



Fractal structure and permeability of membrane cake layers: Effect of coagulation–flocculation and settling as pretreatment steps



Hira Amjad^{a,b}, Zahiruddin Khan^a, Volodymyr V. Tarabara^{b,*}

^a Institute of Environmental Sciences and Engineering, National University of Sciences and Technology, H-12 Campus, Islamabad, Pakistan

^b Department of Civil and Environmental Engineering, Michigan State University, East Lansing, MI 48824, USA

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ABSTRACT

Effect of pretreatment on the flux performance of ultrafiltration membranes is studied and interpreted in terms of structural characteristics of suspended flocs and membrane cakes formed from these flocs. Coagulation–flocculation with different values of $\bar{G}t_{mix}$ and optional pre-settling are applied to approximate pretreatment conditions of inline, direct, and conventional filtration processes. A new method for experimentally determining fractal dimension of membrane cakes, D_{cake} , based on permeate flux data and Happel permeability model is proposed and employed to show that D_{cake} correlates with the fractal dimension of flocs, D_{floc} . The broad range of D_{floc} values ($1.95 < D_{floc} < 2.56$) is found to map onto a narrow range of D_{cake} values ($2.92 < D_{cake} < 2.99$) indicating breakup and restructuring that flocs undergo as they are incorporated into a growing cake. Information on the structure of flocs and membrane cakes complements flux data and may be useful in optimizing feed pretreatment in terms of the tradeoff between the mass of the cake, its permeability and responsiveness to hydraulic and chemical cleaning.

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1. Introduction

Coagulation and flocculation are sometimes described as Victorian era technologies in reference to the long history of their application as water treatment processes [1]. Indeed, it was in the 19th century when G.W. Fuller published his pioneering work on the application of aluminum sulfate prior to sand filtration of Ohio River water in Louisville, Kentucky [2,3]. Much of what is known about the effect of coagulation on the constituents of surface water has been learned in the context of conventional water treatment where screening, coagulation–flocculation and secondary settling precede sand filtration and disinfection. Since the entry of membrane technologies into municipal water treatment practice in the late 1980s, coagulation has been extensively studied [4–8] and applied as pretreatment to reduce fouling of membrane filters.

With membrane filtration processes gaining ground as an alternative to granular medium filtration, the role of pretreatment in reducing operational costs can only be expected to increase. The trend is partly driven by the shift to source waters of lower quality and the ushering in of water reuse as one of sustainable long-term solutions to the water challenge. Because of the high effectiveness and reliability of membranes in treating a broad range of source

waters, the emphasis will be on reducing the cost of such treatment. In this regard, minimizing irreversible fouling and cost-effectively managing reversible fouling of membranes are key challenges.

Natural organic matter (NOM), a major component of most surface waters, is a complex mixture of organic compounds with composition that can vary greatly from one water source to another [9]. NOM is responsible for adsorptive and colloidal fouling of membranes and is a precursor for trihalomethanes – carcinogenic byproducts of disinfection by halogen-based agents such as chlorine gas and hypochlorite. An important component of NOM are humic materials; for an average river they correspond to ~50% of dissolved organic matter [10]. Because of NOM's inherent heterogeneity and variability and because of difficulties with characterizing NOM definitively, humic acid is often used in model studies as a standard surrogate material representing NOM [11]. In particular, humic acid has been used extensively to understand organic fouling of membranes of a range of porosities and chemical makeups (e.g. [12–14]).

It is now well established that properties of flocs (i.e. aggregates that form during coagulation and flocculation of surface water) are very important for understanding the fouling of membranes installed downstream in the treatment train [7]. Size, surface charge, and internal structure of a floc affect its stability [7,15–17] and setting rate [18], and partly determine filterability of

* Corresponding author. Tel.: +1 (517) 432 1755; fax: +1 (517) 355 0250.

E-mail address: tarabara@msu.edu (V.V. Tarabara).

membrane cakes formed from these flocs [19–21]. Given that floc properties are greatly influenced by coagulation and flocculation (e.g. [22,23]), it has been suggested ([20] and references therein) that parameters such as coagulant dose, pH, mixing and intensity can be optimized to improve the performance of membrane filtration.

The internal structure of flocs can be expressed in terms of the fractal dimension, D_{floc} [24]. Fractal dimension of flocs can be measured by a number of techniques [25] such as small-angle neutron scattering (SANS) (e.g. [19]), small-angle X-ray scattering (e.g. [26]), static light scattering (e.g. [27]), image analysis (e.g. [28]) and settling (e.g. [29]). Scattering-based methods provide non-invasive analysis of the entire ensemble of particles but are subject to certain constraints on the shape and size of primary particles and their aggregates. Techniques based on image analysis relate the number or mass of particles at various length scales; these methods are more prone to experimental error but are still used especially when scattering methods are either not applicable or not available.

Several previous studies discussed how the structure and permeability of membrane cakes depend on the fractal dimension of cake-forming aggregates, D_{floc} [15,17,19–21,30–34]. Specific cake resistance was shown to decrease with an increase in floc size and a decrease in D_{floc} [15,31,33]. In accordance with these findings, Choi et al. concluded that flocs of higher fractal dimension are more compressible and, therefore, form cakes with higher specific resistance [21]. Assuming that cake porosity is proportional to floc porosity, Guan et al. proposed a formula for the specific resistance of the cake as a function of floc's fractal dimension [31]. In a separate study, Park et al. showed that for flocs with low D_{floc} the cake porosity was barely dependent on D_{floc} , whereas for high D_{floc} cake porosity decreased with increasing D_{floc} [34]. Cho et al. concluded that at least for the conditions of their study it was likely that "local structures of the cake on membrane remained the same as in the aggregates of the original suspension" [35]. Antelmi et al. used SANS to characterize cake structures and demonstrated that cakes collapse [19].

There were only a few experimental studies where the fractal dimension of a membrane cake, D_{cake} , was determined experimentally. Pignon et al. [36] used SANS, static light scattering and local birefringence techniques and showed that filtration leads to a cake with a void structure that is characterized by a low fractal dimension. The same technique was employed by Antelmi et al. [19] in their study of cake collapse. Mendret et al. [37] used an optical method developed by their team earlier [38] to measure cake porosity as a function of cake's depth. These authors noted that "the deposit thickness growth is not linearly related to the quantity of matter deposited"; however, the fractal dimension was not calculated from this data. Fractal substructures within chemically heterogeneous cakes were also observed in experiments on the autopsy of cakes from pilot filtration tests with surface water [39]. While studies quoted above indicated that membrane cakes can be fractal, no relationships have been established between D_{floc} and D_{cake} . Also, D_{floc} and D_{cake} have not been compared for practically relevant coagulation–flocculation scenarios.

In this study, we propose a simple method for estimating fractal dimension of membrane cakes, D_{cake} . The method is based on data easily measureable in a constant pressure unstirred filtration test. We then apply this new approach to quantify the structure of membrane cakes formed from feed waters subjected to different combinations of coagulation, flocculation and pre-settling that roughly approximate pretreatment conditions of inline, direct, and conventional filtration processes. Specific objectives of our study are: (1) to quantify the structure of membrane cakes formed from flocs generated under different pretreatment conditions including mixing and pre-settling; (2) to identify a possible corre-

lation between fractal dimensions of flocs (D_{floc}) and membrane cakes (D_{cake}) formed by these flocs; (3) to interpret permeate flux and specific cake resistance data in terms of structural characteristics of flocs and membrane cakes; and (4) to explore implications of these data for the optimization of pretreatment processes.

2. Background and approach

In this section we introduce the notion of fractal dimension and equations that connect microstructural characteristics of a membrane cake with its permeability. We then propose a method for estimating the fractal dimension of a growing membrane cake based on variables that can be easily measured in a filtration experiment.

2.1. Fractal dimension of a membrane cake

The fractal dimension, D_{cake} , of a growing deposit defines how its average thickness scales with the number of deposited particles, N [40,41]:

$$L \sim N^\varphi = N^{\frac{1}{D_{cake}-2}}, \quad (1a)$$

where φ is the scaling exponent and $D_{cake} = 2 + \frac{1}{\varphi}$ is the fractal dimension of the deposit. Eq. (1a) has been used to describe and quantitatively differentiate morphologies of deposits formed under conditions characterized by a broad range of particle Peclet numbers [42] and particle–particle interaction energies [39]. The number of particles in the deposit, N , is proportional to its mass, M . In filtration with complete rejection and no back-transport of particles, the mass of deposit equals the product of feed concentration of particles, C_f , and permeate volume, V_p . Thus, when C_f is constant, N is proportional to the permeate volume and Eq. (1a) can be rewritten as

$$L \sim V_p^\varphi. \quad (1b)$$

The average thickness of the cake, L , is given by:

$$L = \frac{C_f V_p}{A_m (1 - \varepsilon) \rho_s}, \quad (2)$$

where ρ_s is particle density, ε is porosity of the cake, and A_m is the area of the membrane. Combining Eqs. (1b) and (2) yields a simple scaling relationship:

$$\ln(1 - \varepsilon) \sim (1 - \varphi) \ln V_p \quad (3a)$$

or

$$\ln(1 - \varepsilon) \sim \frac{D_{cake} - 3}{D_{cake} - 2} \ln V_p. \quad (3b)$$

When the values of permeate flow rate and cake porosity as functions of filtration time are known, Eq. (3b) can be used to determine D_{cake} . To our knowledge, this scaling relationship has not been described in the published literature.

2.2. Specific hydraulic resistance of a membrane cake

In the absence of an osmotic pressure difference across the membrane, the volumetric permeate flux is given by:

$$j = \frac{\Delta P}{\mu(R_m + R_c)}, \quad (4)$$

where ΔP is the transmembrane pressure differential, μ is viscosity of the permeating fluid, while R_m and R_c are hydraulic resistances of the unfouled membrane and the membrane cake, respectively. Specific hydraulic resistance of a membrane cake of thickness L is defined as

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