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Electrocoagulation and crossflow microfiltration hybrid system: fouling investigation

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ABSTRACT

The fouling study of crossflow microfiltration (MF) was comparatively studied with feedwater containing kaolin suspension with and without electrocoagulation (EC) pre-treatment. An acrylonitrile butadiene styrene (ABS) MF membrane of pore size 0.4 μm was used in this study. The experiments were carried out at three different concentration of kaolin (100, 400 and 800 mg/l) and with three different crossflow velocities of 0.5, 1 and 1.5 l/min. When the feedwater was pre-treated by EC, the fouling was found to follow standard law of filtration. Besides the standard filtration law, the fouling mechanism also followed the classical cake filtration model due to formation of a secondary membrane.

Keywords: Microfiltration; Crossflow; Fouling mechanism; Filtration laws; Electrocoagulation

1. Introduction

Microfiltration (MF) has been widely applied in drinking water treatment for the removal of particles, turbidity and microorganisms from surface water and groundwater as an alternative to conventional water treatment processes (coagulation, sedimentation and sand filtration) [1,2]. MF offers several advantages including superior water quality, easier control of operation, lower maintenance and reduced sludge production [3]. However, MF suffers from membrane fouling due to the dissolved and colloidal particles present in the water or wastewater and fouling significantly reduces the process performance.

Chemical flocculation and coagulation are commonly adopted pre-treatment methods for removing the colloidal and suspended participles present in feedwater before membrane application [4]. Electrocoagulation (EC) is an alternative coagulation method to the conventional chemical coagulation. Unlike the conventional chemical coagulation process that requires several moving parts and a large settling tank, EC simply uses two separate electrodes (Al or Fe or Ti) placed at certain distance apart and DC current. EC offers several advantages over conventional chemical coagulation such as: (i) no alkalinity consumption, (ii) no change in bulk pH, (iii) the direct handling of corrosive chemicals is nearly eliminated and (iv) easy adaptability for use in portable water treatment units especially during emergencies [5]. In EC, the coagulant (Fe or Al) is generated by electrolytic oxidation of a sacrificial metal anode to produce coagulating agent to dose the polluted water and in the process generating electrolytic gases (mainly hydrogen at the cathode). The most widely used electrode materials in EC

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process are aluminium and iron, sometimes steel. Titanium metal has also been recently investigated as electrode for EC [6]. The passage of electrical current between the electrodes causes the dissolution of metal into the water. The metal ions, at an appropriate pH value, can form wide ranges of coagulated species and metal hydroxides that destabilize and aggregate the suspended particles or precipitate and adsorb dissolved contaminants.

Crossflow or tangential flow is a process in which the formation of a cake layer is limited or suppressed by a flow of the suspended particles parallel to the filtration surface [7], while water is forced through the filter. Particles deposited on the filter medium are swept away by the cross-flow velocity action, which produces shear and lift forces on the particles as they become attached to the filter medium [7]. Generally, in most practical applications of the crossflow MF, a wide range of colloids and particles exists in the water to be treated. The decline in flux is a major hindrance to the wide implementation of crossflow MF for water and wastewater treatment. In crossflow MF process, there occurs a formation of a secondary or dynamic membrane on top of the primary membrane. Several studies have been done to investigate the formation of dynamic layer on top of the primary membrane during the crossflow MF process. Kaolin, lime and diatomaceous earth were often used as dynamic membranes in crossflow MF [8]. Tanny [9] studied the dynamically formed membrane in the MF with tap water using mineral species such as fluorspar, diatomite, kaolin, silicate flakes and limestone. The formation of dynamic membrane was divided into three categories [9].

Class 1 dynamic membranes are formed when the particles have a particle size greater than the pore size of the membrane. This phenomenon is widely known as concentration polarization.

Class 2 dynamic membranes are created when filtering dilute suspensions of colloidal particles of particle size much smaller than the pore size of the membrane. In this case, the flux decline mechanism was found to behave according to an internal pore clogging phenomenon rather than cake build-up on the membrane surface. Similar results were reported by other researchers [10–12]. This fouling mechanism has been described by the standard filtration law shown by the following model:

$$t/V = 1/Q_0 + k_1 t/2 \tag{1}$$

Where V is the permeate volume (ml), t is the filtration time (min), Q_0 (ml min^{-1}) is the initial flux rate and k_1 is the filtration constant. The development of this model is based on the assumption that the pore volume decreases proportionately to the filtrate volume.

After some time, the colloidal particles will be brought up to the membrane surface, and the flux behaviour will proceed in accordance with the following classical cake filtration model [13]:

$$t/V = 1/K_1\left(V - 2V_f\right) \tag{2}$$

Where V_f (l) is the volume of permeate which produces a hydraulic resistance equal to that of the membrane, and K_1 (l^2 min^{-1}) is the cake filtration constant.

The formation of dynamic membranes was investigated by Al-Malack and Anderson with crossflow MF [4]. They found that dynamic membrane formation obeys the standard law of filtration in the first few minutes of membrane formation (15 min). As time passes, the dynamic membrane formation was found to proceed according to the classical cake filtration model. In another study, the removal of phosphate from water by red mud using crossflow MF was investigated [14]. This study evaluated specific cake resistance in crossflow MF as a function of phosphate concentration by using cake filtration model.

Class 3 dynamic membranes are formed when filtering polymers or polyelectrolyte molecules of equal size to the membrane size.

The objective of this study is to investigate the fouling mechanism of crossflow MF by feedwater containing kaolin suspension with and without Al-based EC. Understanding the fouling mechanism if essential for any membrane filtration so that appropriate fouling control strategies can be developed.

2. Materials and methods

2.1. Synthetic water

Synthetic water was prepared in the laboratory using kaolin and humic acid (composition shown in Table 1). Stock solution was prepared first using 2 g humic acid sodium salt and 40 g kaolin which was then diluted with normal tap water to give a fairly constant turbidity between 80 and 85 NTU before using for EC and crossflow MF experiments.

2.2. MF membrane

A flat sheet MF membrane was used throughout the study. The detailed properties of the flat sheet MF membrane (Pure-Envitech Co., Busan, South Korea) are given in Table 2.

Table 1
Properties of synthetic water

Turbidity	Total organic carbon (TOC)	UV Abs (254 nm)	Conductivity
80 NTU	5.5 mg/l	0.150	0.0, 805 mS/cm

Table 2
General characteristics of MF membrane (as provided by Pure-envitech, South Korea) used in this study

Pore size	pH resistance range	Temperature range	Pressure range	Material
0.4 µm	2–11	2–38 °C	0–60 cm Hg	Acrylonitrile butadiene styrene (ABS)

2.3. EC setup

The EC reactor used in this study consisted of a 5-l Pyrex glass beaker with two aluminium electrodes ($17 \times 9 \times 0.2$ cm) in a monopolar configuration and the spacing between the electrodes was 2 cm [6]. The source of power supply included DC power converter (Q1770, Dick Smith Electronics, Australia). Suspensions were stirred using a magnetic stirrer adjusted to an optimal rate (250 rpm) so as to obtain the highest efficiency of turbidity removal. Experiments were performed at different generation times (5–30 min) and at wide pH range (3–11) for determining the optimum conditions. Prior to each test, 5 l of synthetic wastewater was used in the EC cell which was carried out by varying the process-operating parameters. The current was adjusted by varying the voltage. When not in use, the aluminium electrodes were immersed in acid bath (4% HCl) and prior to each experiment, they were carefully cleaned using steel wool to remove any aluminium oxide that may have formed on the surface. The desired aluminium concentration can be achieved by operating the unit under variable generation time (t) mode in accordance with Faraday's law [5]:

$$m_{Al} = (27 \times I \times t)/Z \times F \quad (3)$$

where m_{Al} is the mass of Al generated (g), I is the current, Z is the number of electrons transferred in the reaction at the electrode and F is the Faraday's constant (96,486 C/eq).

Faraday's law used for calculating the amount of aluminium indicated a good correlation between the theoretically calculated values.

2.4. MF setup

A crossflow membrane filtration unit (Nitto Denko Corp., Japan) was used to study the effect of pretreatment on membrane performance. The schematic diagram of the crossflow MF experimental setup is shown in Fig. 1. Synthetic water, with and without pretreatment, was pumped into a flat sheet membrane module (effective membrane area of 0.07×0.10 m). The operating transmembrane pressure and cross-flow velocity were controlled at 10 kPa and 0.5 l/m, respectively, by means of bypass and regulating valves. The filtrate volume was determined using an electronic balance connected to a computer for continuous data

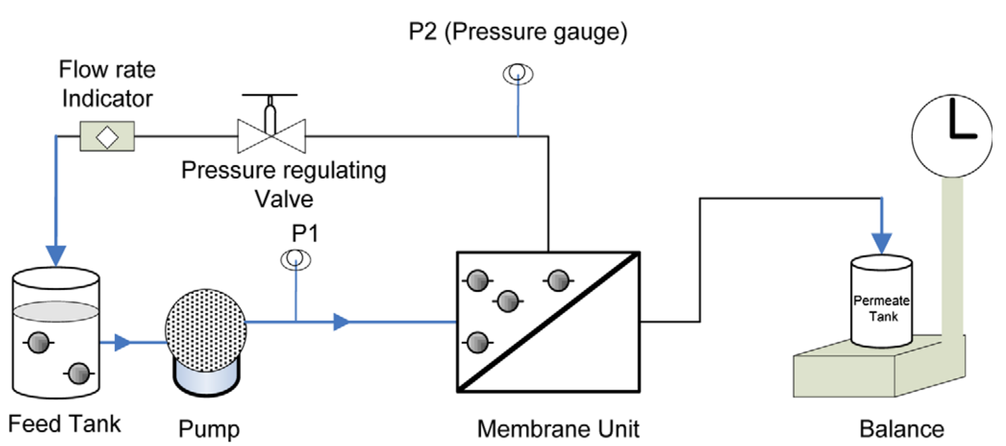

Fig. 1. Schematic diagram of the crossflow MF unit.

logging. New membranes were used in each experiment to avoid the effect of residual fouling and to compare the results obtained under different conditions.

3. Results and discussion

3.1. Crossflow membrane performance

Fig. 2 shows the crossflow MF flux with time and how it varies with Koalin concentrations and crossflow velocity (CV). The Koalin concentration has a strong influence on the MF water flux. The flux declined exponentially and this rate of flux decline was more severe when feedwater containing higher Kaolin concentrations were used. The CV also had a significant influence on the MF flux. The stabilized flux after 100 min of operation increased by up to 240% when CV was increased from 0.5 to 1.5 l/min using 100 mg/l of Kaolin as feedwater as shown in Fig. 2(a). Fig. 2(b) and (c) shows the variation in flux at different CVs with 400 and 800 mg/l of kaolin concentration, respectively. Although, higher fluxes were observed at higher CVs however, the influence of CV was slightly lower when feedwater contains higher Kaolin concentrations. The flux increase after 100 min of operation was only about 120 and 190% for 400 and 800 mg/l kaolin, respectively, when the CV was increased from 0.5 to 1.5 l/min.

3.2. MF fouling mechanisms without pre-treatment

Both the standard law of filtration and classical cake filtration model were used for the evaluation of fouling mechanism for the crossflow MF. Fig. 3 shows the relationship between t/V and t at different kaolin concentrations and at different CVs. For all the cases

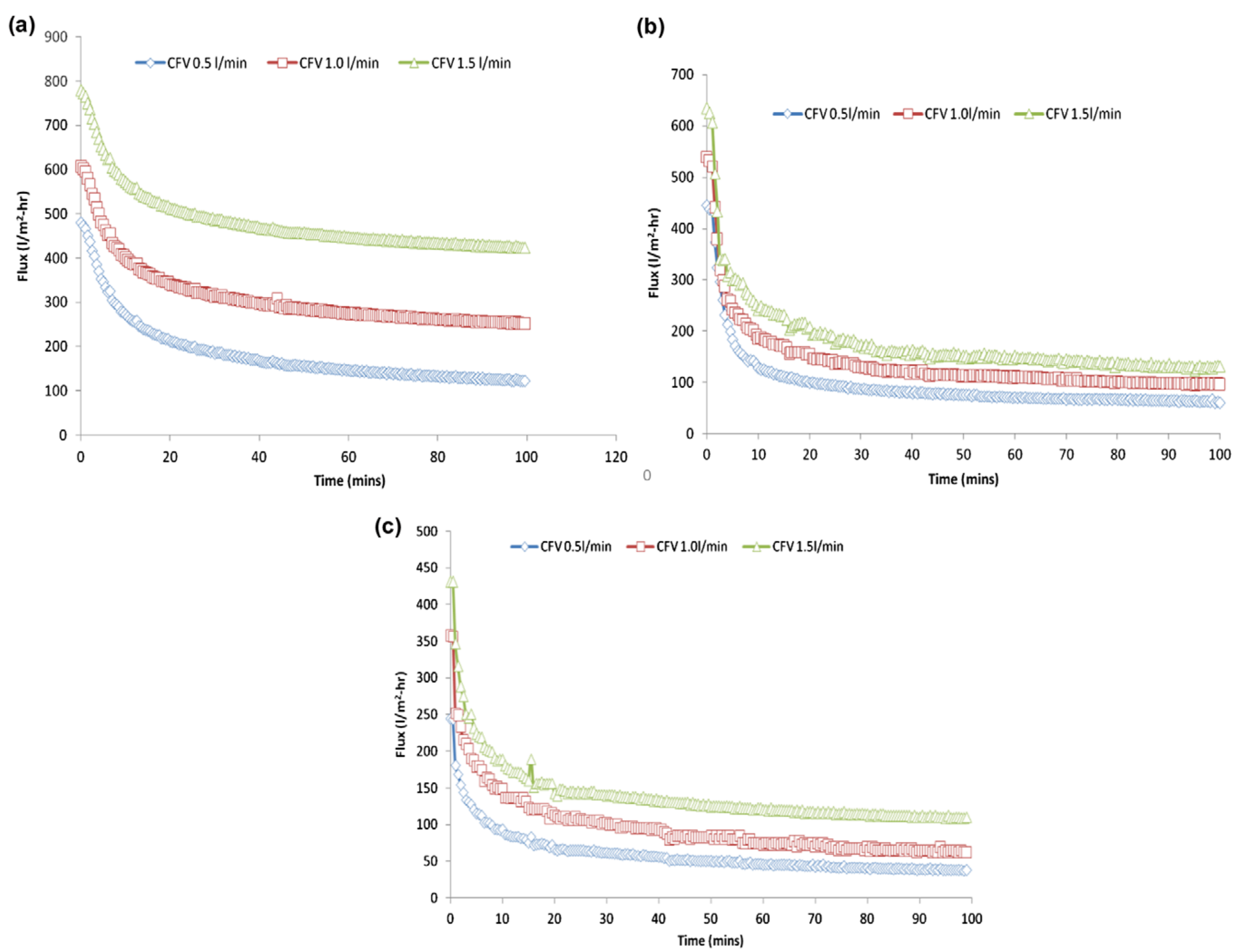

Fig. 2. Variation of crossflow MF flux with time at different crossflow velocities presentedd separately for different colloidal particle concentration (a) 100 mg/l kaolin, (b) 400 mg/l kaolin and (c) 800 mg/l kaolin.

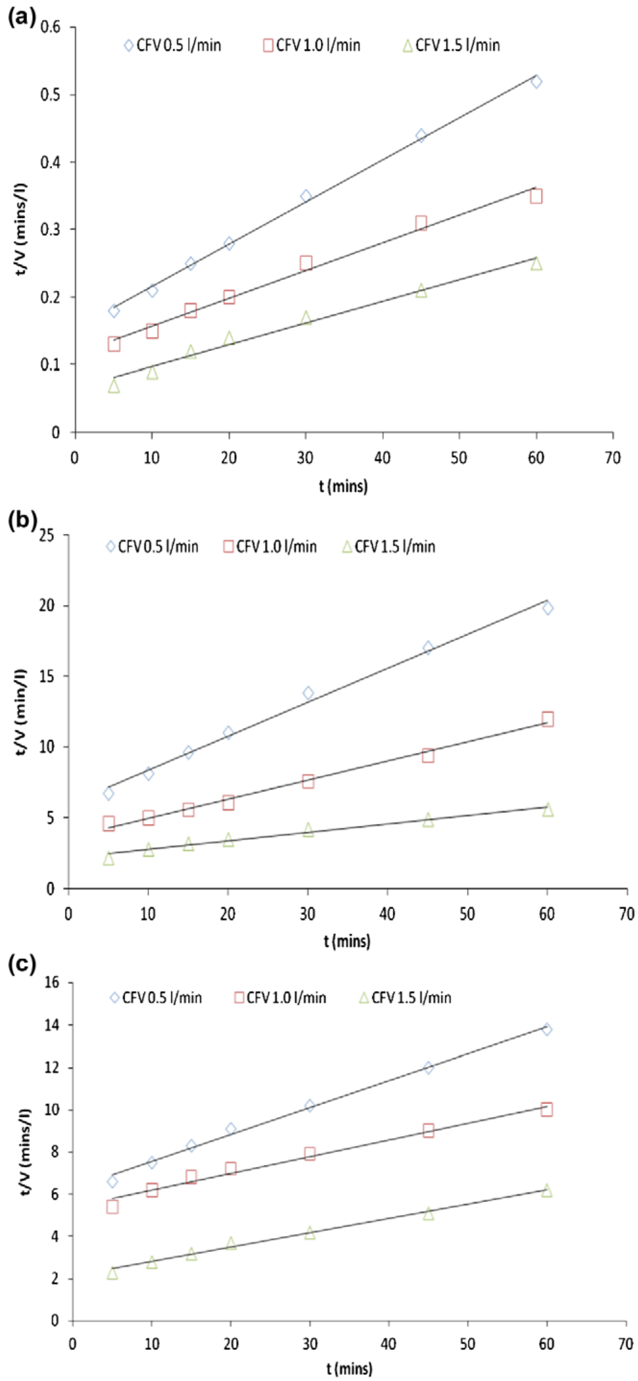

Fig. 3. Relationship between t/V and t for kaolin concentration of 800 mg/l at three different CVs.

in Fig. 3, it shows that the fouling mechanism of the primary membrane was proceeding according to the standard law of filtration which is due to the permeation of kaolin colloidal particles into the pores of the primary membrane. The standard law of filtration states that when filtering a suspension where the size of particles is less than that of the membrane pores, then the particles pass through the pores where it is assumed that the volume of the pores decreases in proportion to the volume filtered [10]. However, fractions of the particles are deposited on the internal pore surface, which results in clogging of the pores.

The summary of the fouling investigation is presented in Table 3. From Table 3, it is observed that the filtration constant k_1 decreased with an increase in CV and increased with an increase in kaolin concentration. It is explained by the fact that increase of CV decreased the hydraulic resistance of the membrane due to the shear effect of the CV on particles. Also increasing the kaolin concentration resulted in increasing the hydraulic resistance of the membrane due to deposition of particles inside the pores and on the membrane surface. It was also observed that the initial flux Q_0 was found to increase with increasing CV.

3.3. MF fouling mechanisms with EC as pre-treatment

The crossflow MF process was investigated for the effect of EC at kaolin concentration of 400 mg/l and at CV of 1.0 l/min. The EC time was varied from 0 to 30 min. Fig. 4 shows the results of MF permeate flux under different EC times. When EC as a pre-treatment was not used, the initial flux was 198 l/m^2 h and decreased to 70 l/m^2 h after 60 min of operation. However, the flux improved significantly when the feedwater was pre-treated by EC. When pre-treated by EC for 10 min, the initial flux increased to 305 l/m^2 h, which is about 77% increase from the untreated feed. The optimum EC time was found to be about 25 min and the corresponding initial MF flux reached 855 l/m^2 h, which represents more than 300% increase from the untreated feed. The flux did not change significantly when the EC time was increased beyond

Table 3
Summary of correlation for the standard law of filtration

Kaolin concentration (mg/l)	Crossflow velocity (l/min)	R^2	Q_0 (ml/min)	k_1 (l/l)
100	0.5	0.997	442	0.259
100	1	0.988	561	0.161
100	1.5	0.981	622	0.030
400	0.5	0.991	431	0.369
400	1	0.992	487	0.150
400	1.5	0.978	521	0.040
800	0.5	0.994	225	0.159
800	1	0.981	342	0.148
800	1.5	0.992	388	0.036

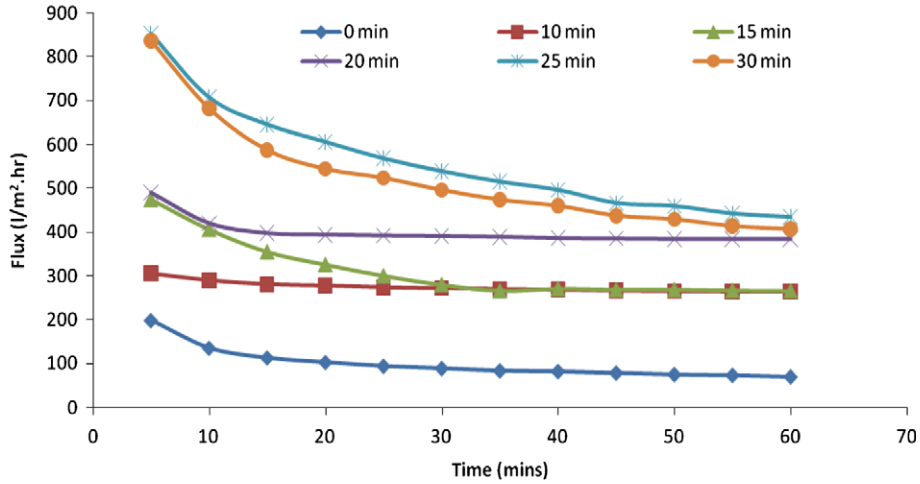

Fig. 4. Permeate flux with respect to time at various EC times (kaolin concentration = 400 mg/l, CV = 1.0 l/min).

30 min which may be due to reversing the destabilization of the colloidal particles [15]. As the experiment was done at pH 8, it is believed that the ions released are converted to $Al(OH)_3$ floc quickly causing the particles and organics to agglomerate, float, settle which can then be easily removed from the synthetic water. Tables 4 and 5 present the summary of results for the crossflow MF.

It was observed from Table 4 that fouling mechanism for all feedwater conditions followed the standard filtration law for the first 30 min which is possibly due to the permeation of colloidal particles into the primary membrane. As the MF has an enormous internal surface area of pores, at the initial stages of the operation the colloids with a size range much smaller than that of the pores will pass through the pores. Once within the pores of the membrane, these colloids become deposited or adsorbed due to various forces such as the electrical double layer effect and hydrodynamic attraction forces, and form a film of colloids on the internal pore surface [10].

Table 4
Results of the fouling mechanism for EC-MF according to standard law of filtration (kaolin concentration = 400 mg/l, CV = 1.0 l/min)

EC time (mins)	R^2	Q_0 (ml/min)	k_1 (1/l)
0	0.973	1,780	0.102
10	0.961	2,030	0.094
15	0.992	4,804	0.083
20	0.964	6,010	0.044
25	0.991	8,050	0.028
30	0.985	8,360	0.026

Table 5
Results of the fouling mechanism for EC-MF according to classical cake filtration model (kaolin concentration = 400 mg/l, crossflow velocity = 1.0 l/min)

EC time (min)	R^2	V_f (l)	K_1 (l^2/min)
0	0.963	0.20	52
10	0.952	0.42	153
15	0.981	2.10	456
20	0.95	4.24	642
25	0.98	6.85	718
30	0.97	7.20	805

Table 5 indicates that, besides the standard filtration law, the fouling mechanism also indicated a good correlation with the classical filtration model especially with the three runs corresponding to 15, 25 and 30 min of EC time. This indicates that, besides the infiltration of colloidal particles, a dynamic membrane was formed on top of the primary membrane due to the agglomerated solids.

The values for Q_0 and V_f show the effect of coagulation on membrane performance. The increase in coagulant dose increased the initial flux value (Q_0) and the volume of permeate which produces a hydraulic resistance (V_f) equal to that of the membrane. Table 5 also shows that the k_1 value was found to decrease with increase in EC time until it reached the lowest value at optimum condition. The results is in agreement with the investigation performed by Grace [16] who concluded that in general all microporous filter media exhibit an initial period of filtration in which fine particles appear in the filtrate. After this initial period, the number of particles in the filtrate decreases rapidly, and the flux decay follows the standard law of filtration, which is then followed by a prolonged period conforming to the cake filtration model.

When the internal surface of the pores becomes saturated with fine colloids, the colloids accumulate on the membrane surface, leading to the formation of a colloidal film on the external membrane surface. At the early stages of filtration, the colloids can be deposited on the membrane surface between the pores, and thus accumulate. Later, these aggregates of colloids can form bridges over the pore openings, resulting in the partial blocking of the pores, a smaller pore structure available to subsequent colloids and particle retention. This bridging of aggregated colloids leads to the eventual formation of a film on the membrane surface [10].

4. Conclusions

A bench-scale study was undertaken to investigate the fouling mechanism of crossflow MF after EC pretreatment for water containing kaolin suspension. Both the standard law of filtration and classical cake filtration model were used to evaluate the fouling mechanism for the crossflow MF. After EC pre-treatment, the fouling mechanism for MF was observed to follow the standard law of filtration possibly due to the permeation of colloidal particles into the primary membrane. Besides the standard filtration law, the fouling mechanism also showed a good correlation with the classical cake filtration model which indicates that besides the infiltration of colloidal particles, a dynamic membrane was formed on top of the primary membrane probably due to the formation of a colloidal film on the membrane surface.

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