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Critical flux of gum arabic: Implications for fouling and fractionation performance of membranes

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ABSTRACT

A flux-stepping method was used to determine the critical flux of 2 wt% gum arabic using flat sheet polysulfone membranes. These were found to be 27 L m^{-2} h^{-1}, 10 L m^{-2} h^{-1} and 22 L m^{-2} h^{-1} for 0.1, 0.5 and 0.8 μm polysulfone membranes, respectively. Increasing the cross flow velocity was found to raise the critical flux, although the effect diminished at higher velocities. These values were then used to assess the fouling and the fractionation performance of the three membranes above and below their critical fluxes.

1. Introduction

The fractionation of gum arabic into its three component parts has long been considered a desirable aim industrially. Gum arabic processing is a 50,000 tonnes per year industry and the processed product is widely used in the food industry as an emulsifying, stabilising, thickening and glazing agent. It has been shown that *Acacia senegal* contains between 10 and 20 wt% arabinogalactan–protein complex (AGP; Average M_W = 1500 kDa), which is the functional component giving gum arabic its exceptional properties as an emulsifying agent (Nishino et al., 2012; Randall et al., 1988). The other two fractions in gum arabic are arabinogalactan (AG; very broad M_W range, average = 280 kDa; 75–90% total gum solids) and glycoprotein (GP; Average M_W = 250 kDa; about 2% total gum solids). The aim of a number of studies has been to modify gum arabic to improve its properties as an emulsifier (Al-Assaf et al., 2007; Fang et al., 2013; Heidebach and Sass, 2013; Katayama et al., 2012; Ward, 2002). Another reason for the industrial interest in gum arabic fractionation is its potential to reduce batch-to-batch variation by separating the AGP and then blending it with other gum constituents in order to regulate the AGP content.

Previous work in our group has reported the first use of synthetic, polymeric membranes to separate the AGP fraction of gum arabic from the other two fractions present (arabinogalactan (AG) and glycoprotein (GP)) in cross-flow diafiltrations (Manning and Bird, 2015). Here, commercial polysulfone (PS)

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membranes of 0.1, 0.5 and 0.8 μm pore size were tested for their fractionation performance. It was concluded that the smaller pore size membrane rejected the AGP to a greater extent, but that after some fouling of the membrane, the larger pore size membrane also showed some rejection. It was also observed that very little gum arabic passed through the 0.1 μm membrane, suggesting that the pore size was too small for the majority of the gum species to be transmitted. The fouling observed was, therefore, likely to be surface cake formation. Contrastingly, the larger pore sized membranes showed greater transmission of gum arabic initially, but this decreased over time. It seems likely that an initial pore blocking mechanism is subsequently followed by cake formation.

Field et al. (1995) first introduced the concept of critical flux. This phenomenon describes filtration upon start up that, below a threshold flux, demonstrates little or no membrane fouling. During filtrations at constant flux, this results in a steady trans-membrane pressure (TMP). If the flux is increased, however, above this critical value, fouling occurs which cannot be removed upon once again lowering the flux. When a membrane is fouled, the TMP increases under conditions of constant flux operation. This critical flux value can be improved by increasing the cross-flow velocity.

There exist two forms of this theory. The first is the 'strong form' of the critical flux, where a flux exists which is equal to the pure water flux under those conditions; the second is the 'weak form' where upon start up, a constant flux is quickly established, after some initial solute adsorption occurs, and continues with no rise in TMP.

Fouling can significantly alter the selectivity of the membrane; operating below critical flux has the advantage of easier control over the filtration conditions. This is interesting from the point of view of fractionation, as the absence of surface deposits may have an effect on the rejection of species. These deposits alter the filtration properties throughout the entire operation as the surface deposits will change in thickness and density, and in-pore blocking will also affect the pore size distribution, giving constantly changing results (Brans et al., 2004).

This paper reports the determination of the critical flux of 2 wt% gum arabic using 0.1, 0.5 and 0.8 μm PS membranes and observes the effect on filtration and fractionation performance operating above and below the critical flux. The effect of CFV upon critical flux is also reported.

2. Materials and methods

2.1. *Materials*

Gum arabic was supplied by *Kerry Ingredients and Flavours* (Cam, Gloucestershire, UK) as a milled, raw product from *A. Senegal* trees in Sudan. Feed solutions were prepared by dissolving the appropriate amount of gum arabic in Milli-Q water and passing this through a 50 μm wound stainless steel pre-filter to remove insoluble debris. A concentration of 2 wt% was chosen for this work to provide a low viscosity feed and to be consistent with previous work (Manning and Bird, 2015). Milli-Q water (Millipore) was used for feed preparation and filtrations.

Polysulfone (PS) membranes of 0.1, 0.5 and 0.8 μm pore size (MFG1, GRM-RT5 and GRM-RT8; *Alfa Laval*, Denmark) were cut to fit a crossflow filtration cell (Ying Kwang Trading, custom made). The cell has parallel plate geometry with an active membrane area of 0.0054 m^2 (0.18 m × 0.03 m) and a channel height of 1.5 mm. The set-up consisted of a 10 L feed tank fitted with an overhead stirrer (Panasonic, model MX8G5B) connected to a gear pump (Cole-Palmer, model 74013-45). Feed temperature was maintained at 40 °C by a heating/cooling system (Polyscience, model 9112) and permeate flux was controlled by a mass flow controller (Brooks Instrument, model 5882).

The membranes were conditioned before use to remove the glycerine coating by passing water at 60 °C over the membrane in the cross-flow cell utilizing the protocol developed by Weis et al. (2005).

2.2. *Experimental procedure*

The flux stepping method (Chen et al., 1997) was used to determine the critical flux of 2 wt% gum arabic using 0.1, 0.5 and 0.8 μm PS membranes. New, conditioned membranes were used for each experiment. First, the TMP readings were recorded every 20 s for Milli-Q water over a range of fluxes. The operating conditions for this membrane pure water permeability were the same as for the fouling experiment of 2 wt% gum arabic. Standard filtration conditions were set at 40 °C, a cross flow velocity (CFV) of 0.37 $m\,s^{-1}$, and 15 min flux steps were carried out from 2 $L\,m^{-2}\,h^{-1}$ (LMH) until a flux where TMP increase over the step period was significant (>0.02 bar min^{-1}), which indicates serious membrane fouling. For the 0.8 μm membranes, the flux stepping started at 10 LMH. Permeate and retentate were cycled back to the feed tank to maintain constant feed volume and concentration.

The effect of altering CFV was investigated by carrying out flux stepping experiments using 0.1 μm PS membranes at varying crossflow velocities (0.18, 0.37, 0.56 and 0.67 $m\,s^{-1}$). A total of 3 repeats of the critical flux measurement of 2 wt% gum arabic with 0.1 μm PS membranes at 0.37 $m\,s^{-1}$ CFV were carried out to test the reproducibility of the experiments.

After determination of the critical flux of 2 wt% gum arabic with the 3 membrane pore sizes under standard conditions, longer diafiltration experiments were carried out both above and below the critical flux in order to study the effect of fouling on the filtration and fractionation performance of the membrane.

2.3. *Data analysis*

The critical flux of 2 wt% gum arabic at 40 °C and 0.37 $m\,s^{-1}$ CFV was determined using the flux stepping method. There are several approaches for estimating the critical flux in this flux stepping method and two of these methods were employed here. Fig. 1, modified from Le Clech et al. (2003), explains the two different parameters based on TMP used to identify the point at which membrane fouling occurred. The first parameter is the change in TMP over the step period (15 min). This can be observed visually from the graphs of TMP vs. time. The critical flux is defined as the flux above which dTMP/dt ≠ 0. This is also calculated using equation 1.

$$\frac{dTMP}{dt} = \left(\frac{TMP_{final} - TMP_{initial}}{t_{final} - t_{initial}}\right) \quad (1)$$

$TMP_{initial}$ is the average TMP over the first minute of the flux step, excluding the first data point and TMP_{final} is the average TMP over the last minute of the flux step. Time is represented by t.

A second parameter, the average TMP in each step (TMP_{av}) can be plotted against flux and the point at which the two are