Pressure relaxation flux recovery in filtration of Alginate solutions by Ultrafiltration and Microfiltration membranes

M.Sc. Thesis in Soil and Water Sciences

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Abstract

Membrane fouling is one of the major obstacles in applying membrane filtration to wastewater treatment and reuse in efficient and economical manner. Membrane Pressure relaxation flux recovery is an interesting phenomenon which may be used to improve Ultrafiltration and Microfiltration systems performance. Yet, the power balance between different mechanisms, i.e. diffusion and de-compaction, which is assumed to controls the process and affect it is far from being understood. Fouling and flux characterization is a precondition for pressure relaxation experiments. In previous studies (Ye et al., 2005, Katsoufidou et al., 2007) Alginate was used a model Polysacharide and Alginate solutions filtration fouling was characterized by fouling models (Hermia, 1982).

The general objective of this work is to understand the flux recovery resulting from pressure relaxation for model Alginate solutions in ultrafiltration and microfiltration processes. The specific objectives were (a) fouling characterization aided by conventional fouling with particulates, (b) comparison between the membrane flux measured with particle relaxation and the flux envisaged without it using the conventional fouling models, and(c) distinction between the de-compaction and diffusion mechanisms and examination of membrane type influence on the phenomenon.

In the first stage of this work, Alginate solutions filtration experiments were conducted in order to characterize fouling and flux by models. Particle size distribution was measured by Malvern's Master ZetaSizer and was found to be in the range of 200nm to 400nm. This size range corresponds with a previous study (Ye et al., 2005) measured size – 200nm. This size range is equal or bigger than all different membrane pore sizes which were used in this work. Therefore, fouling model type should be external to the membrane. The results of the filtration experiments, conducted with solutions containing different concentrations of Alginate and a 150 KDa membrane, was found to fit an external model type – Cake Formation (Hermia, 1982). Calculated cake specific resistances (α_c) – 1.05×10^{16}–1.45×10^{16} m/Kg corresponded with a previous study (Ye et al. 2005) result – $1.11\pm0.08\times10^{16}$ m/Kg.

Calculated cake specific resistance for the highest Alginate concentration (50 mg/L) experiment was the lowest. A possible explanation could be that in high concentrations self-coagulation

occurs, forming a filtration cake made of bigger particles, bigger pores and a reduced specific resistance. A Jar test experiment showed evidence of Alginate self-coagulation.

In the next stage of this work, models were used for flux extrapolations of 25 mg/L and 12.5 mg/L Alginate solutions filtration experiments. Flux extrapolations deviations from the measured flux were less than 3 L/m^2·hr (±10%). Later on, a preliminary pressure relaxation experiment was conducted and the Cake Formation model was used to compare the extrapolated flux (based on the data collected before the pressure relaxation) with the measured flux affected by the pressure relaxation.

In the next stage of this work, pressure relaxation experiments with different membranes and different values of diffusion related parameters were conducted.

In the experiments conducted with 0.2µm membranes a significant flux recovery was measured. Experiments data was found to fit the Intermediate Pore Blocking model. K_i (Intermediate pore blocking model constant, m^{-3}) values calculated using the model ranged 5.31E+03 m^{-3} to 9.71E+03 m^{-3}. When trying to compare these values to the literature it was found that most studies used the Cake Formation model rather than the Intermediate Pore Blocking model, therefore, it wasn't possible to compare these results which similar experiments results. However, the order of magnitude was found to match previous results with other solutions – 4.97E+02 m^{-3} to 7.63E+02 m^{-3} with Kaolin 0.1 w/w% (Chung et al. 2001), 1.62E+04 m^{-3} with mineral oils (Ohya et al. 1998).

As before, the model was used to compare the extrapolated flux (based on the data collected before the pressure relaxation) with the measured flux affected by the pressure relaxation. This analysis showed that the flux recovery was more than 50%. It also showed that when diffusion is increased by applying longer pressure relaxation duration or by using a lower bulk concentration, flux recovery gets bigger as expected.

Flux recovery analysis showed that the de-compaction mechanism effect is stronger than the diffusion mechanism effect by at least one order of magnitude. Calculated cake compressibility factor (n) – 0.25, corresponded with previous study (Ye et al., 2005) results: 0.16 for 0.2µm TE membranes and 0.29 for 0.22µm PVDF membranes. Calculated diffusion coefficient (D) – 4E-12 [cm^2/s] was a few orders of magnitude smaller than previous study (Martinsen et al., 1992) results – 1E-06 [cm^2/s] for 0.25 w/w% BSA solution. A possible explanation for this small diffusion coefficient could be the strong intermolecular interactions between the Alginate particulates and the membrane as explained in previous study (Ye et al., 2005).

In the experiments conducted with 4KDa membranes a slight flux recovery was measured and no model was found to fit all experiments. Therefore, it wasn't possible to use a model for flux recovery evaluation or to calculate mechanisms parameters as conducted for the 0.2µm membranes experiments.

As in the 0.2µm membranes experiments, it was shown that when diffusion is increased by applying longer pressure relaxation duration or by using a lower bulk concentration, flux recovery gets bigger as expected.

A possible explanation for this relatively slight flux recovery may be found in the cake compressibility factors measured in a previous study (Ye et al., 2005). In that study, conducted with 50 mg/L Alginate solutions, cake compressibility factors were – 0 for 100KDa PES membranes, 0.16 for 0.2µm TE membranes and 0.29 for 0.22µm PVDF membranes. It seems that as the membrane pore size get's bigger so does the cake compressibility factor, so we should expect a bigger de-compaction effect for the 0.2µm membranes experiments than for the 4KDa membranes experiments.

The relatively slight flux recovery may be also explained by the high TMP applied in the 4KDa membranes experiments (250 KPa) compared with the low TMP applied in the 0.2µm membranes experiments (33 KPa). It is likely that when the TMP is higher, the de-compaction and the following compaction are faster, and therefore it will be harder to observe any de-compacition related flux recovery in this case.

Additional experiment with a solution containing dominant biofouling component was conducted. The cake formation model was used to compare the extrapolated flux (based on the data collected before the pressure relaxation) with the measured flux affected by the pressure relaxation and by the biofouling. This analysis showed that at first, the measured flux was bigger than the extrapolated flux, meaning that the de-compaction and diffusion were more dominant than the biofouling. Further on, the measured flux declined sharply probably due to the feed solution changes as seen in the turbidity and cake formation model parameters increase. A further analysis for experiments including biofouling could be achieved only after the de-compaction and diffusion mechanisms are studied in a way that their effect could be predicted accurately.

To conclude, it is possible to say that the use of the fouling models (Hermia, 1982) enabled comparing the extrapolated flux (based on the data collected before the pressure relaxation) with the measured flux affected by the pressure relaxation.

For both 0.2µm and 4KDa membranes experiments, it was shown that when diffusion is increased by applying longer pressure relaxation duration or by using a lower bulk concentration, flux recovery get's bigger as expected. However, the effect of the diffusion mechanism was found to be secondary to the de-compaction mechanism.

While a significant flux recovery was measured in the 0.2µm membranes experiments, only a slight flux recovery was measured in the 4KDa membranes experiments. One possible explanation could be a weaker de-compaction mechanism due to a smaller cake compressibility factor (n) in the 4KDa membranes experiments, another explanation could be a faster de-compaction and following compaction mechanism due to higher TMP in the 4KDa membranes experiments.

These possible explanations should be studied further. Regarding the cake compressibility factor (n), it is possible to measure it directly by conducting a set of experiments with different TMP as conducted in a previous study (Ye et al., 2005). In addition, it may be interesting to study the connection between cake compressibility factor (n) and membrane type.

A significant flux recovery of more than 50% was measured in the 0.2µm membranes experiments even when the pressure relaxation duration was very short (1 minute) meaning that the effective flux recovery is significant as well. Therefore, there is a base for a further more applicable study using pressure relaxation cycles on pilot scale system.

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