is fed to the beer column, after being  
157 pumped to 1.5 atm (i.e., the operating pressure of the beer column) and pre-heated to 93.4 °C by the  
158 bottom product obtained from this column, which separates the solvents (ABE) from the beer. The  
159 solvents, which are recovered at the top of the column, are cooled down providing heat to the reboiler  
160 of the downstream acetone column and ethanol column (described in the following) and further  
161 cooled down to 70°C before being expanded to 0.7 atm and mixed with the organic phase (named  
162 ‘ORGANIC’ in Fig. 5) leaving the integrated fermenter-stripping process. This stream is heated up  
163 from 3°C to 70°C and expanded to 0.7 atm before mixing with the ABE stream. The resulting mixture  
164 is treated in the four-column system also described by Mariano and coworkers [26]. In this system,  
165 the distillation columns are operated at different pressures in order to allow heat integration.

166 Acetone is separated in the acetone column, operated at 0.7 atm. As previously stated, the reboiler  
167 duty is met by condensing part of the overhead vapors from the beer column. The top product that  
168 meets commercial specifications is later compressed to atmospheric pressure and condensed.

169 The bottom product from the acetone column is cooled down to 50°C, expanded to 0.3 atm and  
170 further treated in the downstream ethanol column, which is operated at this pressure (vacuum  
171 operation allows the total reboiler duty to be met by condensing a part of the overhead vapors from  
172 the beer column).

173 A two-column distillation system in conjunction with a decanter is used to separate the  
 174 heterogeneous binary butanol/water azeotrope. Thus, after being pumped to atmospheric pressure  
 175 (i.e., the operating pressure of the water and butanol columns and of the decanter), the bottom stream  
 176 of the ethanol column, is added to a decanter together with the top streams from the water and butanol  
 177 columns. In the decanter, the butanol-rich liquid phase separates from the aqueous phase, forming an  
 178 upper layer. The water-rich liquid phase is refluxed to the water column, whose bottom product  
 179 contains almost pure water. On the other side, the butanol-rich liquid phase leaving the decanter is  
 180 refluxed to the butanol column, which produces a high purity butanol product. The overhead vapors  
 181 from the water and butanol columns are cooled down to 80°C, before they are sent back to the  
 182 decanter.

183 The same scheme has been also used for the recovery of the solvents from the fermenter when it is  
 184 not integrated with the gas stripping unit. The only difference lies in the composition of the stream  
 185 fed to the beer column (which is the one of the broth remained in the fermenter when the reaction is  
 186 stopped) and in the absence of the organic stream, which is mixed with the overhead vapors of the  
 187 beer column and sent to the acetone column in the scheme shown in Fig. 5.

188 [FIGURE 5 HERE]  
 189

## 190 4. Modeling

192 The process studied in this work consists of two parts: the integrated fermenter, operated in batch  
 193 mode, and the distillation train for product fractionation, operated continuously.

194 Since the integrated recovery process is a non-conventional unit, it has been modeled by means of  
 195 an in-house Fortran routine. On the contrary, the downstream separation process consists of  
 196 conventional unit operations and, thus, it has been simulated using a commercially available  
 197 simulation software (Aspen Plus® [15]).

198 The kinetic model used for simulating the reactor is the one proposed by Mulchandani and Volesky  
 199 [27], which also accounts for the inhibition due to butanol and butyric acid (subscript 'ba') through  
 200 the following function  $f$ :

$$201 \quad f = \exp(-0.01 \cdot (C_B + C_{ba})) \quad (C_B + C_{ba}) \leq 8 \text{ g/L} \quad (1)$$

$$f = 2.16 - 0.153 \cdot (C_B + C_{ba}) \quad 8 \leq (C_B + C_{ba}) \leq 13.9 \text{ g/L} \quad (2)$$

202 The reactor model equations are of the form given by Eq. (3), in which  $r_i$  and  $C_i$  denote,  
 203 respectively, the rate of reaction and the concentration of the  $i$ -th component inside the reactor (i.e.,  
 204 the microorganism, substrate, butyric acid, acetic acid, butanol, acetone, ethanol and water). To model  
 205 the integrated scheme shown in Fig. 4, the material balance equations have been modified with respect  
 206

207 to the ones for a classical batch fermenter to account for the integration between the fermenter and  
208 the stripping unit by using two parameters: the removal function,  $\eta_i$ , and the dilution factor,  $D_f$ .

209

$$\frac{dC_i}{dt} = r_i - \eta_i \cdot D_f \cdot C_i \quad (3)$$

210

211 The dilution factor corresponds to the ratio between the volumetric flow rate withdrawn from the  
212 reactor and the reactor volume. If the dilution factor is zero, Eq. (3) reduces to the one that  
213 characterizes the behavior of a classic batch reactor.

214 Only the volatile components (namely, acetone, butanol, ethanol and water) have been considered  
215 to be removed by gas stripping (as reported in the literature by Ezeji et al. [6]), assuming the removal  
216 to be instantaneous and the reaction volume to remain constant. For each of these components, a  
217 removal function defined as the ratio between its molar flow rate in the condensed phase and that in  
218 the broth entering the stripping column has been considered. The removal functions have been  
219 obtained by regression of the results obtained from simulations performed in Aspen Hysys® [28] for  
220 different broth compositions in terms of solvents concentrations, which have been varied in the range  
221 of values typically observed in the fermenter. In these simulations, an atmospheric stripping column  
222 with 10 theoretical stages has been used and the liquid and gas phases enter it at 32°C. Moreover, the  
223 liquid-to-gas ratio has been set to 2 (on a molar basis): this value allows to maximize the recovery of  
224 solvents from the fermentation broth [29].

225 Eq. (3) has been written for each considered component and the resulting ordinary differential  
226 equation system has been solved in Fortran using the DLSODES solver from ODEPACK [30].

227 At the end of the fermentation, the mass of each component,  $m_{F,i}$ , remaining in the fermentation  
228 broth inside the reactor is computed as:

229

$$m_{F,i} = C_i(t_F) \cdot V_F \quad (4)$$

230

231 in which  $C_i(t_F)$  represents the concentration of component  $i$  in the fermentation broth at the end of the  
232 fermentation and  $V_F$  is the fermenter volume.

233 In the integrated process, the mass flowrates,  $\dot{m}_{S,i}$ , of the components removed from the reactor by  
234 the stripping column and collected in the condensate are computed as:

235

$$\dot{m}_{S,i} = \eta_i \cdot D_f \cdot C_i \cdot V_F \quad (5)$$

236

237 The solvents stripped during the process are condensed and the obtained organic and aqueous  
238 phases are, then, collected in storage tanks. The final mass of each component in the storage tanks is  
239 computed integrating its mass flowrate over the fermentation time:

240

$$m_{S,i} = \int_0^{t_F} \dot{m}_{S,i} dt \quad (6)$$

241

242 As previously stated, the downstream ABE separation process has been simulated in the Aspen  
243 Plus<sup>®</sup> environment.

244 For the beer column, which consists of 50 theoretical plates and acts as a reboiled stripper, a mass  
245 recovery of 99.9% for the three components acetone, butanol and ethanol in the overhead vapor  
246 stream has been specified.

247 For the acetone column, the acetone mass purity (99.5 wt%) and the recovery (99.5%) have been  
248 specified, in addition to the number of stages (equal to 30).

249 The ethanol column has 50 stages, with the feed stream entering it at stage 14. In addition to the  
250 number of stages, the ethanol mass purity (85 wt%) and the recovery (95%) have been specified for  
251 the overhead product.

252 Finally, the water and butanol columns for the separation of the heterogeneous azeotrope consist of  
253 10 stages, with feed at stage 1. Purities of 99.8 wt% for both the bottom products, namely water and  
254 butanol, have been specified.

255

## 256 **5. Energy Analysis**

257 To compare the performances of the two processes, the equivalent fuel approach has been applied.  
258 It has been already used in previous works [31, 32] in terms of equivalent methane in order to  
259 determine the amount of methane equivalent to a certain duty or work that must be supplied to or can  
260 be recovered from each equipment involved in the process. Similarly, in this work the amount of  
261 equivalent butanol has been determined as explained in the following, making use of its lower heating  
262 value (LHV) and of the cycle efficiencies (Table 2 summarizes the values used for each parameter in  
263 this work).

264



**Table 2.** Parameters used for the calculation of the equivalent amount of butanol consumed by the process.

Coefficient	Value
LHV [MJ/kg]	33.11
$\eta_{CC}$	0.55
$\eta_{boiler}$	0.8
$\eta_{II}$	0.6

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A certain amount of butanol must be consumed for producing low-pressure (LP) steam that provides heat to the reboilers of the beer column, of the water column and of the butanol column and to the organic fraction coming from the stripping unit. To account for this, LP steam has been considered to be generated by a butanol-fired boiler. The amount of equivalent butanol can be computed according to Eq. (7), given the boiler efficiency ( $\eta_{boiler}$ ), the fuel lower heating value and the thermal duty to be provided to the process ( $\dot{Q}$ ).

$$\dot{m}_{FUEL} = \frac{\dot{Q}}{\eta_{boiler} \cdot LHV} \quad (7)$$

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Condensation of the solvents contained in the vapor stream leaving the stripping column in the integrated process configuration shown in Fig. 4 (by cooling it down to 3°C) has been assumed to be achieved by a proper refrigeration cycle requiring electric power ( $\dot{W}_{el}$ ) considered to be provided by a combined cycle. To calculate the amount of butanol equivalent to the required electric power, the coefficient of performance of the refrigeration cycle ( $COP_R$ ) can be calculated from Eq. (8), where  $\eta_{II}$  denotes the second principle efficiency and  $COP_{R,id}$  stands for the coefficient of performance of an ideal Carnot cycle. It is given by Eq. (9) as a function of the temperatures of the hot and cold reservoirs ( $T_{hot}$  and  $T_{cold}$ ), assumed to be 30°C and -2°C, respectively, considering a minimum approach temperature of 5°C.

$$COP_R = COP_{R,id} \cdot \eta_{II} \quad (8)$$

$$COP_{R,id} = \frac{1}{\frac{T_{hot}}{T_{cold}} - 1} \quad (9)$$

284

285

286

287

By definition of the  $COP_R$ , the electric power can be computed given the cooling duty to be removed from the system (Eq. (10)).

$$\dot{W}_{el} = \frac{\dot{Q}}{COP_R} \quad (10)$$

288

289 Then, considering the definition of the efficiency of a combined cycle ( $\eta_{CC}$ ) (Eq. (11)), which is  
 290 given by the ratio between the electric power produced by the gas turbine and the steam turbine and  
 291 the heat received by the gas turbine (given by the product of the fuel flow rate and its LHV), the  
 292 amount of equivalent butanol can be determined according to Eq. (12).

293

$$\eta_{CC} = \frac{\dot{W}_{el}}{\dot{m}_{FUEL} \cdot LHV} \quad (11)$$

$$\dot{m}_{FUEL} = \frac{\dot{Q}}{COP_R \cdot \eta_{CC} \cdot LHV} \quad (12)$$

294

295 The electric energy to be provided to pumps and compressors has been assumed to be obtained by  
 296 means of an equivalent butanol-fired combined cycle power plant. The amount of butanol equivalent  
 297 to the required electric energy ( $\dot{W}_{el}$ ) can be determined knowing the efficiency of the combined cycle  
 298 and the fuel lower heating value (Eq. (13)).

299

$$\dot{m}_{FUEL} = \frac{\dot{W}_{el}}{\eta_{CC} \cdot LHV} \quad (13)$$

300

## 301 **6. Results**

### 302 **6.1. Removal Functions**

303 The removal functions used in the simulation of the integrated recovery process with gas stripping  
 304 have been derived for acetone, butanol, ethanol and water based on the results obtained in Aspen  
 305 Hysys<sup>®</sup> [28], as previously stated. The process scheme used in the simulation is the one reported in  
 306 Fig. 4, where the concentrations of the solvents in the stream fed to the stripping column have been  
 307 varied in such a way to cover all the concentrations experienced in the reactor during fermentation.

308 For each solvent, the removal function has been described with a linear dependence on its  
 309 concentration, with the coefficients expressed as a function of the concentration levels of other  
 310 solvents present in the broth. For water, the removal function has been computed from the water-  
 311 butanol selectivity, in turn expressed as a linear function of butanol concentration in the broth.

312 The expression of the removal functions, of their coefficients and the corresponding coefficient of  
 313 determination,  $R^2$ , obtained by comparison with the simulation results are reported in Table 3.

314

**Table 3.** Removal functions for acetone, butanol, ethanol and water used in the modeling of the integrated fermenter-gas stripping process shown in Fig. 4.

<b>Acetone (<math>R^2 = 0.97</math>)</b>	
<b>Removal function</b>	<b>Coefficients</b>
$\eta_A = m_A(c_B) \cdot c_A + q_A(c_B)$	$a_1 = 4.615 \cdot 10^{-5}$
$m_A = a_1 \cdot c_B + a_2$	$a_2 = 5.990 \cdot 10^{-5}$
$q_A = a_3 \cdot c_B^2 + a_4 \cdot c_B + a_5$	$a_3 = -5.191 \cdot 10^{-5}$
	$a_4 = 3.665 \cdot 10^{-2}$
	$a_5 = 6.299 \cdot 10^{-2}$
<b>Butanol (<math>R^2 = 0.99</math>)</b>	
<b>Removal function</b>	<b>Coefficients</b>
$\eta_B = m_B(c_A) \cdot c_B + q_B(c_A)$	$b_1 = -1.954 \cdot 10^{-4}$
$m_B = b_1 \cdot c_A + b_2$	$b_2 = 7.456 \cdot 10^{-3}$
$q_B = b_3 \cdot c_A^2 + b_4 \cdot c_A + b_5$	$b_3 = 0$
	$b_4 = 2.086 \cdot 10^{-3}$
	$b_5 = 1.112 \cdot 10^{-1}$
<b>Ethanol (<math>R^2 &gt; 0.99</math>)</b>	
<b>Removal function</b>	<b>Coefficients</b>
$\eta_E = m_E(c_B) \cdot c_E + q_E(c_B)$	$e_1 = 1.146 \cdot 10^{-5}$
$m_E = e_1 \cdot c_B + e_2$	$e_2 = 2.188 \cdot 10^{-4}$
$q_E = e_3 \cdot c_B^2 + e_4 \cdot c_B + e_5$	$e_3 = -4.994 \cdot 10^{-5}$
	$e_4 = 2.570 \cdot 10^{-3}$
	$e_5 = 8.187 \cdot 10^{-2}$
<b>Water (<math>R^2 = 0.99</math>)</b>	
<b>Removal function</b>	<b>Coefficients</b>
$\sigma_{WB} = \frac{\eta_W}{\eta_B} = m_{WB}(c_A) \cdot c_B + q_{WB}(c_A)$	$wb_1 = 1.292 \cdot 10^{-4}$
$m_{WB} = wb_1 \cdot c_A + wb_2$	$wb_2 = -4.392 \cdot 10^{-3}$
$q_{WB} = wb_3 \cdot c_A^2 + wb_4 \cdot c_A + wb_5$	$wb_3 = 0$
	$wb_4 = 2.570 \cdot 10^{-3}$
	$wb_5 = 8.187 \cdot 10^{-2}$

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## 6.2. Process comparison

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In this section, the results obtained from the simulations of the two processes (namely, the traditional batch fermentation process and the one integrated with gas stripping for solvents recovery, both followed by solvents separation by a five-column distillation train) are discussed for comparison purposes.

The concentration profiles inside the reactor obtained for the batch fermentation process are reported in Fig. 6. After 15 minutes, the concentrations of butanol and butyric acid reach the critical level for the inhibition of the fermentation microorganism, causing the interruption of the cell growth. At the same time, a reduction in the sugar consumption rate can be observed. The fermentation is ceased after 50 hours, when the butanol concentration is 20 g/L (37 g/L total ABE) and the sugar

326 concentration is 10 g/L, corresponding to a substrate conversion equal to 88%. This situation is  
327 representative of a typical batch fermentation, in which the culture is unable to utilize all sugars due  
328 to the toxic effect of butanol.

329

330 [FIGURE 6 HERE]

331

332 For this case, the composition of the broth at the end of the fermentation is reported in Table 4.  
333 This stream is sent to the beer column of the distillation train for the recovery of the produced  
334 solvents. In the simulation of the train, only the desired solvents and water have been considered: the  
335 actual composition of the broth used in the simulations is reported in Table 5.

336

**Table 4.** Broth composition at the end of the batch fermentation process.

<b>Component</b>	<b>g/L</b>
Biomass	3.05
Sugar	10.00
Butyric acid	1.17
Acetic acid	1.11
Acetone	16.04
Butanol	20.11
Ethanol	1.11

337

**Table 5.** Composition (mole fraction) of the broth at the end of the batch fermentation process sent to the distillation train.

<b>Component</b>	<b><math>x_i</math></b>
Acetone	$4.946 \cdot 10^{-3}$
Butanol	$4.858 \cdot 10^{-3}$
Ethanol	$4.333 \cdot 10^{-4}$
Water	0.9898

338

339 In order to reduce the toxic effect of butanol on cells, an integrated process of fermentation with  
340 gas stripping has been designed and simulated as described in Section 4. For this system, butanol and  
341 the other solvents are produced by the microorganism and removed through the stripping unit. The  
342 rate of butanol removal can be raised by increasing the dilution factor (i.e., the flowrate of the broth  
343 withdrawn from the reactor and sent to the stripping column), causing a reduction of the butanol  
344 concentration in the bioreactor. This would give a dilute butanol condensate, without obtaining the  
345 desired phase separation. Therefore, the dilution factor has been selected in such a way to obtain a  
346 butanol concentration in the fermenter lower than its toxic level, but high enough to obtain a

347 sufficiently concentrated butanol condensate. The selected dilution ratio of  $0.32 \text{ h}^{-1}$  allows to keep a  
348 constant butanol concentration of  $10 \text{ g/L}$  in the reactor during the whole fermentation.

349 The results obtained for this configuration, in terms of concentration profiles in the fermenter, are  
350 reported in Fig. 7.

351  
352 [FIGURE 7 HERE]

353  
354 The gas stripping process begins after 12 h, when the butanol concentration reaches  $10 \text{ g/L}$ , in order  
355 to avoid the inhibitory effect. The butanol concentration remains almost constant due to the  
356 continuous removal by the stripping column. A sugar conversion of 100% is obtained in 25 h,  
357 therefore reducing by 50% the fermentation time.

358 Fig. 8 shows the mass flowrates of the solvents removed through the stripping column during the  
359 fermentation, calculated according to Eq. (5).

360  
361 [FIGURE 8 HERE]

362  
363 The solvents stripped during the process are condensed and the obtained organic and aqueous  
364 phases are, then, collected in storage tanks. The final global composition of the condensate is reported  
365 in Table 6.

366  
**Table 6.** Composition (mole fraction) of the liquid phase collected in the gas stripping condensate of the integrated fermenter-gas stripping process shown in Fig. 4.

<b>Component</b>	<b>Global</b>	<b>Aqueous phase</b>	<b>Organic phase</b>
Acetone	$6.50 \cdot 10^{-3}$	$6.50 \cdot 10^{-3}$	$2.02 \cdot 10^{-2}$
Butanol	$2.75 \cdot 10^{-2}$	$2.75 \cdot 10^{-2}$	$4.12 \cdot 10^{-1}$
Ethanol	$1.90 \cdot 10^{-3}$	$1.90 \cdot 10^{-3}$	$6.07 \cdot 10^{-3}$
Water	0.9641	0.9641	0.5614

367  
368 The total recovery of butanol in the condensate is equal to 44%, the rest remaining in the  
369 fermentation broth inside the reactor.

370 The butanol concentration in the condensate is high enough to obtain the phase separation,  
371 generating an organic phase consisting of  $644.4 \text{ g/L}$  of butanol,  $24.7 \text{ g/L}$  of acetone and  $5.9 \text{ g/L}$  of  
372 ethanol and an aqueous phase consisting of  $101.5 \text{ g/L}$  of butanol,  $18.8 \text{ g/L}$  of acetone and  $4.4 \text{ g/L}$  of  
373 ethanol.

374 However, it is worth highlighting that the organic phase contains only 0.1% of the recovered  
375 butanol, since the organic phase fraction after phase separation is only equal to  $8.8 \cdot 10^{-5}$ . Therefore,  
376 most of the butanol has still to be recovered from the aqueous phase and both these streams are further  
377 treated in the downstream distillation train. For downstream processing, the aqueous phase of the

378 condensate is mixed with the fermentation broth remained in the reactor, giving the final composition  
 379 reported in Table 7, and then fed together to the beer column. The organic phase, due to its high  
 380 solvent content, is fed directly to the acetone column, bypassing the beer column.

381

**Table 7.** Composition (mole fraction) of the fermentation broth remaining in the reactor at the end of the integrated fermenter-gas stripping process and of the final aqueous phase obtained by mixing it with the aqueous phase of the condensate obtained downstream of gas stripping.

<b>Component</b>	<b>Fermentation broth</b>	<b>Final aqueous phase</b>
Acetone	$2.54 \cdot 10^{-3}$	$2.84 \cdot 10^{-3}$
Butanol	$2.38 \cdot 10^{-3}$	$4.29 \cdot 10^{-3}$
Ethanol	$3.61 \cdot 10^{-4}$	$4.78 \cdot 10^{-4}$
Water	0.9947	0.9924

382

383 The results for the two studied configurations in terms of energy requirements are reported in Table  
 384 8.

385

**Table 8.** Comparison of process performances in terms of process productivity and energy consumption between the traditional batch fermentation process and the integrated fermenter-gas stripping process.

	<b>Batch fermentation</b>	<b>Integrated product recovery by gas stripping</b>
<b>Butanol productivity [t/y]</b>	50,000	91,225
<b>Energy requirement for distillation [kgB/h]</b>	2,915.73	6,013.60
<b>Energy requirement for gas stripping [kgB/h]</b>	-	8832.47
<b>Total energy requirement [kgB/h]</b>	2,915.73	14,846.07
<b>Total specific energy requirement [kgB/kgB]</b>	0.47	1.29

386

387 Process simulations for the traditional batch fermentation scheme have been performed for a 50,000  
 388 t/y butanol production plant [11], as representative of a retrofit of a bioethanol plant. Here the energy  
 389 consumption is due to the heat duty at the beer column, water and butanol columns, to the pumps and  
 390 to the vacuum pumps required to get the required void in the acetone and ethanol columns. With this

391 configuration of the distillation train, the total specific energy requirement is equal to 0.47 kg of  
392 butanol per kg of butanol produced.

393 Considering the integrated process, the continuous product removal from the fermenter allows to  
394 reduce the fermentation time by 50%, as previously stated. Therefore, the process productivity,  
395 considering the same reactor volume used in the previous scheme, is almost double. The specific  
396 consumption of the distillation train slightly increases from 0.47 kg<sub>B</sub>/kg<sub>B</sub> of the batch process to 0.52  
397 kg<sub>B</sub>/kg<sub>B</sub>. In fact, even if the condensate obtained downstream of gas stripping undergoes a phase  
398 separation, the flowrate of the generated organic phase that can bypass the beer column is almost  
399 negligible and does not have a significant impact on the energy requirement of the train. Also, the  
400 aqueous phase obtained by mixing the aqueous phase of the condensate with the broth remaining in  
401 the reactor at the end of the fermentation exhibits a lower solvents concentration (as reported in Table  
402 7) with respect to the batch case (Table 5). This can be explained considering that gas stripping has a  
403 low selectivity and removes, together with butanol, a large amount of water from the fermenter. At  
404 the same time, the specific energy input for the gas stripping process is high and equal to 0.77 kg<sub>B</sub>/kg<sub>B</sub>.  
405 This essentially consists of the duty required at the condenser for solvents recovery from the gas  
406 stream, due to the low concentration of ABE in the recovered stream and the associated difficulty of  
407 separating them from the gas stream. As a result, for the integrated recovery process by gas stripping,  
408 the total specific energy requirement is, therefore, higher and equal to 1.29 kg<sub>B</sub>/kg<sub>B</sub>. These results  
409 suggest that the consumption of the integrated process is almost three times greater than the traditional  
410 batch process, as also confirmed by Outram et al. [11].

411 In conclusion, the application of an integrated recovery process allows to increase the process  
412 productivity, but at the expense of an increase in the whole process energy demand.

413

## 414 **7. Conclusions**

415 Over the years, many separation techniques have been proposed or developed to mitigate the  
416 problem of product inhibition which affects the ABE fermentation process. Among them, gas  
417 stripping is considered as a viable solution for that.

418 This work has focused on its application to a classical batch fermenter, to evaluate the advantages  
419 it offers and the possible drawbacks. To accomplish that, the fermentation process has been integrated  
420 with the gas stripping unit by solving material balance equations which involve some removal  
421 functions to take into account the withdrawal of the fermentation broth from the reactor and its  
422 processing in the stripping column. The two liquid phases which are formed by cooling down the  
423 vapor stream leaving the stripping column have been further treated in a five-column distillation train.  
424 The same downstream scheme has been also considered for the recovery of the solvents from the

425 broth remained in the reactor at the end of a classical batch fermentation process (thus not integrated  
 426 with the gas stripping unit), in order to compare the two process solutions. To account for the energy  
 427 requirements of each of them, an energy analysis has been performed on the basis of the net equivalent  
 428 fuel approach.

429 The obtained results have suggested that the application of gas stripping as integrated recovery  
 430 technique allows to increase the process productivity, but at the expense of an increase in the whole  
 431 process energy demand. It may be interesting to carry out the comparison also considering other  
 432 possible operation modes for both the reactor and the gas stripping recovery technique.

433  
 434

## 435 **Nomenclature**

436

### 437 **Acronyms**

<i>ABE</i>	Acetone-Butanol-Ethanol
<i>COP</i>	Coefficient of Performance
<i>EoS</i>	Equation of State
<i>LHV</i>	Lower Heating Value [MJ/kg]
<i>LLE</i>	Liquid-Liquid-Equilibrium
<i>LP</i>	Low-pressure
<i>NRTL</i>	Non-Random Two-Liquid
<i>RK</i>	Redlich-Kwong
<i>VLE</i>	Vapor-Liquid-Equilibrium

438

### 439 **Symbols**

$a_{ij}$	Binary interaction parameter of the NRTL-RK model
$a_k$	Coefficients in the definition of $m_A$ ( $k = 1, 2$ ) and $q_A$ ( $k = 3, 4, 5$ )
$b_{ij}$	Binary interaction parameter of the NRTL-RK model
$b_k$	Coefficients in the definition of $m_B$ ( $k = 1, 2$ ) and $q_B$ ( $k = 3, 4, 5$ )
$C$	Concentration, [g/L]
$D_f$	Dilution factor, [1/h]
$e_k$	Coefficients in the definition of $m_E$ ( $k = 1, 2$ ) and $q_E$ ( $k = 3, 4, 5$ )
$f$	Functions defined in Eqs. (1)-(2)
$m_j$	Coefficients in the removal functions ( $j = A, B, E, WB$ )
$m$	Mass [kg]
$\dot{m}$	Flow rate, [kg/s]
$P$	Pressure, [atm]
$\dot{Q}$	Duty, [kW]
$q_j$	Coefficients in the removal functions ( $j = A, B, E, WB$ )
$R^2$	Coefficient of determination
$r$	Reaction rate, [g/L/h]
$T$	Temperature, [K]
$t$	Time, [h]
$V_F$	Fermenter volume [L]
$\dot{W}_{el}$	Electric power, [kW]
$w_{bk}$	Coefficients in the definition of $m_{WB}$ ( $k = 1, 2$ ) and $q_{WB}$ ( $k = 3, 4, 5$ )
$x$	Mole fraction, [-]



440

441 **Greek symbols**

$\alpha$	Non-randomness parameter of the NRTL-RK model
$\eta_{boiler}$	Boiler efficiency, [-]
$\eta_i$	Removal function for $i$ -th component, [-]
$\eta_{CC}$	Combined-cycle efficiency, [-]
$\eta_{II}$	Second principle efficiency, [-]

442

443 **Subscripts**

$A$	Acetone
$B$	Butanol
$ba$	Butyric acid
$cold$	Cold reservoir
$E$	Ethanol
$F$	Referred to the fermenter/end of the fermentation
$FUEL$	Referred to the fuel
$hot$	Hot reservoir
$i$	$i$ -th component
$R$	Referred to a refrigeration cycle
$R_{id}$	Referred to an ideal Carnot cycle
$S$	Referred to stripped components
$W$	water

444

445

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528

529 **Figure 1.** Comparison between VLE and LLE experimental data [16-23] for the system water-butanol  
530 ( $P = 1$  atm) and prediction by the Aspen Plus<sup>®</sup> [15] NRTL-RK model with default (dotted line) and  
531 regressed (solid line) parameters.

532

533 **Figure 2.** LLE experimental data (symbols from [24]) for the system acetone-water-butanol and  
534 prediction by the Aspen Plus<sup>®</sup> [15] NRTL-RK model with default parameters (dotted line) and with  
535 regressed parameters (solid line) at a temperature of: a) 293.15 K; b) 313.15 K.

536

537 **Figure 3.** LLE experimental data (dots from [25]; triangle down from [16]) for the system ethanol-  
538 water-butanol and prediction by the Aspen Plus<sup>®</sup> [15] NRTL-RK model with regressed parameters  
539 (solid line) at 298.15 K.

540

541 **Figure 4.** Scheme of the integrated fermenter-stripping process.

542

543 **Figure 5.** Scheme of the distillation train used for the ABE separation downstream of the integrated  
544 fermentation-stripping process (green arrows denote electrical energy requirements, red arrows  
545 denote heat duties, blue arrows denote cooling duties).

546

547 **Figure 6.** Concentration profiles obtained from the modeling of the batch fermentation process: (—  
548 · —, violet) sugar, (····, black) biomass, (---, light blue) acetic acid, (---, orange) butyric acid, (—  
549 —, green) acetone, (—, red) butanol, (—, blue) ethanol.

550

551 **Figure 7.** Concentration profiles obtained from the modeling of the integrated fermenter-gas stripping  
552 process: (— · —, violet) sugar, (····, black) biomass, (---, light blue) acetic acid, (---, orange)  
553 butyric acid, (—, green) acetone, (—, red) butanol, (—, blue) ethanol. The vertical dotted line  
554 indicates when the gas stripping starts.

555

556 **Figure 8.** Profiles of mass flowrates removed from the fermenter during the integrated fermenter-gas  
557 stripping process: (---, light blue) water, (—, green) acetone, (—, red) butanol, (—, blue)  
558 ethanol. The vertical dotted line indicates when the gas stripping starts.

559

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562