

# Microfiltration of deformable microgels

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## **Supplementary material**

The model described below aims to correlate the evolution of impedance change as a function of permeated volume. Two successive deposition phases are considered: (1S) pore narrowing due to accumulation of microgels within the porous membrane; and (2) build-up of non-porous gel layer as a result of microgels deposition on the membrane surface. Each of these two cases is assigned with a different equivalent circuit, resulting in two different equations for the growth of electrical impedance. The pore narrowing model (1) describes a scenario in which particles, characterized by electrical conductivity different from that of the solvent, gradually decreasing the open volume of the pores within the membrane ( $V_p$ ) up to a certain penetration depth ( $\delta_p$ ). It is assumed that continuity of the solvent phase is maintained at least within

some of the pores until they are completely blocked, therefore the overall electrical resistance for the pore narrowing scenario ( $R_{PN}$ ) may be described by an equivalent circuit comprising two resistances in parallel (Eq. 1), i.e. the resistance of the solvent ( $R_s$ ) and of the microgels ( $R_{MG}$ ). Furthermore, the conductivity of the membrane material, polypropylene in the current case, is neglected.

$$\frac{1}{R_{PN}} = \frac{1}{R_s} + \frac{1}{R_{MG}} \quad (1)$$

Electrical resistance (which equals to the impedance in the case of zero phase shift) is a function of the material conductivity ( $\sigma$ ), the cross-sectional area ( $A$ ) and the length, which in the case of non-straight pores is the particles penetration depth multiplied by the tortuosity ( $\tau$ ) as shown in Eq. 2.

$$R = \frac{d_p t}{As} \quad (2)$$

With the progression of pore narrowing, the volume of solvent ( $V_s$ ) in the pores is replaced by the volume of microgels ( $V_{MG}$ ), thus in any point in time  $V_s = V_0 - V_{MG}$ . Where  $V_0$  is the total volume of the pores (for the thickness determined by the penetration depth), which can be determined by the membrane area ( $A_m$ ), volumetric porosity ( $\epsilon$ ) and penetration depth ( $V_0 = A_m \delta_p \epsilon$ ). Combining these definitions with Eqs. (1) and (2), the electrical resistance of the narrowing pore segment can be expressed as a function of deposited microgels volume.

$$R_{PN}(V_{MG}) = \frac{t^2 d_p^2}{V_0 s_s - V_{MG}(s_s - s_{MG})} \quad (3)$$

The change in impedance (noting that  $\Delta Z = \Delta R$  for low frequencies as explained above) can then be defined by subtracting the resistance at zero deposited volume from the right hand side of Eq. (3). After rearranging, replacing  $V_0$  with  $A_m \delta_p \varepsilon$  and introducing the ratio of microgels conductivity to solvent conductivity ( $\phi = \sigma_{MG} / \sigma_S$ ) Eq. (4) is obtained.

$$DZ_{PN}(V_{MG}) = \frac{t^2 dV_{MG} (1-f)}{A_m \varepsilon_S [A_m \varepsilon_p - V_{MG} (1-f)]} \quad (4)$$

At the initial stage of filtration, when  $V_{MG}$  is low, a linear increase of  $\Delta Z$  is predicted by Eq. 4, while a steeper increase of  $\Delta Z$  is predicted when  $V_{MG}$  approaches the initial empty volume of the pores  $V_0$ . Note that Eq. 4 is physically valid as long as  $V_{MG} < V_0$ , since following that point the pores are completely full and microgels can deposit only on the surface of the membrane.

The same principles were used in the derivation of a model for the change in impedance imposed by surface deposition ( $\Delta Z_{GL}$ ). Assuming the microgels create a non-porous gel layer on the surface, the equivalent circuit can be described as three resistors in series, i.e. the membrane, the gel layer and the solvent between the feed-side electrode and the gel layer. We recently derived a model for a porous cake layer build-up by hard non-conducting silica particles<sup>14</sup>. For the case of conducting non-porous gel layer, we obtained Eq. (5).

$$DZ_{GL}(V_{MG}) = \frac{V_{MG} (1-f)}{A_m^2 S_S f} \quad (5)$$

Fitting the model to the impedance results

The membrane's porosity and tortuosity were 0.74 and 1.29 respectively as reported by the manufacturer, while the microgels volume was estimated based on their dry concentration (0.155 g/l), approximated water content (90%) and assumed density (1.2 g/ml). Based on the FESEM image shown in Fig. 2, the penetration depth of microgels to the pores ( $\delta_p$ , appearing in Eq. 4) was determined as the full membrane width (170  $\mu\text{m}$ ) for the case of 0.3 bars.