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Concentration of dairy flavour compounds using pervaporation

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Abstract

A dilute aqueous model solution containing acids, esters and ketones (which contribute to dairy flavours) was concentrated by passing through pervaporation membranes. Polydimethylsiloxane membranes gave greater fluxes than polyoctylmethylsiloxane membranes. Fluxes increased with feed temperature and decreased with permeate pressure. Different compounds were concentrated by different amounts, meaning that the permeate composition differed from that of the feed. Esters and ketones passed through the membrane more readily than acids. Low molecular weight esters and ketones were enriched by a greater amount than larger molecules, but the influence of molecular weight was more complex for acids, and depended on the relative importance of sorption and diffusion mechanisms in the membrane.

Keywords: Pervaporation; Acids; Esters; Ketones; Flavour concentration

1. Introduction

Flavour compounds can be lost or adversely affected during many food processing operations (Pyle, 1994). It is sometimes necessary to recover the natural flavour compounds, which can then be added back to the food product. Also, natural flavour compounds can be concentrated to be used as flavouring ingredients (Sibeijn, van der Horst, de Jong, & Smit, 2004). As flavour compounds are typically found at low levels in foods, it is important to be able to concentrate the flavours without concentrating the rest of the food matrix. Some methods currently used to recover or concentrate flavours include techniques based on distillation, partial condensation and gas stripping (Karlsson & Trägårdh, 1997).

Recently, attention has turned to pervaporation as an alternative concentration method. Pervaporation is a membrane separation process that can be used to concentrate certain compounds in a mixed feed. In hydrophobic pervaporation, volatile hydrophobic compounds such as flavours pass through the polymeric membrane more readily than water, and are thereby concentrated in the permeate (Lipnizki, Hausmanns, Ten, Field, & Laufenberg, 1999). The permeation rate of each compound through pervaporation membranes depends on its solubility and diffusivity in the membrane; hence, the enrichment of compounds is governed by their chemical nature as well as by their size. Pervaporation is a selective process; some feed components are concentrated to a greater degree than others, with the selectivity controlled by the membrane type (Karlsson & Trägårdh, 1993).

Compounds permeate through the membrane because their chemical potentials are lower on the permeate side of the membrane than in the feed. This chemical potential difference across the membrane can be expressed as either an activity difference (Lipnizki, Hausmanns, & Field, 2004) or a partial pressure difference (Trifunović & Trägårdh, 2006). This is usually accomplished by keeping the permeate side under vacuum (Feng & Huang, 1997; Lipnizki et al., 1999).

Pervaporation through hydrophobic membranes has been studied by many researchers as a means of concentrating organic compounds from an aqueous feed. Several of these studies focused on flavour compounds. In

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most cases, the feed solutions studied were simple model solutions containing up to only three components other than water, and the results varied widely depending on the membranes used and the compounds tested. For example, Baudot and Marin (1996) used pervaporation to concentrate methylthiobutanoate by a factor of up to 1205 and diacetyl by a factor of 15–20. Rajagopalan, Cheryan, and Matsuura (1994) also studied pervaporation of diacetyl from an aqueous solution, and achieved enrichment factors of around 35. Pervaporation of esters has been studied by several researchers and typically gives high selectivities; Sampranpiboon, Jiraratananon, Uttapap, Feng, and Huang (2000) achieved separation factors (a measure of selectivity) between 80 and 300 for ethyl butanoate and ethyl hexanoate, and Song and Lee (2005) found that ethyl acetate, propyl acetate and butyl acetate were enriched by factors of 60–120. Baudot, Souchon, and Marin (1999) investigated the pervaporation behaviour of four flavour compounds covering a range of physico-chemical properties, and found that selectivity depended on the permeate pressure for high boiling compounds but not for low boiling compounds.

In other cases, feed solutions were real flavour systems, such as fruit essences (Rajagopalan & Cheryan, 1995; She & Hwang, 2006a; Zhang & Matsuura, 1991), cauliflower blanching water (Souchon, Pierre, Athes-Dutour, & Marin, 2002), wine (Karlsson, Loureiro, & Trägårdh, 1995), tea (Kanani, Nikhade, Balakrishnan, Singh, & Pangarkar, 2003; She & Hwang, 2006a) and a marine alga (Beauchêne, Grua-Priol, Lamer, Demaimay, & Quémeneur, 2000). Most flavour compounds in the feed were concentrated by pervaporation, whereas feed components with low volatilities did not pass through the membrane. However, it is difficult to predict pervaporation performance with complex feed solutions, because interactions between feed components are possible (Karlsson & Trägårdh, 1993; Kedem, 1989). Therefore, results with real feed mixtures did not always match those with model feeds (Kanani et al., 2003; Souchon et al., 2002).

The objective of this study was to compare pervaporation of nine flavour compounds in a model feed solution, under a range of operating conditions (membrane type, feed temperature and permeate pressure). Compounds from three functional groups (organic acids, esters and ketones) were selected, enabling comparisons between different types of permeating molecules. Compounds from these three functional groups contribute to the flavour of cheese and other dairy products (Keen, 1998; Urbach, 1997).

2. Materials and methods

2.1. Model feed solution

The multicomponent feed solution used for all experiments consisted of approximately 100 mg L^{-1} each of acetic acid (Sigma-Aldrich), butanoic acid (Aldrich), hexanoic acid (Fluka), octanoic acid (Sigma), ethyl butanoate (Fluka) and ethyl hexanoate (Aldrich), and approximately 10 mg L^{-1} each of ethyl octanoate (Aldrich), 2-heptanone (Fluka) and 2-nonanone (Fluka); all were dissolved in distiled water. Sigma-Aldrich, Aldrich, Sigma and Fluka brand chemicals were all supplied by Sigma-Aldrich Co. (St Louis, MO, USA). All chemicals had a purity of greater than 98%. A fresh 5-L batch of model feed solution was used for each pervaporation experiment.

2.2. Pervaporation apparatus

The pervaporation set-up is shown in Fig. 1. The feed was recirculated past the membrane and back into the feed vessel at approximately 1 L min^{-1} (corresponding to a Reynolds number of about 500), using a diaphragm pump (Cole-Parmer Instrument Company, Vernon Hills, IL,

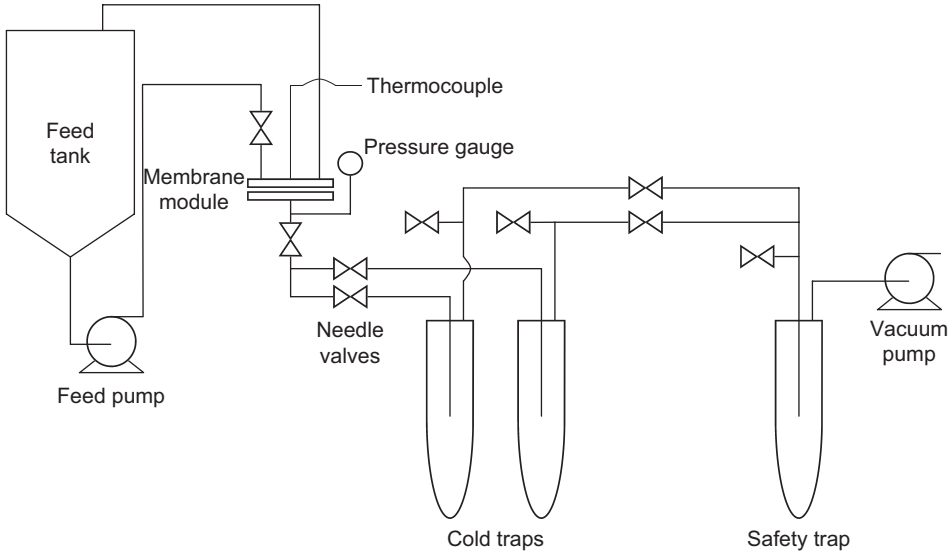

Fig. 1. Schematic diagram of pervaporation unit.