Interaction mechanisms associated with organic colloid fouling of ultrafiltration membrane in a drinking water treatment system

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HIGHLIGHTS

- The XDLVO approach is effective for predicting UF membrane fouling caused by NOM.
- Hydrophility and polarity of membrane are benefits to alleviate membrane fouling.
- Particle size and total surface energy of colloid influences membrane fouling
- LW is dominant to total interaction energy contribution if separation is beyond 5 nm.
- AB is a main contributor to total interaction energy if separation is under 3 nm.

ARTICLE INFO

ABSTRACT

The Extended Derjaguin Landau Verwey Overbeek (XDLVO) approach was introduced to predict organic colloid fouling of membranes in an initial ultrafiltration (UF) phase. Two polymeric UF membranes, made of polyvinylchloride (PVC) and polyvinylidene fluoride (PVDF) respectively, were selected to investigate membrane fouling by filtering water samples with different organic colloid compositions. The experiment was performed to determine the fouling contributions of van der Waals (LW) interactions, electrostatic (EL) interactions, as well as double layer and short-range acid–base (AB) interactions, to the total interaction energy caused by organic colloids attaching to UF membrane surfaces. The results showed that LW interaction energy predominated when the distance between the membrane surface and organic colloid was >5 nm, while AB accounted for a key contribution to total interaction energy over short distances (<2.5 nm). The influence of EL interaction energy was ignored in the total interaction energy composition. The surface energy, among all characteristics of membrane material, was a dominant factor affecting membrane fouling. The experimental results of initial ultrafiltration of raw water from the actual water source were in accordance with the predictions based on XDLVO theory, indicating that it was a feasible option for predicting membrane fouling during the initial ultrafiltration phase.

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1. Introduction

Membrane technology is used worldwide to facilitate compliance with current and anticipated water quality regulations. Ultrafiltration (UF) membrane techniques attract considerable attention, due to their capacity to remove particulates by size exclusion, which usually produces a filtrate that is free of turbidity and bacteria [1–4]. Nonetheless, membrane fouling implies a substantial loss of hydraulic permeability and requires frequent replacement of membrane modules, resulting in increased costs. Membrane fouling is a serious impediment to the use of low-pressure membrane systems as a substitute for conventional

drinking treatment processes. Previous studies have shown that natural organic matter (NOM) is the main cause of membrane fouling [5–8]. The mechanisms of NOM fouling involve a number of influencing factors, such as molecular weight distribution, Zeta potential, and other characteristics of the solution that is being filtered. It is not clear whether or not the membrane fouling formation mechanisms are caused by the interaction of multiple influencing factors. The XDLVO approach has therefore been introduced to investigate the fouling mechanism and to provide theoretical support for the control of membrane fouling that has resulted from complex organic colloid interactions [9,10]. Colloids are fine particles in an approximate size range of 1–1000 nm. In aqueous solutions NOM usually bonds together to form colloidal aggregates of tens or hundreds of nanometers in size. The attachment of a colloidal particle to a surface can be described by the XDLVO theory, which takes the following three interactions into consideration (as membrane fouling contributors): van der Waals (LW) interactions,

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electrostatic (EL) interactions, and double layer and short-ranged acidic–basic (AB) (electron donor/electron acceptor) interactions. These three interactions account for the total energy composition, in membrane fouling caused by organic colloids attaching to the UF membrane surface. The LW interaction, caused by intermolecular forces, is the attractive force between a particle and a surface (or between two colloidal particles). The EL interaction, from an electrical double-layer force, is a repulsive force for similarly-charged particle/surfaces, while it is attractive for oppositely-charged particles/surfaces. The AB component, arising from the short-range electron-acceptor–electron-donor interaction, can either be attractive, for hydrophobic colloid/surfaces, or repulsive for hydrophilic surfaces, due to hydration forces. The positive interaction force, between organic colloids and the UF membrane surface, represents a repulsive action to reject membrane fouling, while a negative one causes an attraction that aggravates fouling.

Previous studies have shown that the XDLVO theory can be used to predict membrane fouling. Jonathan [10] used the XDLVO approach to describe the influence of colloidal fouling on reverse osmosis membranes. Lin [11] made use of the XDLVO theory when studying the fouling behavior of membranes caused by soluble microbial products (SMPs). Seok-Tae [12] established a bench-scale membrane filtration system to enable direct microscopic observation of microbial cell deposition and release in cross-flow microfiltration. There is, however, a paucity of available information on the interactions between NOM and UF membranes, when considering the XDLVO theory. According to the assumptions of the XDLVO theory, the surface to which colloidal particles attach should be smooth, which is similar to the UF membrane surface during the initial ultrafiltration phase. As is well known, the deposition of colloidal particles may cause the membrane surface to become gradually rougher during the middle and final ultrafiltration phases. In this paper we have therefore used XDLVO theory to investigate initial UF membrane fouling caused by NOM, and to determine the predominant factors that influence the total interaction energy composition. Two commercial polymeric UF membranes, of PVC and PVDF respectively, were selected to investigate membrane fouling, by filtering water samples of various organic colloid composition. The polarities of membrane surface and organic matter were measured using the Lifshitz–van der Waal acid–base approach [13], which was used in an XDLVO analysis to evaluate membrane–colloid interactions. During the initial phase, the objectives of this paper were to investigate the factors and mechanisms that influenced organic colloidal fouling of UF membranes. The feasibility of the XDLVO approach for predicting membrane fouling was estimated from the experimental results of the ultrafiltration of raw water from the actual water source.

2. Materials and methods

2.1. Theory

The XDLVO theory describes the total interaction energy, or adhesion energy, between a colloid and a membrane surface as the sum of LW, AB and EL interaction [10].

$$U_{mlc}^{XDLVO} = U_{mlc}^{LW} + U_{mlc}^{EL} + U_{mlc}^{AB}, \tag{1}$$

where U^{XDLVO} is the total interaction energy between a membrane surface and a colloid immersed in water, U^{LW} is the LW interaction term, U^{EL} is the EL interaction term, and U^{AB} is the AB interaction term. The subscripts 'm', 'l' and 'c' correspond to the membrane, the bulk feed solution, and the colloid, respectively. All three variables are expressed in mJ/m^2.

The LW interaction energy between a membrane surface and a colloid in an aqueous environment can be expressed as follows [14]:

$$U_{mlc}^{LW}(y) = 2\pi\Delta G_{y_0}^{LW} \frac{y_0^2 a_c}{y}, \tag{2}$$

where y_0 is the minimum equilibrium cut-off distance, usually assigned a value of 0.158 nm (± 0.009 nm) [10–12]; y is the separation distance between the flat plate (membrane) and the sphere (colloid), expressed in nm; a_c is the radius of the spherical colloid, expressed in nm; and $\Delta G_{y_0}^{LW}$ is the LW adhesion energy per unit, expressed in mJ/m^2.

The AB interaction energy between a membrane surface and a colloid can be expressed as follows [14]:

$$U_{mlc}^{AB} = 2\pi a_c \lambda \Delta G_{y_0}^{AB} \exp\left[\frac{y_0 - y}{\lambda}\right], \tag{3}$$

where λ is the characteristic decay length of AB interaction in water, commonly measured as 0.6 nm for aqueous systems [10,14]; and $\Delta G_{y_0}^{AB}$ is the AB adhesion energy per unit, expressed in mJ/m^2.

The EL interaction energy between a membrane surface and a colloid in an aqueous environment is expressed as follows [14]:

$$U_{mlc}^{EL}(y) = \pi\varepsilon_0\varepsilon_r a_c \left(2\zeta_c\zeta_m \ln\left(\frac{1 + e^{-\kappa y}}{1 - e^{-\kappa y}}\right) + \left(\zeta_c^2 + \zeta_m^2\right) \ln\left(1 - e^{-2\kappa y}\right) \right), \tag{4}$$

where $\varepsilon_0\varepsilon_r$ is the dielectric permittivity of the suspending fluid, usually assigned as 6.95×10^{-10} $C^2 \cdot J^{-1} \cdot m^{-1}$ [10,14]; κ is the inverse Debye screening length, assigned as 0.104 nm^{-1} [10,14]; ζ_c and ζ_m, expressed in mV, are the surface potentials of a membrane and a colloid, respectively.

When the separation between two surfaces is close to the minimum equilibrium cut-off distance, the LW/AB adhesion energy per unit can be expressed as:

$$\Delta G_{y_0}^{LW} = 2\left(\sqrt{\gamma_l^{LW}} - \sqrt{\gamma_m^{LW}}\right)\left(\sqrt{\gamma_c^{LW}} - \sqrt{\gamma_l^{LW}}\right) \tag{5}$$

$$\Delta G_{y_0}^{AB} = 2\sqrt{\gamma_l^+}\left(\sqrt{\gamma_m^-} + \sqrt{\gamma_c^-} - \sqrt{\gamma_l^-}\right) + 2\sqrt{\gamma_l^-}\left(\sqrt{\gamma_m^+} + \sqrt{\gamma_c^+} - \sqrt{\gamma_l^+}\right) - 2\left(\sqrt{\gamma_m^+\gamma_c^-} + \sqrt{\gamma_m^-\gamma_c^+}\right), \tag{6}$$

where γ^{LW}, γ^+ and γ^- are the surface tension parameters, expressed in mJ/m^2; and the subscripts 'm', 'l' and 'c' correspond to the membrane, the bulk feed solution and the colloid, respectively.

Surface energy can be calculated using the contact angle parameter and the acid–base approach. According to the extended Young Equation [15], the contact angle of a liquid on a solid surface can be expressed as:

$$\left(\gamma_L^{LW} + 2\sqrt{\gamma_L^+\gamma_L^-}\right)(1 + \cos\theta) = 2\left(\sqrt{\gamma_S^{LW}\gamma_L^{LW}} + \sqrt{\gamma_S^+\gamma_L^-} + \sqrt{\gamma_S^-\gamma_L^+}\right), \tag{7}$$

where θ is the contact angle and the surface tension parameters (γ_S^{LW}, γ_S^+, γ_S^-) of a solid can be obtained by determining the contact angles between the solid and three probe liquids with well-known surface tension properties (γ_L^{LW}, γ_L^+, γ_L^-). In this study, the UF membrane surface was treated as a solid, for the purpose of attaining surface energy parameters.

2.2. Membrane

Hollow fiber UF membranes, made of PVC and PVDF respectively, were provided by the Litree Purifying Technology Co., Ltd. (Suzhou, China), for this study. The UF membrane has a molecular weight cut-off (MWCO) of 50,000 Da and an effective surface area of 0.133 m^2. Bench-scale UF experiments were designed as an outside-in type of membrane reactor. Detailed characteristics of the membrane are summarized in Table 1.