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Protein fouling of cellulose acetate microfiltration membranes modified by the deposition of amino acid L-DOPA

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Introduction:

Flux decline of the membranes due to fouling is a major drawback in the wide-scale application of the membrane separation process in the aqueous environments. A typical case is the protein fouling of microfiltration membranes when used in the applications such as food industries or water treatment. Filtration-induced protein deposition and protein adsorption are two distinct mechanisms which simultaneously affect the membrane flux decline. A number of works have tried to modify the surface of the microfiltration membranes in order to reduce the membrane fouling. Some of them demonstrated a lower flux decline for the modified membranes compared to the unmodified ones. However, some other works found the membrane fouling to be independent of the membrane surface chemistry while it occurs primarily due to the physical deposition of protein aggregates.

This study aims to investigate the role of membrane surface modification in the protein fouling of microfiltration membranes. It reports on using zwitterionic amino acid L-DOPA to modify the microfiltration cellulose acetate membranes. It was motivated by our previous study which showed that adhesion of poly L-DOPA to the surface of polyamide reverse osmosis membrane presents a zwitterionic interface with low attraction to proteins.

Methods:

Cellulose acetate microfiltration membranes (pore size: 0.2 µm from Toyo Roshi Co) were modified by soaking them in a 2 g/L L-DOPA solution. DOPA solution was prepared by dissolving L-DOPA in the Tris-HCl (10mM, pH 7.9) buffer solution. The membranes were characterised by water flux measurement, static protein adsorption and the filtration experiments. The water flux results were obtained by testing the membranes in 20 kPa transmembrane pressure in a dead end filtration system and the data were recorded after the membranes had been compacted for 1 hour. The static protein adsorption was expressed in terms of flux loss; The test was carried out by measuring the pure water flux of the membranes before and after being submerged for 3 hours in the BSA solution (model protein foulant). As the Final characterization, a series of dead end filtration experiments were performed using BSA in the feed to evaluate the effect of modification on the fouling behaviour of the membranes.

Results:

Table 1 shows the water flux of the original cellulose acetate membrane and two coated membranes. The results demonstrated an increase in water flux for the sample coated for 16 hours. It is likely that the coated L-DOPA layer is very thin relative to the membrane pore diameter and it resulted in the increase in membrane wettability without blocking the pores, so that increase the membrane permeability. However, for the 24- hour coated sample, a slight decrease in water flux was observed which can be attributed to the formation of the thicker poly

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L-DOPA layer on the membrane surface or into the pores causing pore blockage or wall thickening.

Table 1

The water flux results for the original and coated membrane samples measured in 20 kPa

Membrane sample	Virgin membrane	16 hours coated membrane	24 hours coated membrane
Water flux ($g/min.cm^2$)	1.54	1.65	1.43

Fig. 1 shows The percentage of flux loss for the pre-adsorbed membranes. The results demonstrate a higher amounts of flux loss for untreated membranes than that of the modified membranes which means higher amounts of protein had been adsorbed to the untreated membranes. Membranes which had been pre-adsorbed with 0.04 g/L BSA solution showed very small percentage of flux loss in comparison with the ones pre-adsorbed with 0.2 g/L BSA solution.

Figs. 2 and 3 display the plots of flux versus time curves for the tested membranes during the filtration of BSA solutions as the feed. During the filtration of 0.2 mg/ml BSA solution, an immediate fouling for the uncoatd membrane occured and the flux declined to nearly zero after 100

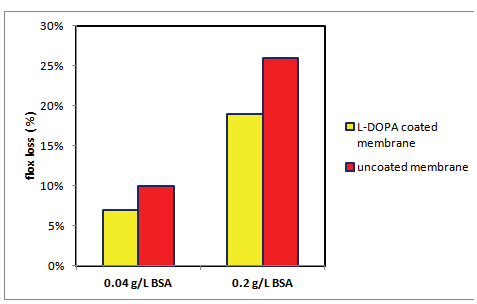

Fig. 1: The percentage of flux loss of the preadsorbed membranes

minutes of filtration. The flux of the coated membrane showed a slight decline with time and it reached about half of the inintial flux after 100 minutes filtration. During the filtration of 0.04 mg/ml BSA solution, both coated and uncoated membranes experienced lower flux decline rates than the samples tested with 0.2 mg/ml BSA solution. From Fig 3 it is also observed that even though the flux of the coated membrane, declined slightly less than that of the original membrane, no obvious difference between the flux decline profile of the coated and uncoated membranes happened.

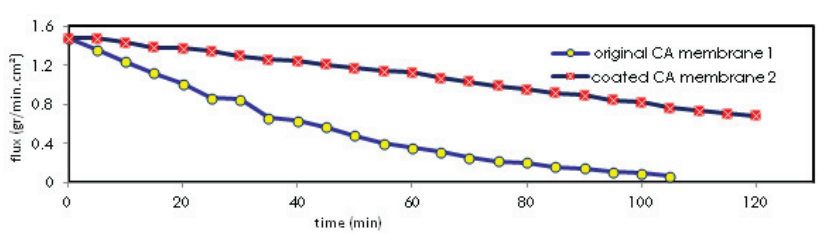

Fig. 2: The flux decline profile for the coated and uncoated membranes during the filtration of 0.2 mg/ml BSA solution.

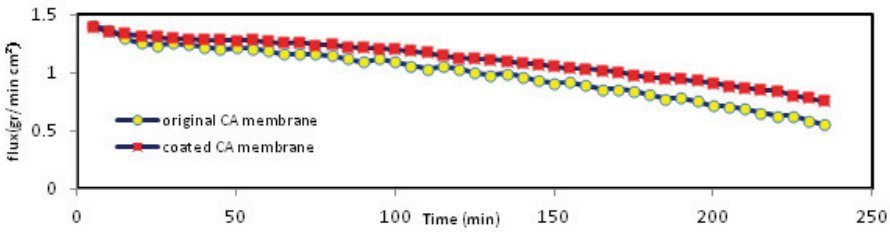

Fig. 3: The flux decline profile for the coated and uncoated membranes during the filtration of 0.04 mg/ml BSA solution.

Discussion:

Filtration experiments exhibited an improvement in the antifouling property of the coated membranes. However, it was more significant when the membranes were tested with a more concentrated BSA solution (0.2 mg/ml). During the filtration of 0.04 mg/ml only a slight diffrence in the decline rate of the coated membrane was observed. The filtration results of less concentrated BSA solution (0.04 mg/ml) seem to indicate a great dependence of the membranes fouling propensity on the membrane structure rather than on the chemical interactions. In fact in actual microfiltration, adsorption and deposition of proteins occur simultaneously and it is difficult to distinguish the amount of protein adsorbed or deposited. It is assumed that in the low concentrations of BSA, physical deposition is the dominate fouling mechanism and the protein adsorption driven by the concentration diffusion is of less importance. The results of the pre-adsorption experiments also confirm this observation because both coated and uncoated membranes showed a very small amounts of flux loss after 3 hours immersing in 0.04 mg/ml BSA solution.

In summary, the L-DOPA coating improved the antifouling properties of CA microfiltration membranes. However its improvment effectivness can be dictated by operating parameters.

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