

# Membrane fouling mechanism of submerged membrane bioreactor during erythromycin removal

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## Abstract

**Background:** Based on the previous studies, antibiotics can have affected biological properties of biomass and fouling properties of mixed liquor in aeration tank. The present study was conducted to explore the fouling mechanisms of membrane bioreactor (MBR) system during the treatment of wastewater containing erythromycin (ERY) antibiotic under several mixed liquor suspended solids (MLSS) concentrations.

**Methods:** A lab-scale two-chamber MBR system equipped with a polypropylene hollow fiber submerged membrane was fed with synthetic wastewater containing different initial concentrations of ERY. MBR system was operated under the constant flux mode and different MLSS concentrations (5.0-13.0 g/L) and the obtained results were evaluated using different individual and combined fouling models.

**Results:** The variation of MLSS concentrations had not significantly affected the kind of best-fitted model. From the individual models, the standard model indicated the best performance for permeate prediction under different MLSS concentrations ( $R^2_{adj} > 0.997$ ). For all studied MLSS concentrations, the  $R^2_{adj}$  values of combined fouling models were higher than 0.986 and demonstrated good fitness performance of combined models compared to individual models. Overall, the cake-intermediate model showed the lowest fitness, and cake-complete and complete-standard models were the most successful models in filtrated volume prediction in comparison with other combined fouling models.

**Conclusion:** This study indicated that mechanistic models are suitable for fouling prediction of MBR systems in ERY removal and under a wide range of MLSS concentrations and provide valuable information on fouling mechanisms of full-scale MBR systems.

**Keywords:** Membrane bioreactor, Membrane fouling, Modeling, Erythromycin

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## Introduction

Membrane systems such as membrane bioreactor (MBR) system are the new technologies that play an important role in increasing the efficiency of water and wastewater treatment systems in solid-liquid separation and the removal of emerging pollutants (1,2). MBR system is the combination of an activated sludge process and a membrane unit that was replaced by conventional treatment systems to alleviate their operational problems and improve the effluent quality and quantity consistency (1,3). But membrane fouling as the most challenging issue against the application of membrane units, leads to the flux decline and imposes operational and capital costs due to reversible and irreversible fouling (3,4).

Membrane fouling is defined as the blockage of

membrane pores due to the entrapment of compounds present in the inlet flow to pores, followed by an increasing flow resistance, and then, flux decreasing (4,5). Fouling quality and quantity depend on its mechanisms that are affected by the combination of membrane and inflow properties such as suspended solids, colloids, macromolecules, extracellular polymeric substances (EPS), soluble microbial products (SMPs), fouling-related microbial communities, and operational conditions such as applied pressure, the ratio of mixture dilution, and sludge retention time (SRT), etc (6-8). Thus, the characterization of membrane fouling mechanisms for its control and prevention is important (9).

For this purpose, recently, conceptual and mathematical models are developed for covering the complex effects



of operational conditions and membrane properties on fouling mechanisms for different fluids filtration (10). For example, four mechanisms have been identified for membrane fouling including standard blocking, intermediate blocking, complete blocking, and caking. These mechanisms are different in particle sizes and stages of the fouling process (11). Most studies on MBR systems aimed to identify, investigate, control, and model membrane fouling. So, development of models for the characterization and control of membrane fouling mechanisms is very important (12).

In the past few years, some comprehensive models have been developed for covering the complex effects of operational conditions and membrane properties on fouling mechanisms (13). According to the study of Kim et al (12), the combined effects of the different individual fouling mechanisms at constant pressure originated from Darcy's law can be explained by the combined fouling models such as cake-complete, cake-intermediate, complete-standard, intermediate-standard, and cake-standard models. Hu et al (8) modeled the membrane fouling using various models, from the straightforward individual models stemming from Darcy's law to the complex combined ones. The effects of applied pressure and SRT at the constant pressure on fouling were also investigated. The results showed that the cake-standard and complete-standard models had better fits at different pressure and at different dilution ratios, respectively. Wu et al (10) studied the combined mechanistic model to demonstrate the fouling mechanisms and stated that the model was promising in both experimental performances and predicting flux reduction. Also, Kim et al (12) reported that the four mechanisms originated from Darcy's law could precisely predict and reflect each individual fouling mechanism.

In the recent decade, pharmaceutical industries and domestic wastewater treatment plants have been the primary sources of antibiotics to water bodies (in parent or metabolite form). Pharmaceuticals in the environment alter the microbiome and lead to microbial resistance in microorganisms. Conventional treatment processes cannot reduce antibiotics significantly (14). Antibiotics such as erythromycin (ERY) could affect the performance of treatment plants. It is reported that ERY reduced the specific evolution rate of chemical oxygen demand (COD) and ammoniacal nitrogen by 79% and 41%, respectively. Also, ERY destructed the floc structure in the activated sludge process (15). This compound also has indicated acute inhibition in the microbial community of wastewater treatment plants (16). However, in several studies, the MBR system has shown remarkable performance in removing pharmaceuticals such as ERY (removal efficiency of 25-91%) from wastewater (17,18). These findings show that the MBR system can be a promising option for removing antibiotics from domestic and pharmaceutical wastewater.

Pharmaceuticals can interact with the foulants and change their characteristics, exerting direct or indirect effects on fouling behavior. The presence of pharmaceuticals would induce chemical stress on biomass and change their characteristics, alter the composition of foulants, and subsequently, affect the metabolism of microbial species and the abundance of microbial communities (19,20). Previous studies have shown that the presence of pharmaceuticals resulted in high fouling rates compared to the absence of them in the MBR systems (21,22). Many studies have recognized EPS and SMP as the fundamental constituents responsible for fouling in MBR systems treating pharmaceutical wastewater (23,24).

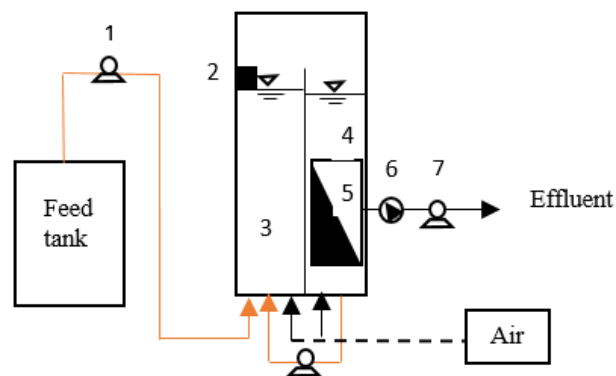
However, according to our knowledge, there are few studies investigated the modeling of fouling mechanisms of the MBR system during the removal of antibiotics. The present study was conducted to explore the individual and combined models for fouling modeling of mixed liquor in the MBR system during the treatment of wastewater containing ERY antibiotic.

## Materials and Methods

### MBR system setup and operation

A lab-scale two-chamber MBR system with a working volume of 5.0 L was constructed from Plexiglas and operated for 365 days. The MBR was equipped with a membrane module, feed and extract pump, pressure gauge, flowmeter, air compressor equipped with air stones, and water level controller (Figure 1). The main characteristics of the membrane module are summarized in Table 1.

At first, the MBR was acclimated with activated sludge derived from full-scale wastewater treatment plant (Isfahan, Iran) to achieve mixed liquor suspended solids (MLSS) equal to 3.0 g/L based on the previous study (25). Before sludge acclimation, the parent sludge is sieved and washed with tap water to eliminate any debris and impurities. MBR was operated for 30 days at feed COD of 250.0 mg/L and hydraulic retention time (HRT) of 24 hours without ERY addition. The feed media was prepared by tap water and contained per liter: 0.681 g glucose as a



**Figure 1.** Schematic diagram of MBR system: (1) Feed pump, (2) Flow level meter, (3) Aeration tank, (4) Sedimentation tank, (5) Membrane module, (6) Pressure gage, (7) Peristaltic pump.

**Table 1.** Characteristics of membrane module used in the present study

Properties	Specification
Material	Polypropylene
Type	Hollow fiber
Capillary thickness	40-50 $\mu\text{m}$
Capillary outer diameter	450 $\mu\text{m}$
Capillary pore diameter	0.01-0.2 $\mu\text{m}$
Ventilation rate	$7.0 \times 10^{-2} \text{ cm}^3/\text{cm}^2 \cdot \text{S} \cdot \text{cm Hg}$
Porosity	40-50%
Lengthways strength	120000
Designed flux	6-9 $\text{L} \cdot \text{m}^2 \cdot \text{h}$
Area of membrane module	0.1 $\text{m}^2/\text{module}$
Operating pressure	-0.01-0.03 MPa
Abnormal pressure	- <0.05 Kpa
Length of pipe	0.37 m

carbon source, 0.05 g urea, and 0.01 g  $\text{K}_2\text{HPO}_4$  as a nutrient (C: N: P ratio of 100:5:1). Other trace elements including  $\text{CuSO}_4 + 5\text{H}_2\text{O}$  (16 mg/L),  $\text{MgCl}_2 + 6\text{H}_2\text{O}$  (14.6 mg/L),  $\text{CaCl}_2$  (13.5 mg/L),  $\text{FeCl}_3$  (4 mg/L),  $\text{Na}_2\text{MoO}_4 + 2\text{H}_2\text{O}$  (0.002 mg/L), and  $\text{MnSO}_4 + \text{H}_2\text{O}$  (0.002 mg/L) were also added (26).

HRT was gradually decreased during the MBR operation and at the same time, ERY concentration was increased until HRT and ERY concentration reached 6.0 h and 1000.0 mg/L, respectively. For routine operation, the membrane blockage was controlled by continuous monitoring of transmembrane pressure (TMP) and backwash was conducted every 60 secs for 10 secs. When the TMP reached 0.4 bar, the membrane module was cleaned by immersing the blocked membrane in  $\text{NaClO}$

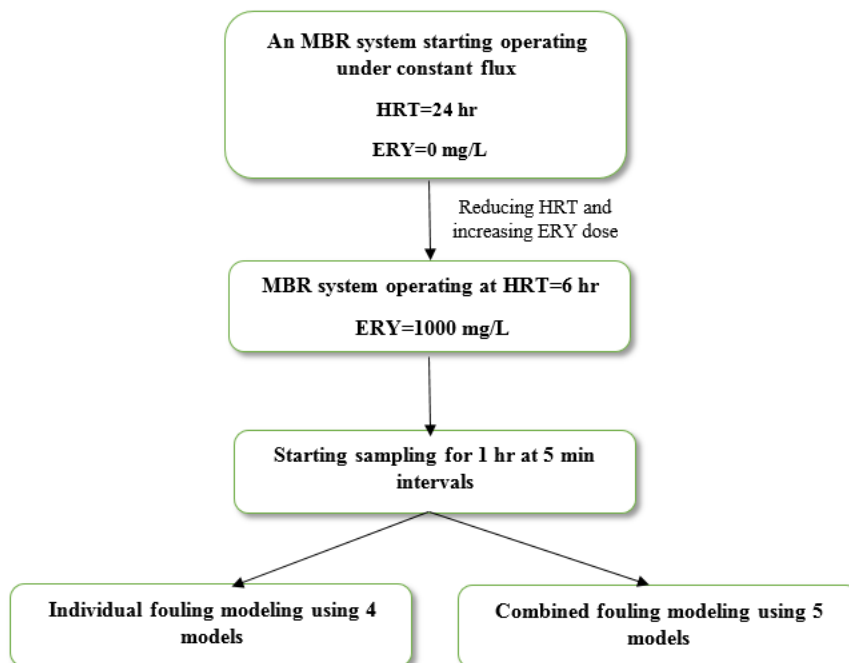
solution (1000.0 mg/L), soaking with citric acid (3 g/L) for 1 hour, and finally, washing with clean water. In order to find the fouling mechanisms and after achieving COD removal higher than 85%, the MBR operated at different MLSS concentrations ranging from 5.0 to 13.0 g/L under constant flux mode ( $5.0 \text{ L}/\text{m}^2 \cdot \text{h}$ ) and maximum suction lift of 500.0 mbar. For water sampling, the backwash pump was temporarily turned off and the filtered volume from MBR versus time was recorded using a graduated cylinder for 1 hour at 5-minute intervals. All the experiments were carried out in triplicates for any MLSS concentrations. During MBR operation, the dissolved oxygen concentration was supplied by the air compressor to reach as near as possible saturation. The flowchart of MBR system operation and modeling is illustrated in Figure 2.

### Fouling model

Based on the Darcy's law, in a membrane system, the flow rate ( $Q$ ) can be computed according to Eq. 1 (27).

$$Q = \frac{PA}{R\mu} \quad (1)$$

where,  $A$  and  $P$  are membrane area and TMP, respectively, and  $R$  and  $\mu$  are the resistance of membrane filtration and solution viscosity, respectively. In the present study, four individual models including standard blocking (Eq. 2), complete blocking (Eq. 3), intermediate blocking (Eq. 4), and cake filtration (Eq. 5) were employed to determine the mechanisms of membrane fouling. The mentioned models are derived from the Darcy's law under constant pressure (27). In the standard model, membranes have straight cylindrical pores that decline in radius as solid matter accumulates on the pore walls (28,29). The volume

**Figure 2.** Flow diagram of stages of MBR operation and modeling.

can be calculated as a function of time from Eq. (2):

$$V = \left( \frac{1}{J_0 t} + \frac{K_s}{2} \right)^{-1} \quad (2)$$

When a membrane fouls by the complete or intermediate blocking mechanism, a portion of the pores are unavailable for flow. The available membrane area declines with permeate volume in the complete model (29). For constant trans-membrane pressure operations, the equations can be inserted into the Darcy's law, Eq. (1), and integrated to derive equations for volume versus time. Eq. (3) describes complete blocking and Eq. (4) describes intermediate blocking (3).

$$V = \frac{1}{J_0 K_c} \left( \sqrt{1 + J_0^2 K_c t} - 1 \right) \quad (3)$$

$$V = \frac{J_0}{K_b} (1 - \exp(-K_b t)) \quad (4)$$

In the cake filtration model, the resistance to flow is increased by the presence of a cake layer on the membrane surface. The total resistance  $R$  will be the sum of the membrane resistance and the cake resistance. This can be calculated as a function of volume and time.

$$V = \frac{1}{K_i} (\ln(1 + K_i J_0 t)) \quad (5)$$

where  $V$  is filtered volume,  $J_0$  is the initial flux, and  $K_s$ ,  $K_c$ ,  $K_b$ , and  $K_i$  are standard blocking constant, cake filtration constant, complete blocking constant, and intermediate blocking constant, respectively.

In addition, in order to comprehensive understanding of the fouling mechanism under real conditions, five combined models (consisting of two individual models) namely cake-complete (Eq. 6), complete-standard (Eq. 7), cake-intermediate (Eq. 8), complete-intermediate (Eq. 9), and intermediate-standard (Eq. 10) were used (27). It must be noted that in each combined model, the dominant individual model is the one with a higher fitted parameter.

$$V = \frac{J_0}{K_b} \left( 1 - \exp \left[ \frac{-K_b}{K_c J_0^2 t} \left( \sqrt{1 + K_c J_0^2 t} - 1 \right) \right] \right) \quad (6)$$

$$V = \frac{J_0}{K_b} \left( 1 - \exp \left[ \frac{-2K_b t}{2 + K_c J_0 t} \right] \right) \quad (7)$$

$$V = \frac{1}{K_i} \left( \ln \left[ 1 + \frac{K_i}{K_c J_0} \left( \sqrt{1 + K_c J_0^2 t} - 1 \right) \right] \right) \quad (8)$$

$$V = \frac{\exp(K_i J_0 t) - 1}{K_i} - \frac{J_0}{K_b} (\ln(1 - K_b t)) \quad (9)$$

$$V = \frac{1}{K_i} \left( \ln \left[ 1 + \frac{2K_i J_0 t}{K_s J_0 t} \right] \right) \quad (10)$$

The fitness of the fouling models and experimental data

was done using nonlinear methods, which were evaluated using the Simplex method and the Levenberg–Marquardt algorithm using the fitting facilities of the Microcal Origin 2018 software. The suitability of the fouling models was evaluated using the adjusted determination coefficient ( $R^2_{adj}$ ). Eqs. (11) and (12) are the mathematical expressions for  $R^2$  and  $R^2_{adj}$  respectively (30).

$$R^2 = \left( \frac{\sum_i^n (V_{i,exp} - \bar{V}_{exp})^2 - \sum_i^n (V_{i,exp} - V_{i,model})^2}{\sum_i^n (V_{i,exp} - \bar{V}_{exp})^2} \right) \quad (11)$$

$$R^2_{adj} = 1 - \left( 1 - R^2 \right) \cdot \left( \frac{n-1}{n-p-1} \right) \quad (12)$$

where,  $V_{i,model}$  is the individual theoretical  $V$  value predicted by the model,  $V_{i,exp}$  is individual experimental  $V$  value,  $\bar{V}_{exp}$  is the average of all experimental  $V$  values measured,  $n$  is the number of experiments, and  $p$  is the number of parameters in the fitting model.

## Results

### Variation of filtrated volume versus time

MBR system was operated under different MLSS concentrations ranging from 5.0 to 13.0 g/L under a constant flux of 5.0 L/m<sup>2</sup>.h. Variations of extracted solution from the MBR system was volumetrically measured and the results are shown in Figure 3.

As illustrated, when the concentration of MLSS in aeration tank increased from 5.0 g/L to 13.0 g/L, the filtrated volume from the MBR system depleted from 744 ± 15 mL to 403 ± 20 ml after 60 minutes. This behavior is mainly related to MLSS accumulation in aeration tanks and the high production of EPS and SMP (31). In addition, with progressing filtration time, the volume of the filtration decreased. Overall, around 60% of filtration

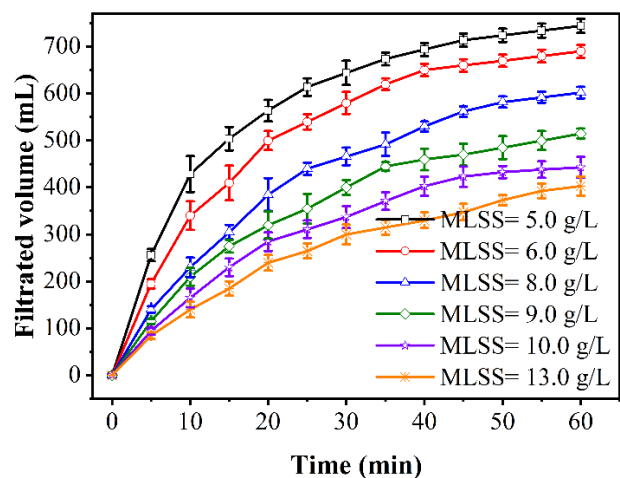


Figure 3. Variation of filtrated volume versus time during MBR operation under different MLSS concentrations.

volume was collected in 25 min due to pore blocking (32,33).

### Prediction using the individual fouling models

As previously stated, in order to determine the dominant mechanisms in the fouling of the membrane module, the experimental data of filtrated volume from the MBR system were fitted with four individual models and displayed in Figure 4. In addition, the parameters of individual fouling models under different MLSS concentrations are presented in Table 2.

As can be seen in Figure 4 and summarized in Table 2, for all studied MLSS concentrations, the values of the  $R^2_{adj}$  parameter ranged from 0.914 to 0.999, indicating that the individual fouling models appropriately fitted with

experimental data. In overall, the standard model indicated the best performance for all studied MLSS concentrations ( $R^2_{adj} > 0.997$ ). In addition, the lowest fitting performance was observed for the cake model when the MBR system operated under the MLSS concentration of 5.0 g/L. Other individual fouling models including the standard model, complete model, and intermediate model have shown well-fitting performance.

### Prediction using the combined models

The experimental data under five different MLSS concentrations (5.0-13.0 g/L) was further examined with combined fouling models namely cake-complete, complete-standard, cake-intermediate, complete-intermediate, and intermediate-standard. Figure 5

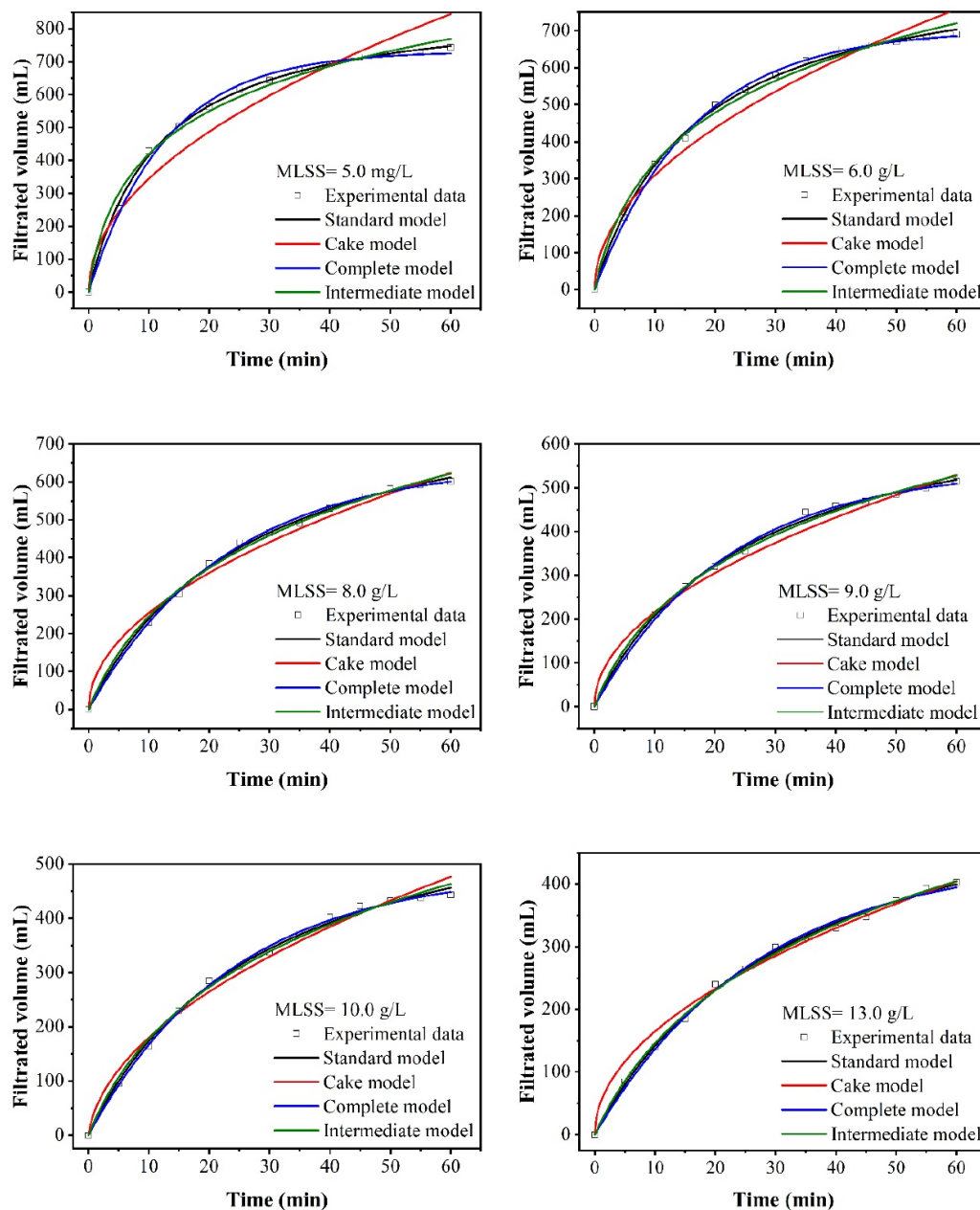


Figure 4. Fitted individual fouling models under different MLSS concentrations.

represents the fitted plots of experimental data with different combined fouling models, and the estimated parameters are summarized in Table 3.

As presented in Table 3, for all studied MLSS concentrations, the values of  $R^2_{adj}$  were higher than 0.986, indicating good fitting of combined fouling models with experimental data. Compared to individual fouling models (Table 2), the combined fouling models showed better prediction of filtrated volume from the MBR system under various MLSS concentrations. Overall, the cake-

intermediate model showed the lowest fitness (based on the  $R^2_{adj}$ ) in studied MLSS concentrations. On the other hand, cake-complete and complete-standard models were the most successful models in filtrated volume prediction in comparison with other combined fouling models.

### Discussion

The present study was carried out to evaluate the performance of a lab-scale MBR system for the treatment of wastewater containing ERY. The MBR system was

**Table 2.** Estimated parameters of individual fouling models

Models	Parameter	MLSS Concentration (g/L)					
		5.0	6.0	8.0	9.0	10.0	13.0
Standard	$J_0$	77.81	53.80	32.72	28.89	23.30	18.36
	$K_s$	$2.24 \times 10^{-3}$	$2.22 \times 10^{-3}$	$2.25 \times 10^{-3}$	$2.70 \times 10^{-3}$	$2.95 \times 10^{-3}$	$3.19 \times 10^{-3}$
	$R^2_{adj}$	0.999	0.998	0.998	0.997	0.997	0.998
Cake	$J_0$	138.99	144.04	38.89	87.59	15.61	7.96
	$K_c$	$8.41 \times 10^{-5}$	$1.04 \times 10^{-4}$	$1.54 \times 10^{-4}$	$2.14 \times 10^{-4}$	$2.37 \times 10^{-4}$	$3.67 \times 10^{-4}$
	$R^2_{adj}$	0.914	0.958	0.983	0.981	0.983	0.987
Complete	$J_0$	57.65	43.04	28.34	24.79	20.42	16.33
	$K_b$	$7.88 \times 10^{-2}$	$6.11 \times 10^{-2}$	$4.37 \times 10^{-2}$	$4.55 \times 10^{-2}$	$4.18 \times 10^{-2}$	$3.68 \times 10^{-2}$
	$R^2_{adj}$	0.995	0.998	0.998	0.998	0.997	0.996
Intermediate	$J_0$	133.82	76.15	39.79	35.66	27.77	21.38
	$K_i$	$4.77 \times 10^{-3}$	$4.16 \times 10^{-3}$	$3.66 \times 10^{-3}$	$4.47 \times 10^{-3}$	$4.70 \times 10^{-3}$	$4.91 \times 10^{-3}$
	$R^2_{adj}$	0.992	0.991	0.996	0.995	0.994	0.998

**Table 3.** Estimated parameters of combined fouling models

Models	Parameter	MLSS Concentration (g/L)					
		5.0	6.0	8.0	9.0	10.0	13.0
Cake-complete	$J_0$	75.86	47.17	30.42	26.67	20.51	19.76
	$K_c$	$1.38 \times 10^{-4}$	$8.14 \times 10^{-6}$	$1.52 \times 10^{-5}$	$2.33 \times 10^{-5}$	$1.72 \times 10^{-6}$	$1.26 \times 10^{-4}$
	$K_b$	$9.51 \times 10^{-1}$	$6.37 \times 10^{-2}$	$4.31 \times 10^{-2}$	$4.52 \times 10^{-2}$	$4.17 \times 10^{-2}$	$3.23 \times 10^{-2}$
	$R^2_{adj}$	0.999	0.999	0.998	0.997	0.998	0.998
Complete-standard	$J_0$	60.56	46.59	30.22	26.69	20.49	18.36
	$K_b$	$5.39 \times 10^{-1}$	$6.18 \times 10^{-2}$	$4.10 \times 10^{-2}$	$4.25 \times 10^{-2}$	$4.17 \times 10^{-2}$	$1.33 \times 10^{-4}$
	$K_s$	$8.22 \times 10^{-3}$	$3.37 \times 10^{-4}$	$4.65 \times 10^{-4}$	$6.14 \times 10^{-4}$	$3.04 \times 10^{-5}$	$3.20 \times 10^{-3}$
	$R^2_{adj}$	0.999	0.999	0.998	0.998	0.998	0.998
Cake-intermediate	$J_0$	11.04	4.09	3.35	3.23	2.97	2.74
	$K_c$	$2.16 \times 10^{-2}$	$1.58 \times 10^{-3}$	$1.52 \times 10^{-3}$	$1.73 \times 10^{-3}$	$1.79 \times 10^{-3}$	$2.01 \times 10^{-3}$
	$K_i$	$2.89 \times 10^{-3}$	$3.15 \times 10^{-3}$	$2.77 \times 10^{-3}$	$3.38 \times 10^{-3}$	$3.56 \times 10^{-3}$	$3.65 \times 10^{-3}$
	$R^2_{adj}$	0.986	0.992	0.996	0.996	0.994	0.998
Complete-intermediate	$J_0$	57.65	43.04	28.34	15.08	11.79	10.26
	$K_b$	3.28	3.28	3.28	0.192	0.125	0.222
	$K_i$	$1.37 \times 10^{-3}$	$1.42 \times 10^{-3}$	$1.54 \times 10^{-3}$	$2.93 \times 10^{-3}$	$3.66 \times 10^{-3}$	$3.08 \times 10^{-3}$
	$R^2_{adj}$	0.994	0.998	0.998	0.998	0.997	0.997
Intermediate-standard	$J_0$	77.81	53.80	32.72	28.89	23.31	19.58
	$K_s$	$2.24 \times 10^{-3}$	$2.21 \times 10^{-3}$	$2.24 \times 10^{-3}$	$2.66 \times 10^{-3}$	$2.95 \times 10^{-3}$	$7.79 \times 10^{-1}$
	$K_i$	$5.18 \times 10^{-6}$	$1.03 \times 10^{-5}$	$1.46 \times 10^{-5}$	$1.89 \times 10^{-5}$	$1.03 \times 10^{-6}$	$3.15 \times 10^{-3}$
	$R^2_{adj}$	0.999	0.997	0.998	0.998	0.996	0.998

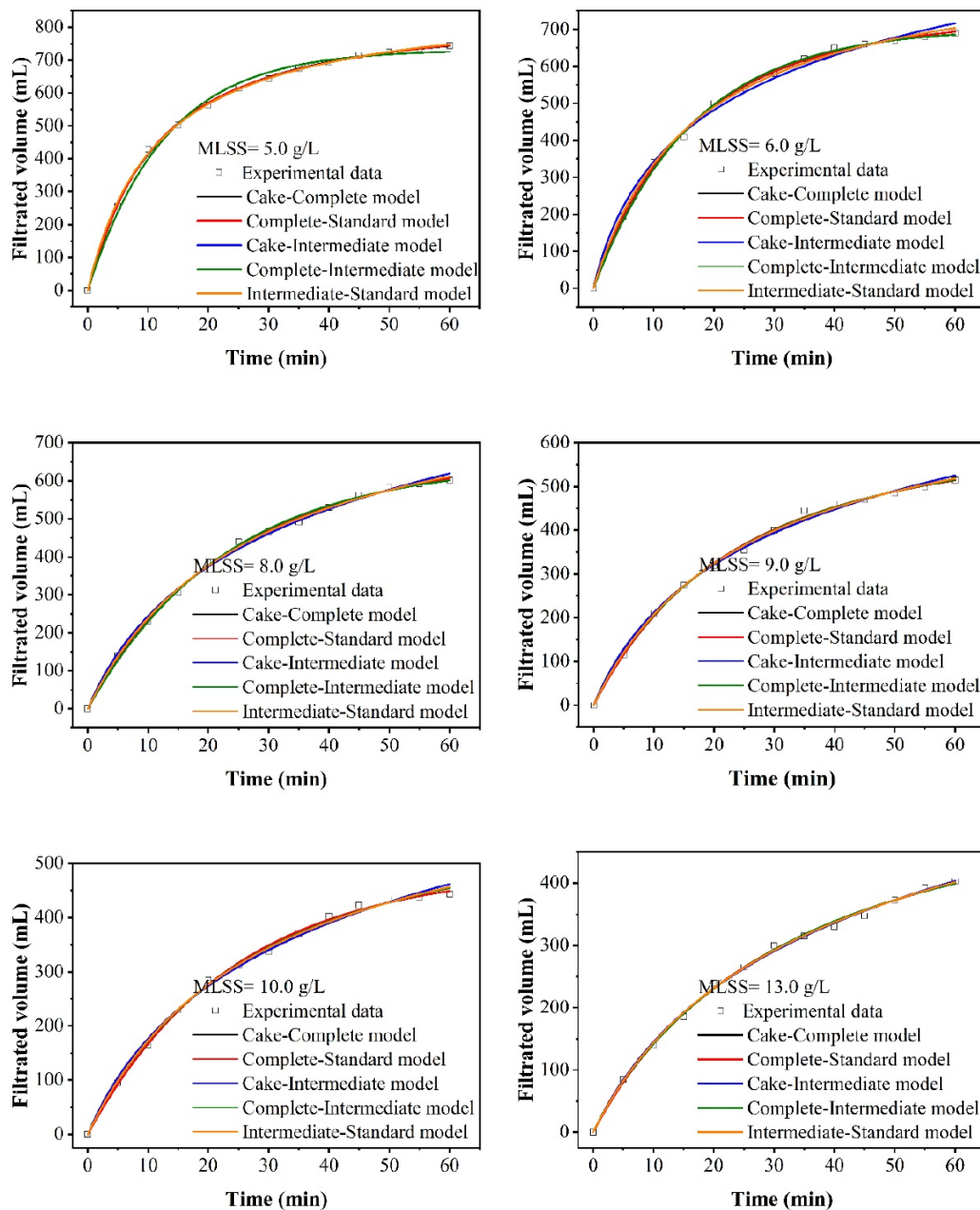


Figure 5. Fitted combined fouling models under different MLSS concentrations.

operated under different MLSS concentrations (5.0-13.0 g/L) under a constant flow rate. The experimental data on filtrated volume versus time was recorded and evaluated with different individual and combined fouling models to determine the mechanisms of membrane fouling.

Based on the obtained results, the individual fouling models showed an  $R^2_{adj}$  value higher than 0.914, indicating that the individual models were capable of predicting permeation of the MBR system with relatively high accuracy without considering the MLSS concentration. In addition, from individual models, the standard model indicated the best performance for the predication of permeate of the MBR system for all of the studied MLSS concentrations. On the contrary, Hu et al (34) operated a lab-scale MBR system in the absence of ERY under

four different SRTs and investigated the membrane fouling mechanisms of an MBR system by individual and combined models derived from the Darcy's law. They reported that under constant pressure, the complete and standard blocking models showed the lowest fitting performance than the intermediate and cake models. In addition, they found that that the characteristics of MLSS don't significantly affect the fitness of models.

Also, Palani et al (35) studied a lab-scale MBR conducted under the constant current density for pharmaceutical wastewater from Alathur Industrial Estate, Chennai. In this study, the propensity of fouling was evaluated by Hermia models. In accordance with this study, the standard blocking model was very well fitted with the standard error of 0.00882, which also reconfirmed the

lesser internal fouling in IMBBR.

As reported in Tables 2 and 3, the initial membrane flux ( $J_0$ ) was significantly decreased as a function of MLSS concentration increased. Based on the standard model, the  $J_0$  reduced from 77.81 m/s to 18.36 m/s as the MLSS concentration in the aeration tank increased from 5.0 to 13.0 g/L. Iorhemen et al (36) indicated that higher MLSS concentration leads to diminish membrane permeability, and also, enhances membrane fouling. This behavior is may be related to the decrease in the amount of EPS in MLSS, the increase in the ratio of protein to polysaccharide in bounded EPS, the worsening of the sedimentation properties of sludge, the increase in hydrophobicity of its, and finally, the decrease in membrane flux (37,38).

The results of combined fouling modes indicated that all models well fitted with the experimental data compared to individual fouling models (Tables 2 and 3). However, cake-complete and complete-standard models were the most suitable model for the prediction of permeate of the MBR system. Hu et al (8) used the combined models for the prediction of fouling phenomena of an MBR system during the treatment of synthetic wastewater. They found that the combined models have a better fitting performance than the individual models. From the combined models, the cake-standard, intermediate-standard, and cake-intermediate models provided the highest capability for the prediction of experimental data at the shortest SRT. For the longest SRT, the cake-standard model provided slightly better performance for the data prediction than other combined models. Based on the fitness performance, the cake-complete model presents a good ability for permeate prediction, and flux declined in a manner between the extremes of cake filtration and complete blocking (27). In addition, Hu et al (34) indicated that the combined cake-intermediate and intermediate-standard models were more effective in describing the experimental data. The difference between the results of the present study and previous studies may be attributed to different experimental conditions (ERY presence, MLSS concentrations, SRT, applied pressure, etc), properties of the membrane, and also, characteristics of the feed wastewater (39,40).

Fakhri et al (41) studied the removal of some antibiotics such as ERY from wastewater by the MBR system, and also, investigated the membrane biofouling. They showed that compared with the control system, the MBR system containing ERY reached a higher TMP. This condition may be related to antibiotics stressing the microorganisms in the MBR system, resulting in more EPS/SMP secretion. Another study has also reported that the presence of pharmaceutical compounds in influent wastewater leads to a higher production rate of EPS and results in higher fouling rates. The fouling rate is in proportion to the concentration of pharmaceuticals. Pharmaceuticals alter the composition of the foulants and affect microbial

metabolism, thereby inflicting direct/indirect effects on fouling. Also, these compounds have been found to alter the size of sludge flocs, with the unknown exact mechanism for the floc size change (6,22).

For this reason, this study was conducted since the membrane fouling mechanisms in MBR systems treating antibiotics can be different from those treating conventional pollutants (6). The results of the present study showed differences in comparison with other mentioned studies. One major reason could be the nature of wastewater; as mentioned before, antibiotics stimulate the production of EPS and SMP (42).

Some studies used the activated sludge model (ASM) and its modifications to a fully-fledged membrane fouling model. In some studies, ASM1 integrated with the fouling model with a modified ASM1-based model were used in the lack of the influence of SMP on the irreversible fouling of the membrane and connection between both of them (39,43-45). Also, in Janus study, an integrated mathematical model of MBR, in which the ASM, membrane fouling, and air scouring were considered, was used. The effects of membrane cleaning on membrane fouling and resistance have not been considered in this model. Originally, integrated mathematical models for MBR systems can simulate processes and aid the design and operation of full-scale membrane bioreactors (46,47).

Although the results of this study could be applied in the prediction of the fouling mechanisms of full-scale MBR systems, some limitations should be mentioned. First, the MBR system was operated only under one TMP and future studies could vary the TMP to increase our knowledge. Secondly, in contrast with artificial intelligence (AI)-based technologies, the effect of other possible influencing factors on membrane fouling (as inputs of AI algorithms) including operating conditions, water and wastewater quality parameters, membrane & biomass properties, DO concentration, and mixing intensity, was not evaluated in the present study (48).

## Conclusion

In the present study, the fouling mechanisms of the MBR systems for the treatment of wastewater containing ERY were investigated. The MBR system was operated under several MLSS concentrations to evaluate the effect of mixed liquor on membrane fouling. The membrane fouling was assessed using the individual and combined fouling models based on the Darcy's law. After experimental data modeling with individual modeling, most of the models were capable of appropriately predicting permeate of the MBR system. However, combined models showed better fitting performance than individual models. The standard model, and also, cake-complete and complete-standard models are the best individual and combined models, respectively, for experimental data prediction. This study indicated that mechanistic models are suitable



for the fouling prediction of MBR systems under a wide range of MLSS concentrations. However, there are still some areas that future studies can focus on evaluating the effect of varying TPM, different membrane properties, treating a complex of antibiotics, etc. Mechanistic models based on operating parameters such as membrane flux have been developed to evaluate the membrane fouling behaviors in membrane treatment systems. However, due to the complexity of this phenomena, it is rather challenging to accurately predict the occurrence or evolution of membrane fouling using these classical mathematical models. Therefore, it is very necessary to develop more effective approaches that can overcome the shortcomings of traditional models and accurately predict the performance of membrane processes (48).

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### Competing interests

The authors declare that they have no conflict of interests.

### Ethical issues

The study protocols were approved by the Ethics Committee of Isfahan University of Medical Sciences, Isfahan, Iran (Ethical code: IR.MUI.RESEARCH.REC.1397.300).

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