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Differential analysis of deposition layers from micellar casein and milk fat globule suspensions onto ultrafiltration and microfiltration membranes

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Abstract

Particle deposition behaviour on membrane filters is of the utmost importance for the flux and selectivity. For milk filtration not only the particle behaviour as such but also the behaviour in time is a factor to take into account. Therefore we applied a differential analysis to the flux decline during dead-end filtration of latex, casein micelles and milk fat globules.

Based on our analysis for latex we distinguished two regions, one best described by pore blocking and one by cake filtration. Besides these two regions, no other time dependent behaviour was observed, as was expected for latex. However, for casein micelles and milk fat globules various other time dependent effects were observed and quantified.

Pore blocking of an UF membrane by micellar casein was relatively small, as concluded from the values of the pore blocking parameter of 0.1–0.2. In contrast, the cake resistance was relatively high, 35 times higher than expected from the Carman–Kozeny theory. This may be attributed to a decrease of the size of the casein micelles, leading to the formation of very compact layers. The specific cake resistance of micellar casein was lower for a MF membrane where depth filtration played a role.

Milk fat globules generate layers with a very high cake resistance, both for liquid and for partly crystallised fat. For liquid fat, spreading of milk fat globules onto the membrane and deformation of milk fat globules seem to be the main cause for the high cake resistance. When a large part of the milk fat was crystallised ($T = 5$ °C), partial coalescence of milk fat globules in the cake layer seems to play a role.

Differential cake analysis proved to be a valuable method for the evaluation of concentrated particle layers, resulting in better knowledge about the specific (time dependent) particle behaviour in these layers. In this way, parameters are identified that are essential for the design of microfiltration or ultrafiltration processes.

Keywords: Membrane filtration; Milk; Colloidal particles; Pore blocking; Cake filtration; Modelling

1. Introduction

Microfiltration is a widely applied technique in the food and bioprocess industry for the separation and fractionation of particles in suspensions, such as present in milk. Milk contains about 15% v/v colloidal particles, mainly casein micelles (proteins) and (emulsified) fat globules. The development of the uniform transmembrane pressure (UTP) concept in the 1970s and 1980s of the last century

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(Sandblom, 1974) enabled successful microfiltration of this concentrated suspension on an industrial scale. This opened the way for the application of microfiltration for e.g. the reduction of bacteria and spores (Maubois, 1991), fractionation of fat globules (Goudédranche, Maubois, & Fauquant, 1998) and the separation of casein micelles and serum proteins in milk (Maubois, 1991).

The performance of microfiltration processes is governed by particle deposition onto the membrane. Therefore, prediction of flux (decline) and separation performance requires a good understanding of this particle deposition process. Not only the deposition rate, but also the properties of the deposited layer play an important role.

Studies on the properties of deposition layers of (other) proteins (Guell & Davis, 1996; Hlavacek & Bouchet, 1993; Ho & Zydney, 1999, 2000; Tracey & Davis, 1994) and emulsion droplets (Arnot, Field, & Koltuniewicz, 2000) are mostly carried out by modelling of dead-end filtration processes with a constant pressure drop. Two different modelling approaches can be distinguished: (1) based on the filtration equations of Hermia (1982), and (2) based on the sieve mechanism of Flippov, Starov, Lloyd, Chakravarti, and Glaser (1994). Both approaches recognise two different mechanisms: pore blocking and cake filtration. The first mechanism is usually relevant in the beginning of the deposition process, and the second one after the deposition process has progressed for some time. Both approaches as well as both mechanisms can be expressed in one characteristic filtration law, describing how the change in resistance $\left(\frac{d^2t}{dV^2}\right)$ is related to the resistance (inverse flux, $\frac{dt}{dV}$):

$$\frac{d^2t}{dV^2} = k \cdot \left(\frac{dt}{dV}\right)^n \quad (1)$$

The values of the constant k and the exponent n depend on the filtration mechanism. For pore blocking, n can vary between 1 and 2, and for cake filtration $n = 0$. The two approaches of Hermia and Flippov are only different in the formulation of pore blocking. Hermia distinguishes three blocking mechanisms, complete, standard and intermediate blocking, each with a fixed value for n of 2, 1.5 and 1, respectively. Flippov uses a single parameter to describe pore blocking for a given filter and suspension. This parameter has physical meaning: it is the ratio of the area of influence of a particle above a pore and the pore area itself. In other words, the value of the single parameter is a measure for the relative flux decline caused by the deposition of a single particle onto the membrane as related to the relative projection area of this particle on the membrane. The exponent n does not have a specific value in this approach.

The abovementioned models are valid for non-compacting and non-compressive particle layers. For soft and polydisperse colloids such as casein micelles and fat globules, these models may be lacking sufficient detail. Models that do take compaction and compression during a filtration run into account are also available (see e.g. Philip & Smiles, 1982; Smiles & Kirby, 1987; Smiles, Raats, & Knight, 1982), but these models require detailed data on the concentration distribution in the particle layers. Since in general, the layers formed during milk microfiltration are very thin, analysis of the concentration distribution is technically complex. This may be the reason that presently, no systematic study on the properties of milk deposition layers has been published.

In this paper, we present a new differential approach for the analysis of the properties of deposition layers, based on simple flux measurements in dead-end filtration experiments. In this method, one single flux curve is separated into a series of curves, such that a time-sequence of flux curves is obtained, each representing a different stage of the filtration process. The size of the time interval of each stage is taken such that the constant k and the exponent n can be considered constant. In this way, pore blocking and cake filtration can be evaluated separately in each stage of the process, but one can also discern the effects of compaction and compression of the particles on the flux decline.

We applied this approach to dead-end filtration of colloidal suspensions of casein micelles and milk fat globules. In this way we focus on the properties of these industrially relevant components, that, surprisingly enough, have not been studied systematically. With our differential flux analysis approach we were able to quantify the particle behaviour and found very large differences between the various particles under study.

2. Materials and methods

2.1. Preparation of colloidal suspensions

2.1.1. Latex particles

Three different suspensions were used; suspensions of latex particles, casein micelles and of milk fat globules. The latex particles (surfactant-free, sulfate polystyrene, density 1.06 g/ml, Interfacial Dynamics Corporation, USA) had mean diameters of 0.42 and 0.60 μm and were suspended in a mixture of demineralised water with 23.1% w/w glycerol, which has the same density as the latex particles. The viscosity of the water–glycerol mixture is 2 mPa s (25 °C).

2.1.2. Casein micelles

The suspension of casein micelles was prepared from pasteurised skimmilk. The skimmilk contained 3.6% w/w protein (Kjeldahl method), consisting of 2.7% w/w casein and 0.7% w/w serum protein. The skimmilk was microfiltered (Alfa Laval MFS-1 system, filtration temperature 50 °C) with a 0.2 μm ceramic MF membrane (SCT Membralox P19-40, tubular αAl_2O_3 membrane, tube diameter 4.0 mm, membrane area 0.20 m^2) up to a concentration factor ($=Q_{feed}/Q_{retentate}$) of 7 (viscosity measurements) or of 4 and subsequently diafiltered (100%) and diluted (100%) with protein-free milkserum (dead-end filtration