

# **Diffusive Tortuosity Factor of Colloidal Cake and Biofilm**

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Membrane fouling of fine colloids and biocolloids is ubiquitous in pressure-driven membrane processes, such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO). In MF/UF, colloidal fouling is initiated by rapid formation of the concentration polarization (CP) layer, and is significantly enhanced by gradual development of the cake layer of colloidal maximum packing. In RO/NF, the hydraulic resistances from the CP and cake layers are, however, negligible compared to the membrane resistance. Nevertheless, the cake layer causes remarkable permeate flux decline by playing an obstructive role in back diffusion of solute ions: the solutes need to evade the colloids by taking tortuous paths within the cake layer. Therefore, their back diffusion is hindered; the concentration polarization is enhanced; the osmotic pressure is elevated; and then the permeate flux is eventually suppressed to a certain extent. These phenomena become more significant in biocolloidal fouling due to extra polymeric substances (EPS) production that generates very low porosity of the biofilm. In this light, of great importance is estimating the hindered diffusion coefficient of solutes within the cake layer as a function of cake structure and porosity.

In this study, we define the diffusive tortuosity factor (DTF) as a ratio of the diffusion coefficient in bulk phase to that in the cake layer, and consider four different structures as possible configurations of colloids and biocolloids within the cake layer and biofilm, respectively: those are simple cubic (SC), body centered cubic (BCC), face centered cubic (FCC), and disordered cake structure. The porosity of the colloidal cake layer is controlled by changing inter-particle distance of colloids, and the low porosity of the biofilm is mimicked by allowing overlaps among the biocolloids and taking account of only the void space within the biofilm. Then, we perform random walk simulations of non-interacting solutes and calculate DTF of each structure.

We find by simulations that DTF can be well described by Maxwell's theoretical prediction in the colloidal cake layer; however, simulation results surpass the theory and almost diverge while the porosity of the biofilm decreases to zero. Importantly, the disordered cake structure has the highest DTF with respect to volume fraction as much as 0.94 (porosity of 0.06), which implies that the solute diffusion within the biofilm of a disordered, compressed structure is most hindered so that the concentration polarization and osmotic pressure of solutes are significantly enhanced on the membrane surface, causing noticeable permeate flux decline.

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