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EFFECT OF CONVECTIVE MASS TRANSFER ON BEER DIAFILTRATION

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The industrial application of dialysis for the de-alcoholization of beer is based on a diafiltration method, because a small positive transmembrane pressure on the beer side of the membrane is necessary to prevent ingress of water by osmosis and resulting dilution of the beer. This pressure difference introduces the new driving and convective mass transfer mechanism, which takes place simultaneously with the diffusive mechanism (specific for dialysis). The question is the influence which it has on the overall mass transfer phenomenon and selectivity of the alcohol/extract separation. An experimental investigation of beer diafiltration using two different membranes (cellulose based Cuprophane and polysulfone) was performed at different transmembrane pressure differences and flow rates. On the basis of the experimentally measured inlet and outlet concentrations the convective components of the alcohol and extract mass fluxes as well as the overall separation factor were determined. This data was analysed and compared. The results indicate that convective mass transfer stimulated extract transfer and losses and thus diminished the efficiency of selective alcohol/extract separation in beer dialysis.

Key Words: *Diafiltration, beer, diffusion, convection, Cuprophane, polysulfone.*

NOTATION

A_m (m^2)	membrane surface area
c (g/cm^3)	concentration
J_{uf} ($cm^3/min/m^2$)	ultrafiltration volumetric flux
L_p ($cm^3/min/m^2/bar$)	membrane hydraulic permeability
N ($cm^3/min/m^2$)	normalised mass flux
Q (cm^3/min)	volumetric flow rate
TMP (bar)	transmembrane pressure difference
ΔP	pressure drop

Subscripts

avg	average
A	alcohol
b	beer
E	extract
n	nominal
s	solute
uf	ultrafiltration
w	water
1	feed
2	dialysate

Superscripts

i	inlet
o	outlet
D	dialysis
F	ultrafiltration
D/DF	diffusive component of diafiltration
C/DF	convective component of diafiltration

INTRODUCTION

Dialysis is an accepted method for the production of alcohol free, low-alcohol and low-calorie beers[3,7]. Theoretically it operates with zero transmembrane pressure difference (TMP) but in industrial conditions a TMP is applied in order to compensate for the osmotic flow of the dialysing medium and the resulting dilution of the beer. This process is often called diafiltration[2,8]

While dialysis is a process principally governed by diffusion, in diafiltration both diffusive and convective mass transfer take place simultaneously as a result of two driving forces: a concentration gradient and transmembrane pressure gradient. Hence diafiltration has some parallels with ultrafiltration. The influence of convective mechanism on mass transport and the selectivity of the alcohol/extract separation is largely unknown. In this paper, overall and convective mass fluxes were calculated using the results of an experimental investigation of beer diafiltration using cellulose based (Cuprophane—regenerated cellulose) and polysulfone hollow fibre membranes. Selected membranes have different permeability. Cuprophane is the typical dialysis membrane with low hydraulic permeability, low TMPs, which can be applied, and diffusive transport mechanism as a dominant one. Polysulfone is the ultrafiltration membrane, which enables higher TMPs and higher convective portion of overall mass transport. The results give some insight into the mass transfer mechanism in/on the membrane for a complex system such as beer and enable a better understanding and optimisation of diafiltration.

EXPERIMENTAL METHODS

The experimental investigation was performed using laboratory scale for continual dialysis under pressure. The system was equipped with valves, flow and pressure indicators on the inlet and outlet sides of membrane modules and tanks. The main characteristics of the modules are given in Table I.

In all experiments a standard beer with 12% (w/w) of original extract was used. The beer was passed through the hollow fibre capillaries while water, used as the dialysing medium, was passed counter currently around the fibres. The operating conditions are given in Table II. Alcohol and extract contents in the outlet streams were determined by recommended analytical procedures[1].

TABLE I. Main technical characteristics of dialysers

Membrane material	Regenerated cellulose	Polysulfone
Membrane structure	Symmetric	Asymmetric
Membrane type	Hollow fibre	Hollow fibre
Total membrane surface area	1.3 m²	1.25 m²
Hollow fibre diameter	200 µm	200 µm
Membrane thickness	8 µm	40 µm
Molecular cutoff	500	5000
Hydraulic permeability	16.8 ml/(min bar m²)	133.2 ml/(min bar m²)

TABLE II. Operating conditions

	Regenerated cellulose	Polysulfone
Beer flow rate	100–650 ml/min	100–650 ml/min
Beer/water flow ratio	1 : 1	1 : 1
Transmembrane pressure difference	0–0.4 bar	0–0.7 bar
Absolute pressure	1 bar	1 bar
Temperature	5°C	5°C

Main Definitions and Equations

Beer is a complex combination of materials, both in true solution and in the colloidal state. The aim in beer dealcoholisation is to reduce alcohol content with minimal reduction of the extract content in the beer, in order to preserve taste and sensory characteristics. In analysis used in this paper beer was considered to be a quasi three-component system consisting of water (solvent) and two solutes: alcohol and extract. Mass transfer parameter in dialysis are usually expressed by clearance, rather than by solute mass flux, because it eliminates the problem of variable inlet concentrations[4,5]. Clearance depends on the membrane surface area and therefore in order to obtain data comparable for two membrane modules with slightly different membrane surface areas and also slightly different feed concentrations, a new quantity termed normalised flux was introduced[6]. Normalised flux is defined as the ratio of the solute mass flux transferred through the membrane to the solute concentration in the feed stream. It is obvious that normalised mass flux is proportional to clearance and can be obtained by dividing the clearance values by the value of the total membrane surface area of the module.

Normalised mass flux of solute N_s in the process of diafiltration is obtained from the equation:

$$N_s = Q_1^o \frac{c_{s_1}^i - c_{s_1}^o}{A_m c_{s_1}^i} + J_{uf} - Q_2^o \frac{c_{s2}^o - c_{s_2}^i}{A_m c_{s_1}^i} + J_{uf} \frac{c_{s2}^i}{c_{s_1}^i} \qquad (1)$$

Q represents volumetric flow rate, A_m the membrane area and c_s solute concentration (in our case s is alcohol A, or extract E). Subscript 1 relates to the feed solution and subscript 2 to the dialysate, while superscripts i and o correspond to the inlet and the outlet streams, respectively. J_{uf} is the ultrafiltration volumetric flux and is proportional to the transmembrane pressure difference TMP:

$$J_{uf} = L_p TMP \qquad (2)$$

where L_p is the membrane hydraulic permeability. Due to functional losses the transmembrane pressure difference varies along the module and therefore an average transmembrane pressure difference TMP_{avg} was used. This was calculated assuming a linear relationship:

$$TMP_{avg} = TMP_n - \tfrac{1}{2}\Delta P_b + \Delta P_w \qquad (3)$$

TMP_n is the nominal transmembrane pressure difference, i.e. the difference between the beer and water inlet pressures, and ΔP_b and ΔP_w are the beer and water side pressure drops, respectively.

The convective component of the normalised mass flux was determined using the following relationship[8]:

$$N_s^{C/DF} = R_{xs} J_{uf} S_s \qquad (4)$$

where: R_{xs} is the average concentration relation for component s, which can be calculated in the following way:

$$R_{xs} = \frac{Q_1^i}{Q_{uf} B}\left[1 - (1 - Q_{uf}/Q_1^i)^B\right], \; B = 1 + \frac{\ln(c_{s_1}^o / c_{s_1}^i)}{\ln(1 - Q_{uf}/Q_1^i)} \qquad (5)$$

and S_s is the sieving coefficient, which was calculated as the ratio of the solute mean concentrations on the dialysate and beer side:

$$S_s = \frac{c_{s_2}^o + c_{s_2}^i}{c_{s_1}^o + c_{s_1}^i} \qquad (6)$$

In our experiments, the dialysate was pure water ($c_{s_2}^i = 0$) and the outlet beer and dialysate flow rates were equal.

The extraction ratio represents the fraction of maximum solute concentration change that can be attained under a given set of operating conditions. It can be evaluated using the following equation:

$$E_s = \frac{Q_1^i c_{s_1}^i - (Q_1^i - Q_{uf}) c_{s2}^o}{Q_1^i c_{s_1}^i} = \frac{(Q_1^0 + Q_{uf}) c_{s_1}^i - Q_1^0 c_{s_1}^0}{(Q_1^0 + Q_{uf}) c_{s_1}^i} \qquad (7)$$

which for negligible ultrafiltration, reduces to:

$$E_s = \frac{c_{s_1}^i - c_{s_1}^0}{c_{s_1}^i} \qquad (8)$$

This parameter can be used as a measure of dialyser effectiveness. The separation process is more advantageous if the extraction ratio of the separated component is higher.

The overall separation factor can be defined as the ratio of the extraction ratio of two solutes (alcohol and extract in the present case):

$$\lambda_{AE}^0 = \frac{E_A}{E_E} \qquad (9)$$

and can be used to quantify the selectivity of the separation process. High rate of selectivity requires high value of the separation factor, which means that loss of valuable component is minimal.

Results and Discussion

The alcohol and extract normalised fluxes, calculated from the experimental results using the previous equations are presented in Figures 1 to 4.

Figure 1 shows that with low molecular weight cutoff, in the case of cellulose based dialysing membranes, the convective fluxes of both alcohol and extract were negligible compared to the overall flux. Furthermore the overall alcohol flux was considerably higher than the corresponding extract flux. The overall alcohol flux increased with increasing flow rate and with increasing TMP, while the extract flux was independent of both (Fig. 2). In the case of diafiltration using the polysulfone

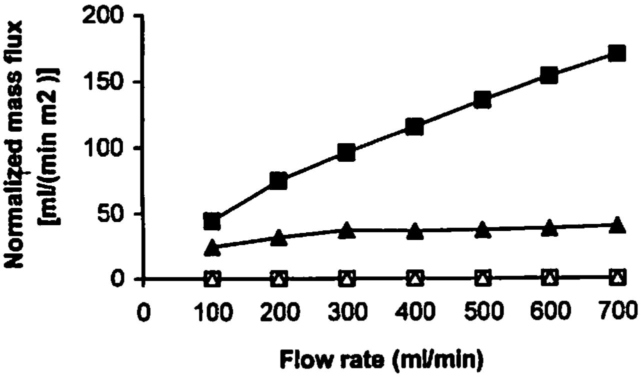

FIG. 1. Overall and convective alcohol and extract normalised mass fluxes obtained for cellulose membrane by different flow rates (Q^0) and 0.3 bar transmembrane pressure difference (TMP). Overall alcohol (-■-) and extract mass flux (-▲-); convective alcohol (-□-) and extract mass flux (-△-).

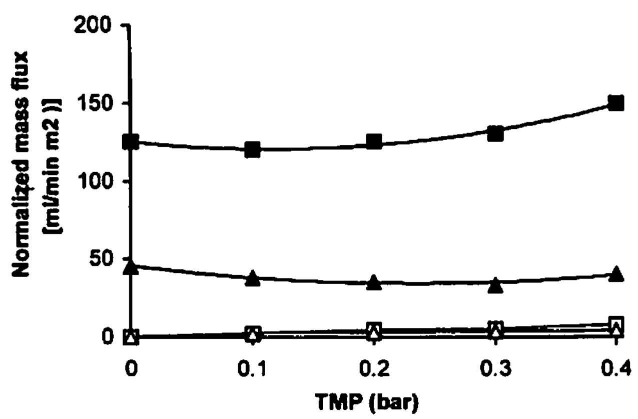

FIG. 2. Overall and convective alcohol and extract normalised mass flux obtained for cellulose membrane by different transmembrane pressure difference (TMP) and 200 ml/min flow rate (Q^0). Overall alcohol (-■-) and extract mass flux (-▲-); convective alcohol (-□-) and extract mass flux (-△-).

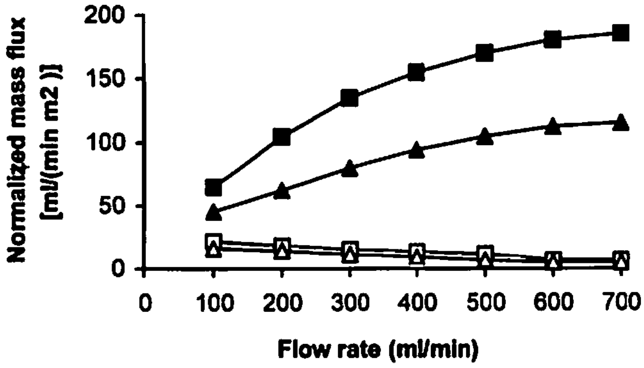

FIG. 3. Overall and convective alcohol and extract normalised mass fluxes obtained for polysulfone membrane by different flow rates (Q^0) and 0.3 bar and transmembrane pressure difference (TMP). Overall alcohol (-■-) and extract mass flux (-▲-); convective alcohol (-□-) and extract mass flux (-△-).

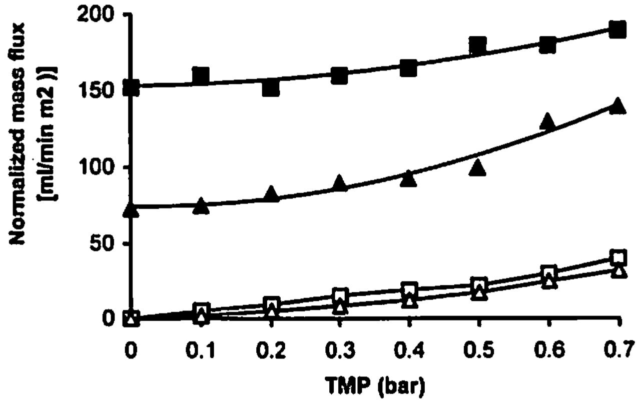

FIG. 4. Overall and convective alcohol and extract normalised mass flux obtained for polysulfone membrane by different transmembrane pressure difference (TMP) and 200 ml/min flow rate (Q^0). Overall alcohol (-■-) and extract mass flux (-▲-); convective alcohol (-□-) and extract mass flux (-△-).

membrane, the convective alcohol and extract fluxes cannot be neglected and differed only slightly (Fig. 3). Both decreased with increase of flow rates (owing to pressure drops which diminished the TMP) and increased with increase of TMP. In contrast to diafiltration using the cellulose membrane the overall fluxes of both alcohol and extract increased with increasing flow rates and TMP (Fig. 4).

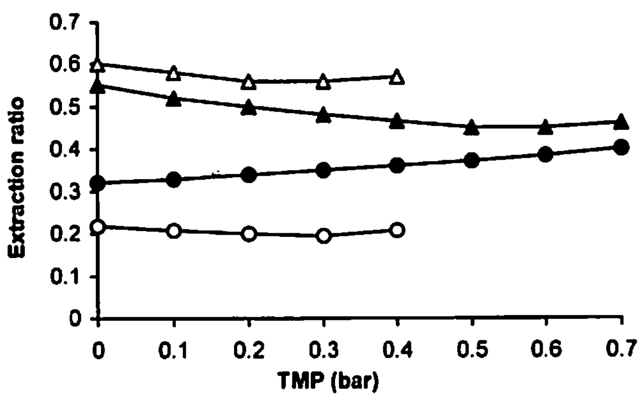

FIG. 5. The relation between the alcohol and extract extraction ratios and the transmembrane pressure difference (TMP) for cellulose and polysulfone membranes obtained by 200 ml/min flow rate (Q^0). Alcohol (-△-) and extract (-○-) extraction ratio by cellulose membrane; alcohol (-▲-) and extract (-●-) extraction ratio by polysulfone membrane.

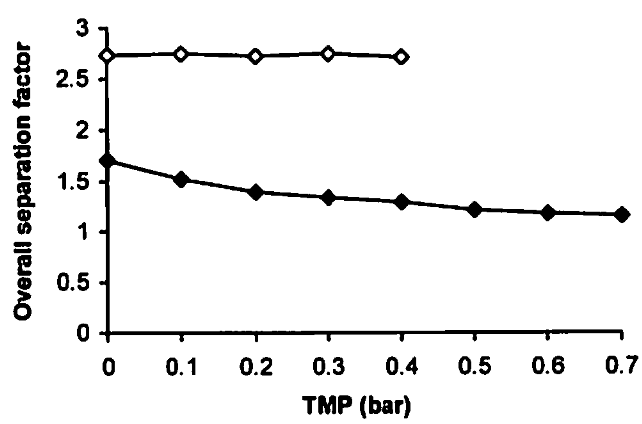

FIG. 6. Overall separation factors for cellulose (-◇-) and polysulfone (-◆-) membranes obtained by different transmembrane pressure difference (TMP) and 200 ml/min flow rate (Q^0).

Values of extraction ratios and overall separation factors obtained for different experimental conditions are given in Figures 5 and 6. The influence of TMP on the selectivity of the process depended on the membrane porosity and molecular mass of the separated component. In the case of dialysing membranes with small molecular cut-offs and negligible convective mass transfer, the TMP had no significant influence on the alcohol/extract separation. However in the case of higher values obtained with more porous membranes with greater influence of the convective transport mechanism, TMP generally diminished the effect of the separation. This is because the transport mechanism of larger molecules and conditions existing in/on the membrane surface depended much more on convection, than on the small relatively mobile alcohol molecule.

Conclusions

The portion of convective mass transfer in the overall flux is affected by membrane properties, the transported molecule and the TMP, but generally speaking convective mass transport diminishes the selectivity of alcohol/extract separation in beer diafiltration. Increasing the TMP can slightly support alcohol removal by introducing pressure difference as a new driving force, but this effect is dominated by extract molecules, which have much higher molecular mass and volumes. In other words, the TMP increase, aimed to achieve higher efficiency of the plant, is not justified.

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