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# Selective separation of flavour-active compounds from strip gas using frictional diffusion



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#### ABSTRACT

Attaining constant flavour composition in products that are produced batch-wise, such as beer, is not trivial given the inherent variability in fermentation.  $CO_2$  stripping is feasible but unselective. Condensation of the flavour is possible but energy intensive. We here propose the use of frictional diffusion (also called FricDiff), which is based on differences in diffusion rates in a sweep or carrier gas such as  $CO_2$  through an inert porous medium. Application of a slight counter-flow of the sweep gas can be used to adapt the selectivity between different flavours. It is shown that from a difference in diffusion rate of 25%, a selectivity of more than 10 can be obtained between ethyl acetate and isoamyl acetate, albeit at the cost of the flavour flux through the porous barrier.

### 1. Introduction

Flavours are an important element in foods, but their profile may vary, due to variation in the production process, or in raw materials [1]. An example is the brewing of beer. The fermentation process is generally operated as batch process, and slight variation in the fermentation or in the exact composition of the raw materials, such as the malt, may give rise to variations in flavour profile. To avoid the beer to vary in quality to the consumer, the flavour of the beer may be adapted by selectively removing some of the flavours, while retaining others.

Volatile flavours can be controlled through various recovery, separation or removal processes [2]. Vacuum distillation that is known to protect nutrients, is a classical method of volatile separation; however, the selectivity is toward lighter compounds such as ethyl acetate and ethanol. This implies that a second step separation step is needed to process the effluent vapour and obtain the desired fraction and return the resulting fraction to the beer.

Some membrane separation techniques such as reverse osmosis [3] have been used to selectively remove the ethanol whereas, nanofiltration has been described to separate flavours next to ethanol [4]. Using these two methods would also require a recycling loop as described in the previous section for vacuum distillation. Pervaporation allows more selective removal of flavours, depending on the membrane, but is relatively intensive in energy, and the separation is mostly dependent on the properties of the membrane; hence it does not leave much flexibility

for adaptation of the separation to mitigate batch to batch variation [5,6].

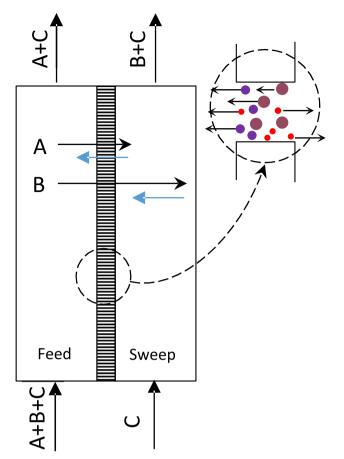
From the above it is clear that there is no single step separation technique that can be used to target flavour compounds specifically. Here we propose to start with stripping the beer, with for example CO<sub>2</sub>, which is a naturally occurring component in the beer itself is an attractive primary stage. This will remove the flavours in proportion to their volatility. The actual composition can be changed by selectively removing them from the stripping gas. Since compression and cooling processes are highly energy intensive, recovery of compounds directly from the gas phase is favourable. Alternatively, the condensate can be treated as reported by Saffarionpour and co-workers [7] using adsorption, for example with active carbon or zeolites, to selectively remove the flavours. While this does allow flexibility in terms of separation, it is a semi-batch process, in which the columns regularly need to be regenerated, which complicates process operation.

For correcting dynamic variations, as opposed to structural deviations in flavour profile, a flexible process is necessary, which can be quickly adapted to changes in fresh beer quality, without costing too much energy or other auxiliaries. Geboers et al. [8] proposed the frictional diffusion (FricDiff) process principle for azeotrope breakage. Different from existing methods, it has the possibility to adapt the selectivity between different components, and therefore it may also be of value for the selective, adjustable removal of flavours from a strip gas.

As can be seen in Fig. 1, frictional diffusion relies on differences in

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**Fig. 1.** The principle of frictional diffusion. The strip gas D coming from the beer containing flavours A and B, is contacted to a porous barrier. The same gas C is also flowing over the other side of the barrier. The flavour components A and B diffuse through the gas that is present in the pores of the barrier. Depending on their rates of intermolecular and Knudsen diffusion, they will diffuse at different rates. A slight difference in pressure over the barrier changes the selectivity between the two flavour molecules.

diffusion rate between the flavours. By simultaneously imposing a small convective flow of the strip gas against the flavour diffusion, selective flavour removal can be achieved. Thus, any change in the flavour profile of the raw material can be mitigated by the adaptation of the pressure over the barrier.

We here present the feasibility of the frictional diffusion principle for dynamic adaptation of flavour removal, adopting the Maxwell-Stefan approach that Geboers and Kerkhoff introduced. We will show the possibility of having selective removal of flavour, show how this can be adapted through imposing a counter flux of the strip gas, and how different system parameters may influence the results.

Fig. 2 illustrates the overall system that we envision the frictional diffusion module will be part of. CL1 is the stripping column from which our feed solution stems, and that contains  $\rm CO_2$  and flavours. This gas is next contacted in M1 with a secondary gas phase in our case carbon dioxide through a porous barrier. Based on the concentration gradient between the feed side and pure carbon dioxide diffusion of various components takes place. Components with low diffusivity can be retained in the feed by applying elevated sweep gas pressure. It is also possible to tune the driving force for separation of a certain component by enriching the sweep gas with that component. The process is expectd to further contain a sweep gas profiler (CL2) and a scrubber (CL3).

Since  $CO_2$  is a naturally occurring gas in fermentation, it is our sweep gas of choice, also to avoid waste. In this paper, we focus on the gas contactor only, in which the sweep gas can either be a pure gas or

can be profiled by adding ethanol and/or water. The effects that can be created in this way will be evaluated using the FricDiff approach that is discussed next.

#### 2. Theory

#### 2.1. The frictional diffusion concepts

Frictional diffusion was introduced by Geboers et al. [8] initially as an alternative technique for azeotrope mixture separation. Within this approach, a feed mixture and a sweep gas are separated by a non-selective porous layer (barrier). Components will diffuse through the barrier with different velocities, depending on the diffusivities. Pressure can be imposed over the barrier to influence the permeation rates, which will induce flow. This may lead to suppression of the more slowly diffusing components, while faster diffusion components may still be able to reach the other side of the membrane. The detailed concept is presented in earlier publications [9–13].

We assume a flat sheet membrane made of an inert material (more properties will be detailed later) with negligible pressure drop or differences in concentrations along its length, due to relatively fast crossflow on both sides. Axial concentration gradients at the sweep side and at the feed side are also assumed to be small and have not been considered in this study. The motion of the gases inside the pores of the barrier can be described with [8]

$$\nabla p_i = RT \frac{\tau^2}{\varepsilon} \left[ -\sum_{j=1}^n \frac{(p_j N_i - p_i N_j)}{p_t D_{ij}} - f_{im} N_i \right]$$
(1)

Here,  $p_i$  is the partial pressure of component i,  $N_i$  the molar flux,  $p_t$  the total pressure,  $\tau$  the tortuosity of the pores inside the barrier, here taken 1.3 a typical value found in FricDiff investigations [9,10,12,13], and  $\epsilon$  the porosity of the barrier, chosen at 0.5 which is a very acceptable value for membrane porosity [14]; R and T are the gas constant and the temperature, respectively. The thickness of the barrier L is 0.5 mm.  $\nabla p$  is the local partial pressure gradient. The term  $f_{im}N_i$  represents the friction between the diffusing component i and the pore walls in the barrier, through viscous friction and through Knudsen interaction (collisions between molecules of i and the barrier pore walls). For the wall-friction coefficients  $f_{im}$  Kerkhof and Geboers [15], proposed

$$f_{im} = \left(D_i^K + \frac{p_i r_p^2}{8\kappa_i}\right)^{-1} \tag{2}$$

In which  $r_p$  is the radius of a pore which is assumed to be cylindrical,  $p_i$  is the partial pressure of component i, and  $\kappa_i$  is the fractional viscosity of component i. The Knudsen diffusivity may be approximated under the assumption of complete diffusive reflection at the wall and the absence of any molecule-molecule interaction [16] by:

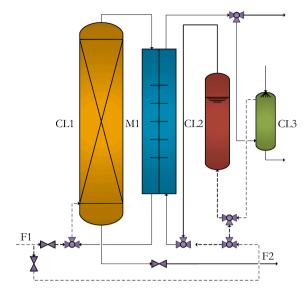
$$D_i^K = 0.89 r_p \left( \frac{8}{\pi} \frac{RT}{M_i} \right)^{1/2} \tag{3}$$

with  $M_i$  the molecular weight of component i. The fractional viscosity  $\kappa_i$  is defined with

$$\kappa_i = \frac{x_i \eta_i^0}{\sum_{j=1}^n x_j \xi_{ij}} \tag{4}$$

with  $\eta_i^0$  the viscosity of the pure gas i, and  $\xi_{ij}$  the Wilke [17] parameter, given by

$$\xi_{ij} = \frac{\left[1 + \left(\frac{\eta_i^0}{\eta_j^0}\right)^{0.5} \left(\frac{M_j}{M_i}\right)^{0.5}\right]^2}{\left[8\left(1 + \frac{M_i}{M_j}\right)\right]^{0.5}}$$
(5)



Code	Description
CL1	Stripping Column
CL2	Sweep Gas Profiler
CL3	Scrubber
M1	Gas Flow Contactor
F1	$CO_2$
F2	Product Beer

Fig. 2. Schematic representation of the proposed location for FricDiff (Dash lines are CO<sub>2</sub>).

The viscosities of the pure components,  $\eta_i^0$  , are calculated using the DIPPR method:

$$\eta_{G_i} = \left[ \frac{B_{1i} T^{B_{3i}}}{1 + \frac{B_{3i}}{T}} \right] \quad \text{for} \quad T_{min} > T > T_{max}$$
(6)

in which  $B_1$ ,  $B_2$ ,  $B_3$  are dependent on the component i (see Appendix A Table A2). The binary Maxwell-Stefan diffusivities  $D_{ij}$  can be described using the following correlation [18]:

$$D_{ij} = \frac{3.16 \cdot 10^{-8} T^{1.75}}{p_t (v_t^{1/3} + v_j^{1/3})^2} \left( \frac{1}{M_i} + \frac{1}{M_j} \right)^{\frac{1}{2}}$$
(7)

in which  $v_i$  is the molar diffusion volume of component i (Table 1).

The Maxwell-Stefan diffusion coefficients obey the reciprocal relations  $D_{ij} = D_{ji}$ . To evaluate the use of frictional diffusion, the following definition of the selectivity between components i and j is used:

$$\alpha_{i,j} = \frac{N_i/x_i}{N_j/x_j} \tag{8}$$

## 3. Results and discussions

## 3.1. Simplified system with $CO_2$ and two flavours

Substituting Eq. (2) in 1, we assume that the flavour components i are dilute and that we have only carrier gas c ( $p_1 \ll p_c$ ), we can separate the fluxes of the several flavours, since they will only have interaction with the sweep gas. Thus, for each flavour we obtain

$$\frac{dp_i}{dx} = RT \frac{\tau^2}{\varepsilon} \left[ \frac{(p_i N_c - p_c N_i)}{p_i D_{ij}} - \frac{N_i}{D_i^K + \frac{p_i r_p^2}{8\kappa_i}} \right]$$
(9)

Since that flavours are dilute,  $p_c \approx p_t$ , and we get

$$\frac{dp_i}{dx} = RT \frac{\tau^2}{\varepsilon} \left| \frac{p_i N_c}{p_i D_{ij}} - \frac{N_i}{D_{ij}} - \frac{N_i}{D_i^K + \frac{p_i r_p^2}{8\kappa_i}} \right| \tag{10}$$

Here, we should bear in mind that  $N_c$  is either zero (no pressure difference), or negative, in case we want to reduce the flavour diffusion to the sweep side by imposing a counter-flux against the direction of diffusion of the flavours. Thus, we find that

$$\frac{dp_i}{dx} = \left(\frac{RT}{p_i D_{ic}} \frac{\tau^2}{\varepsilon} \cdot N_c\right) \cdot p_i - \left(RT \frac{\tau^2}{\varepsilon} \cdot N_i\right) \cdot \left(\frac{1}{D_{ic}} + \frac{1}{D_i^K} + \frac{p_i r_p^2}{8\kappa_i}\right)$$
(11)

Assuming a tubular geometry, with a moderate gas flow rate of 1m/s, the Biot number can be estimated through

$$Bi = \frac{hL\tau^2}{D_{ic}}$$
 (12)

and is found to be 4.13, confirming our assumption that mass transfer is limited by internal mass transfer. Hence, we will assume that the partial pressures at the entrance of the barrier pores at the feed side are equal to the partial pressures in the feed and that the partial pressures at the end of the pores at the sweep side are equal to the partial pressures in the sweep phase. We assume that the partial pressures in the sweep phase are negligibly small.

Eq. (9) is integrated using a fourth-order Runge-Kutta algorithm

 Table 1

 Gas Phase Diffusion Coefficient (Eq. (1)) and diffusion volumes.

	ν*	Water	Ethanol	Ethyl acetate	Isoamyl acetate	Isoamyl alcohol	$CO_2$
Water	1.27E-05	-	1.59E-05	1.14E-05	1.05E-05	8.81E-05	2.19E-05
Ethanol	5.04E-05	1.59E-05	_	5.58E-06	5.21E-06	4.34E-06	1.10E-05
Ethyl acetate	9.28E-05	1.14E-05	5.58E-06	_	3.58E-06	2.93E-06	7.85E-06
Isoamyl acetate	1.12E-04	1.05E-05	5.21E-06	3.58E-06	_	2.76E-06	7.28E-06
Isoamyl alcohol	1.54E-04	8.81E-06	4.34E-06	2.93E-06	2.76E-06	_	6.10E-06
$CO_2$	2.69E-05	2.19E-05	1.10E-05	7.85E-06	7.28E-06	6.10E-06	-

<sup>\*</sup> Diffusion volumes are calculated using Lightfoot [19].

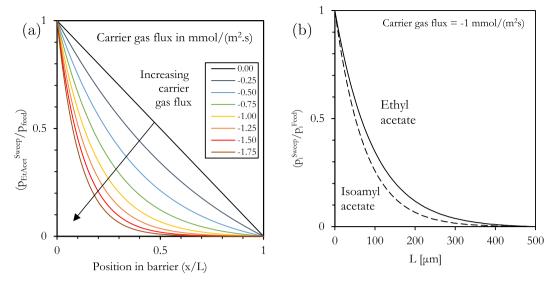


Fig. 3. Normalized partial pressure profiles of flavours. Left hand graph: partial pressure profiles of ethyl acetate with increasing counter-flux of  $CO_2$  through the barrier. Right hand graph: partial pressure profiles of ethyl acetate (–) and isoamyl acetate (–) through the barrier, with a sweep-to-feed flux of the carrier gas  $(CO_2)$  of  $1 \text{ mol/(m}^2\text{s})$ . The slight difference in the diffusion coefficient of the two flavours causes a stronger drag on isoamyl acetate than on ethyl acetate; hence its overall flux through the carrier is reduced disproportionally, giving an unexpectedly large selectivity.

with 100 steps over the barrier (smaller steps did not change the results), in which the flavour flux is varied until the partial pressure of the flavour at the sweep side is exactly zero, using a nonlinear minimization procedure with its convergence tolerance set at  $10^{-15}$ . Typical concentration profiles for ethyl acetate an isoamyl acetate through the barrier are shown in Fig. 3; Fig. 3a shows the normalized partial pressure profiles of ethyl acetate with different counter-fluxes of the strip gas; Fig. 3b shows the different partial pressure profiles through the barrier for ethyl acetate and isoamyl acetate, at one particular counter-flux.

Fig. 3 Shows that at zero counter-flux, the concentration profile is completely straight, which is logical given that the barrier was assumed to be homogeneous. At a non-zero counter-flux of the strip gas, the partial pressure profiles become non-linear, and reduce the flux of the flavours towards the sweep gas side. Faster diffusion components such as ethyl acetate are less hindered by the counter-flow, but the profile of isoamyl acetate, which has a somewhat lower mutual diffusion

coefficient with CO2, is reduced markedly stronger.

By changing the sweep-to-feed flux of the carrier gas, we can therefore change the ratio of the two flavour fluxes. A zero-carrier gas flux gives unbiased diffusion of the flavours through the stagnant carrier gas inside the barrier pores, resulting in a selectivity of 1.282, which is very close to the ratio of the two flavour- $CO_2$  mutual diffusion coefficients, which is 1.287.

An increase in the sweep-to-feed carrier gas flux affects the slower diffusing components disproportionally strong relative to faster diffusing molecules. Therefore, imposing a counter-flux increases this selectivity. Fig. 4a shows that the selectivity between ethyl acetate and isoamyl acetate can become better than 10, even though their diffusion coefficients are only 29% different. Fig. 4b confirms that this is because of the difference in the intermolecular diffusion coefficient with  ${\rm CO}_2$ . If a barrier would be used with smaller pores, then the Knudsen diffusion becomes more important. Knudsen diffusion takes place between the flavours and the barrier pore walls, which are stagnant. This is in

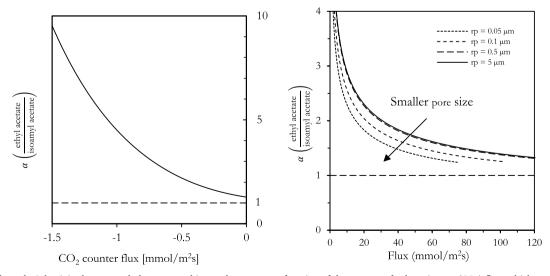


Fig. 4. Left hand graph: Selectivity between ethyl acetate and isoamyl acetate as a function of the sweep-to-feed carrier gas (CO<sub>2</sub>) flux; which are negative because the carrier gas flows from sweep to feed, while the flavours diffuse from feed to sweep phase. Right hand graph: Influence of the pore size of the barrier on the selectivity and flavour fluxes obtained. At smaller pore size, Knudsen diffusion starts to become more important' at larger pore size, intermolecular diffusion rates dominate (calculated with -1 mmol/m<sup>2</sup>s CO<sub>2</sub> counter flux).

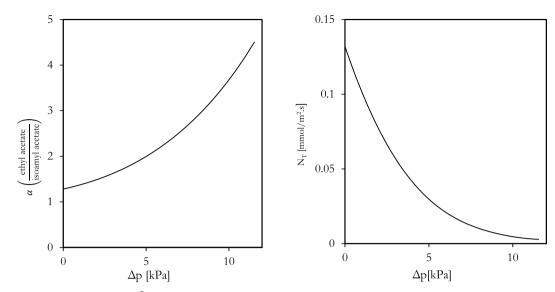


Fig. 5. Selectivities (left) and fluxes in mmol/(m<sup>2</sup>s) (right), as a function of the applied pressure over the barrier. With a pressure drop of only 6 kPa, one can already obtain a substantially elevated selectivity and still a reasonable flux.

contrast to the intermolecular diffusion between the flavours and the strip gas  $CO_2$ : this  $CO_2$  gas can flow, and this flow can compensate the intermolecular diffusion between flavours and  $CO_2$ . For Knudsen diffusion, there is however no influence of any flow of the strip gas. Therefore, a barrier with pores that are smaller than around  $0.5\,\mu m$ , will show a reduced effect of the counter flow of  $CO_2$ .

The fact that we can alter the separation selectivity between the two flavours by changing the sweep-to-feed counter-flux of the carrier gas makes it fundamentally different from other separation processes. For example, a membrane-based vapour permeation or pervaporation process will have an intrinsic selectivity based on the permeability of the components, which are properties of the membrane material. An adsorption process, such as a molecular sieve, will exhibit selectivity based on the surface adsorption affinities of the components, which, once more, are material properties in this case of the adsorbents. In frictional diffusion, the separation is created by the process conditions, especially the pressure of the strip/sweep gas over the membrane, to create the counter-flux of the strip/sweep gas. The typical pressures needed to achieve these counter-fluxes are quite moderate, as is shown in Fig. 5.

While the pressures needed to impose relevant counter-fluxes of the

strip/sweep gas, in this case  $CO_2$ , the specific requirements of the process will dictate what compromise is needed between the selectivity and the flux of the flavours through the barrier. A larger selectivity will imply a lower flux, and hence a larger barrier surface area will be required. Since there are no large pressure differences in the process, and the barrier is only contacted with gases on both sides, one may choose for modules that have a very high surface-to-volume ratio, for example using hollow fibres, which may have a surface area – to volume ratio between 7 000 and 13 000  $m^2/m^3$  [20].

We here assumed a barrier that is  $50\,\mu m$  thick. Fluxes can be improved by using a thinner barrier, for example by using a porous top layer on a more open supportive membrane; however, the carrier counter-flux will also be proportionally larger. The carrier flux is an important parameter for system design, as the system will feed some sweep gas (CO<sub>2</sub>) from the sweep side towards the strip side. This may not be a problem, as some of the CO<sub>2</sub> could leave the system as it will dissolve in the beer that is being stripped. If this is not sufficient, one could allow the volume of the feed side strip gas phase to slowly expand (during batch treatment) or one could remove the CO<sub>2</sub> using a small bleed stream. This bleed stream could be recycled again, by condensation of the flavours or by using a selective membrane process,

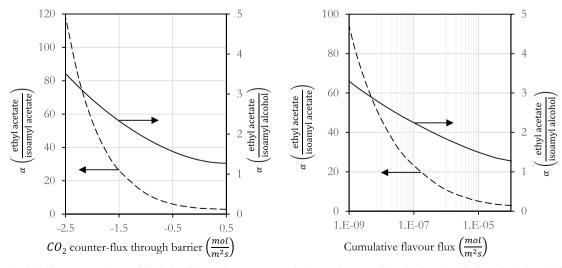


Fig. 6. Selectivities of ethyl acetate over isoamyl alcohol and isoamyl acetate versus the imposed counter-flux of carbon dioxide through the barrier (left hand graph), and versus the total flux of all three flavours combined (right hand graph; both in  $mol/(m^2s)$ ). One can see quite similar behaviour as in the simplified case.

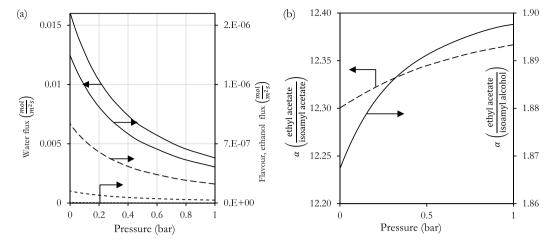


Fig. 7. The fluxes can be increased strongly by operating at reduced pressure. This decreases the friction between the gases, and thus increases the diffusive velocities. The fluxes at zero pressure are solely determined by Knudsen diffusive rates. All values were calculated using a  $CO_2$  counter-flux of  $-0.1 \text{ mol/}(\text{m}^2\text{s})$ .

which would make the  $CO_2$  available again for the sweep side. Since this bleed stream is quite small, this will not significantly impact the overall energy consumption of the process.

#### 3.2. Full system

We can now take the full system into account, using the carrier gas that was contacted with beer. The beer was assumed to have 4 g/L ethanol, 50 mg/L ethyl acetate, 100 mg/L isoamyl alcohol, and 10 mg/L isoamyl acetate. Using Wilson's model to estimate the activity coefficients at 4.78, 90.38, 1245 and 3998, the partial vapour pressures in the carrier gas after having equilibrated with the beer, would be 2339, 44.2, 8.98, 7.13 and 3.27 Pa, for water vapour, ethanol, ethyl acetate, isoamyl alcohol and isoamyl acetate, respectively. Calculating the fluxes with the full Eq. (1), using the same procedure by integrating the set of differential equation using 4th order Runge-Kutta, and then varying the fluxes until the concentrations at the strip side matched the

one in the strip phase, shows that also in such a complex system one can use the  $CO_2$  counter-flux to adjust the selectivities between the different flavours (see Fig. 6a). Application of a larger counter-flux of course again results in lower overall flavour fluxes through the barrier; hence a larger barrier surface area would be needed (Fig. 6b).

We could of course change the thickness of the barrier as shown earlier. There is however another possibility, which is by operating a reduced  $\mathrm{CO}_2$  pressure. Fig. 7 shows that by doing this, one lowers the friction between the different gases, which increases the diffusive velocity of the diffusing components. At very low pressures, one approaches the fluxes obtained based on pure Knudsen diffusion, in which the fluxes are determined by the molecular weights (Fig. 7a). Interestingly, reducing the  $\mathrm{CO}_2$  pressure hardly influences the selectivities of the flavours. Please note that in Fig. 7b, the scales are strongly enlarged; the selectivity between ethyl acetate and isoamyl alcohol changes just 1.6%, and that between ethyl acetate and isoamyl acetate only 0.5%. This this gives us the possibility to improve the fluxes without much

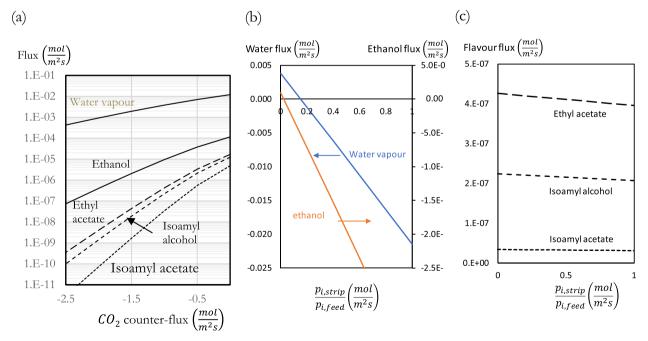


Fig. 8. The left-hand graph shows the fluxes as a function of the  $CO_2$  counter-flux. One can see that by far the largest fluxes are water and ethanol vapour. The middle graph shows that if the vapour pressures of water and ethanol are enlarged from 0 to 100% of their vapour pressure on the feed side, their fluxes can become zero or negative. Careful adjustment of these vapour pressures, therefore, can stop these components from moving through the barrier. The right-hand graph shows that this hardly affects the fluxes of the flavours (or their selectivities).

change of the selectivities.

Fig. 8 shows that when using pure  $CO_2$  as strip phase, also water and ethanol vapour are transferred to the strip side. This can be easily adjusted by allowing a certain vapour pressure of water and ethanol at the strip side as well. Fig. 8b and c show that by imposing on the strip side a fraction of the vapour pressure of water and ethanol on the feed side, one can effectively stop water and ethanol vapour from being transferred. At the same time, the fluxes of the flavours are hardly affected. This implies that by adjusting the composition of the strip gas, one can select only those components that one would wish to transfer to the strip gas.

#### 4. Conclusion

We showed at frictional diffusion (FricDiff) may be a suitable technique that allows the creation of a strip process that can be dynamically adapted to varying requirements on flavour removal. The strip gas is contacted with a porous, inert barrier that is on the other side in contact with the same gas, but without flavours. Different diffusion rates of the flavours through the gas-filled pores of the barrier yield a separation between the flavours. By imposing a small positive pressure over the barrier, a small counter-flux of the sweep/strip gas is created, and both the fluxes of the flavours and the selectivity between these

changes.

It is shown that the selectivity rises disproportionally with the counter-flux, but that the flavour fluxes go down. The pressure needed over the barrier is below 0.15 bar, which means that the selectivity can be quickly adapted to the exact needs of the moment. The absolute pressure (i.e.., not the difference) can be used to increase all fluxes. It is shown that all fluxes rise strongly when reducing the overall pressure of the carrier gas.

Water and ethanol are volatile as well and therefore will be present in the carrier gas after contacting it with beer. Their fluxes can be quite large, but can be completely countered by allowing a certain partial vapour of these two components in the strip side as well. It was shown that this will hardly influence the fluxes of the flavour components.

## **Declaration of Competing Interest**

The authors declared that there is no conflict of interest.

#### Acknowledgements

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#### Appendix A

(See Table A1).

**Table A1**Typical concentration of major volatiles in beer [21].

Ethyl Acetate	Ethanol	Isoamyl Acetate	Isoamyl Alcohol
[mg/L]	[g/L]	[mg/L]	[mg/L]
15–44	36.6–39.9	~4	62–112

Table A2
Parameters used in vapour viscosity DIPPR method (taken from AspenPlus® database.)

Component	Water	Ethanol	Ethyl acetate	Isoamyl Acetate	Isoamyl alcohol	$CO_2$
B1 [Pa.s]	1.71E-08	1.06E-07	3.21E- 06	8.93E-08	8.90E-08	2.14E-6
B2 [-]	1.11	0.81	0.36	0.789	0.80	0.46
B3 [K]	0	52.7	667	89.73	77.65	290
$T_{min}[K]$	273.16	200	189.6	194.65	155.95	194.67
T <sub>max</sub> [K]	1073.15	1000	1000	1000	1000	1500

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