

# Heat and mass transfer in osmotic distillation with brines, glycerol and glycerol–salt mixtures

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## Abstract

Osmotic distillation (OD) is a mass transfer operation in which the water activity difference is the driving force for mass transport through the membrane, however, heat transfer is also concerned in that the mass transport implies evaporation at the feed side and condensation at the extract side, thus, creating a temperature difference across the membrane which acts a counter driving force for mass transfer (thermal effect).

This study presents OD experiments with various extractants in a plate and frame module expressly designed to investigate separately the simultaneous effects of the heat and mass transfer.

In co-current operation, the temperature difference between the two streams approaches an asymptotic value for large residence time, i.e., for long modules or low flow rates.

In asymptotic conditions, which correspond to no heat flux through the system, the problem looks like a pseudo isothermal case, this allows to obtain the relevant membrane transport properties and to evaluate the role played by the concentration polarisation.

The temperature difference created by the thermal effect, even of few degrees, greatly reduces the driving force for mass transfer through the membrane, the resulting loss of flux is relevant and increases with the temperature. The concentration polarisation plays a comparable role in the OD with calcium chloride or glycerol at room temperature, while is less important in OD with glycerol–NaCl mixtures, and negligible in the case of sodium chloride.

Among the extractant used,  $\text{CaCl}_2$  is the most effective, however, the advantage over the Glycerol–NaCl mixture is not so high as would appear comparing the respective theoretical driving forces for mass transfer.

The length of module needed to approach asymptotic conditions (thermal entry length) can be estimated to be lower than the typical length of industrial apparatuses. The flux achievable in OD applications is, thus, close to the asymptotic value, larger fluxes can only be obtained in lab devices.

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## 1. Introduction

Osmotic distillation (OD) is a well known membrane technique to concentrate at room temperature aqueous mixtures such as fruit or vegetable juices. Water is removed from the feed by a hypertonic solution flowing downstream

a porous hydrophobic membrane typically made of polytetrafluoroethylene (PTFE) or polypropylene [1].

Due to the hydrophobicity of the membrane material, the two liquids facing the membrane do not enter the pores, where a thin gas layer is entrapped. This gas layer (gas membrane) is the true selective barrier. The driving force for mass transfer is represented by the water vapour pressure difference, related to the water activities in the two liquids phases.

Highly concentrated Calcium Chloride solutions were widely used as extractant [2,3], indeed, this salt is quite effec-

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tive, owing to the large solubility. However, the use of nearly saturated  $\text{CaCl}_2$  solutions on industrial scale poses serious problems: the solutions are highly corrosive, the solubility strongly depends on the temperature, so that an accidental temperature drop causes crystallisation with obstruction in the pipes and fittings. Sodium chloride was used in research works [4,5], however, the use of NaCl is not convenient in industrial scale, since the driving force available is limited because of the low solubility.

In order to overcome the problems associated with the use of brines, alternative extractants were proposed [6], among these glycerol showed good performances: it is completely miscible with water and fulfils the no toxicity requirement, the volatility is negligible and the surface tension is quite close to that of water. The last properties are crucial, indeed, negligible volatility means negligible counter diffusion of glycerol towards the juice and easy regeneration, whereas the penetration pressure through the membrane pores is directly related to the surface tension. The viscosity of glycerol solutions is actually quite large; by using ternary mixtures of glycerol–water–NaCl, or other salts it is possible to obtain the same flux achievable with glycerol alone, but with substantially lower viscosity [6]. Also propylene glycol was considered as a possible extractant, it is nearly equivalent to glycerol as regard as the driving force, however, the volatility is not negligible and the penetration pressure is quite small [6].

The water activity of the relevant solutions can be calculated with good accuracy by predictive methods, in [6] the Wilson method was used for glycerol solutions and a modified ASOG method [7] for electrolyte solutions. For the ternary mixtures glycerol–water–NaCl the product rule showed to be adequate:

$$\gamma_w = \gamma_w^{\text{el}} \gamma_w^{\text{Gly}} \quad (1)$$

in which  $\gamma_w^{\text{el}}$  is the water activity coefficient, calculated by the ASOG method, of the water–electrolyte solution on glycerol free basis, and  $\gamma_w^{\text{Gly}}$  the water activity coefficient, calculated by the Wilson method, of the water–glycerol solution on electrolyte free basis.

OD is not a simple mass transfer operation, in that heat transfer is also concerned even in the case of equal bulk temperatures. Indeed, we have water evaporation at the feed side, diffusion through the pores and condensation at the extractant side. As a consequence of the phase changes at the membrane walls the feed cools down and the extractant warms up. A temperature difference between the membrane sides is, thus, created, reducing the driving force for mass transfer (thermal effect).

The thermal effect was studied in [5] for capillary modules in co-current operation; that work showed that, whatever the inlet temperatures may be, equal or not, the temperature difference between the two streams approaches an asymptotic value as the module length increases.

The phenomenon is more difficult to analyse by experiments in plate and frame modules, indeed, heat transfer through the spacers, supports and module walls superimposes to the heat transfer through the membrane.

Preliminary results for a plate and frame module were reported in [8]: by using NaCl solutions near the saturation, the asymptotic temperature differences between the two streams were identified for different inlet temperatures. The data allowed to characterise the membrane in terms of permeability  $K_m$  and heat transfer coefficient  $h_m$ .

In this paper, we report the results obtained with a plate and frame module using different extractants (NaCl,  $\text{CaCl}_2$ , Glycerol, Glycerol–NaCl mixtures) and discuss the role of heat and mass transfer in the various cases.

## 2. Theory

The mass transport phenomena involved in OD are well understood [5,9,10] and are represented by the mass transfer through the stagnant air entrapped within the membrane pores and the mass transfer through the liquid boundary layers.

Assuming that molecular diffusion is the prevailing mechanism of mass transfer through the membrane pores, the mass flux ( $N$ ) can be expressed as a function of the water vapour pressures at the membrane interfaces,  $P_{w1}^I$ ,  $P_{w2}^I$  as:

$$N = K_m \frac{P_{w1}^I - P_{w2}^I}{P_{A_{lm}}} \quad (2)$$

where  $P_{A_{lm}}$  is the logarithm mean of the partial pressure of the air within the pores and  $K_m$  the membrane permeability.

Theoretical values of the membrane permeability can be calculated from the membrane properties:

$$K_m = \frac{\varepsilon M_w}{\chi \delta} \frac{PD}{RT} \quad (3)$$

in which  $\varepsilon$  and  $\delta$  are the membrane void fraction and the thickness, respectively and  $\chi$  the tortuosity factor.

For the diffusion in gas phase the dependence of the diffusion coefficient,  $D$  on temperature and pressure is of the type  $PD/T^b = \text{constant}$  [11]. In the case of water vapour in air, the exponent  $b$  can be assumed equal to 2, in the small temperature range here considered [5], as a consequence the membrane permeability linearly increases with temperature, in other words the ratio  $K_m/T$  is a constant membrane property.

The description above does not imply that molecular diffusion is the only mechanism of mass transfer through the membrane, it is well recognized that Knudsen resistance gives a contribution depending on the pore size. Alves and Coelho [12] recognized that the Knudsen resistance represents 58% of the overall resistance for 0.1  $\mu\text{m}$  pore size, 39% for 0.22  $\mu\text{m}$  pore size and only 24% for 0.45  $\mu\text{m}$  pore size.

In any case, Eq. (2) can be used to describe the phenomenon, considering the membrane permeability  $K_m$  as an

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