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**Modeling of MF/UF membrane fouling by a protein: A new multiscale approach**

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Pressure-driven membrane processes have strongly gained importance in industrial separations over the past three decades. Numerous improvements in the technology – for instance, development of highly selective and permeable membranes, improvement in peripheral technology – have led to widespread adaptation of this process in chemical, environmental, pharmaceutical and biomedical applications.

Membrane fouling and subsequent permeate flux decline are inevitably associated with the pressure-driven membrane processes. Despite the various studies on membrane fouling and related phenomena, the fundamental mechanisms and processes involved are still not fully understood. Typical observations of permeate flux over time reveal a rapid initial decline followed by a more gradual long-term decline. This indicates the increase of the membrane resistance with time. The resistance of membrane is modeled via different mechanisms (pore blockage, concentration polarization, and cake formation). These models depend on the experimental parameters and do not describe the fouling process completely. It should be noted that the characteristics of the system change with time because of the evolution of molecules (such as proteins) or their interactions with environment (a porous medium and a fluid). Furthermore the problem is multiphysics (hydrodynamics, mass transport, physical chemistry) and multiscale (molecule, membrane pores and membrane).

The fouling of membrane during the filtration of complex media containing microorganisms is frequent (wastewater, cleaning solutions, microorganisms cultures etc). Microorganisms can attach, grow, multiply, and relocate on the membrane surfaces they can excrete extracellular polymer substances (EPS). EPS are primarily composed of polysaccharides and proteins. The fouling can be located on the surface and in the volume of the membrane. The fouling of membranes by proteins is also present in many post treatment of food products (extraction of proteins from milk, concentration of cooking juice for example).

The objective of this work was to focus on the fouling of ultrafiltration and microfiltration membranes by proteins. In contrast of small molecules that behave like rigid molecules, most proteins do not simply attach or detach from an interface with certain adsorption and desorption probabilities. Instead, the complex composition and structure of proteins causes more complex phenomena such as structural re-arrangements, changing surface affinities during adsorption, positive cooperative effects, overshooting adsorption kinetic or surface aggregation.

Firstly we were interested in better understanding the interactions between the membranes and proteins and the mechanisms of fouling. Secondly the modeling of the membrane process was developed in order to describe and predict the performance of the filtration system. The proposed model could then be validated by new filtration experiments.

BSA (Bovine Serum Albumin) solutions in Milli-Q water was used as a model of proteins in the project. The commercial PES UF/MF membranes were used. The membrane nominal cut-offs obtained from manufacturer were 0.1 and 0.01  $\mu\text{m}$ . Ultrafiltration experiments performed on a plate module (Ray flow X100, Orelis-Novasep) allowing the use of 100  $\text{cm}^2$  flat membrane. Fouling with 2 L of the BSA solution was performed over night at ambient temperature at constant TMP of 1.5 bar. Adsorption isotherms in batch mode were carried out to characterize the

interactions. The mechanisms (pore blocking, cake formation, concentration polarization) occurring in the fouling membranes were identified with filtration experiments. The membrane properties (permeability, porosity, pore diameter, thickness of the different parts of the membrane) were analyzed by means of experiments (filtration, SEM, AFM etc).

The method of volume averaging is used for the modeling approach. It is one of the techniques for modeling of the porous media. It provides a rigorous foundation for the analysis of a porous system. The development is based on classical continuum physics, and it provides both the spatially smoothed equations and a method to predict the effective transport coefficients that appear in those equations.

In this project, local equations in the porous medium (membrane) were identified and described at the local scale thanks to experimental results of measurements of interactions and identification of mechanisms. The model of membrane fouling is based on diffusion of species and adsorption at the fluid-solid interface. The method of volume averaging was then applied for upscaling the local equations and obtaining the macroscopic equations with effective coefficients.

**Keywords:** Ultrafiltration and microfiltration processes, Fouling, Modeling, Proteins