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Role of zeolite in reducing membrane fouling in a hybrid membrane bioreactor system applied for wastewater treatment

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ABSTRACT

This study investigates the effectiveness of zeolite for controlling the membrane fouling in a hybrid membrane bioreactor. A control membrane bioreactor without zeolite (CMBR) and a hybrid MBR with zeolite were studied, which were called 4 g L^{-1} zeolite (Z4-MBR), 8 g L^{-1} zeolite (Z8-MBR) and 12 g L^{-1} zeolite (Z12-MBR) within long- and short-term filtration experiments. On average, the Z4-MBR, Z8-MBR and Z12-MBR reduced soluble microbial products (SMP_C) by 18.96%, 42.11% and 19.44%, respectively. Furthermore, the ZMBR systems developed lower operational trans-membrane pressure (TMP), lower zeta potential, increased the relative hydrophilicity of sludge and enhanced the concentration of EPS. Reduced membrane fouling and ease of membrane operation are concluded from this study.

Keywords: Membrane bioreactor; Zeolite; Trans-membrane pressure; Soluble microbial products; Zeta potential

1. Introduction

Membrane bioreactor (MBR) processes combine biological wastewater treatment with micro/ultrafiltration process to treat wastewater biologically and to separate biomass physically from mixed liquor in an integrated configuration. MBR technology features various distinct advantages over the conventional activated sludge process, including excellent effluent quality, good disinfection capability, higher volumetric loading, reduced footprint and sludge production, process flexibility towards influent changes, and improved nitrification. Over the past decades, more attention has been paid to post-treatment of anaerobic digestion effluent using different strategies since the effluent is considered as a resource for water production in some regions [1]. Membrane filtration has been considered a promising solution to treat anaerobic effluent to meet the increasingly strict discharge standards, because membranes could remove physical, chemical and microbiological contaminants [2]. Notwithstanding the significant progress of MBR technology, membrane fouling remains the primary hindrance for its universal and large scale applications. Membrane fouling would reduce system productivity. It also increases the energy required for gas scouring and frequency of cleaning, which might shorten the membrane lifespan, resulting in higher replacement costs. Membrane fouling in MBRs is a result of the interactions between the sludge suspension and membrane unit [3–5]. Combination of supplementary processes such as adsorption or coagulation with MBR can reduce the membrane fouling. PAC, zeolite, chitosan and polymeric coagulants have been used as adsorbents/coagulants in wastewater plants for enhancing effluent quality or reducing membrane fouling [6,7]. Zeolite consists of smectite minerals and functions as cation exchanger and coagulant aid, thanks to its thixotropy, permeability and viscosity properties [6]. Both natural and synthetic zeolites have the ability to remove some cations from solutions by adsorption and cation exchange processes [8]. Clinoptilolite, a type of zeolite, enjoys a higher storage capacity, lower price and strong adsorbabil-

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ity [11]. Rezaei et al. studied the effect of clinoptilolite on the performance of MBR [10–11]. The results indicated that addition of zeolite improved ammonium removal by 24%. Damayanti et al. studied PAC, zeolite and *Moringa oleifera* as biofouling reducers in MBR [6]. The results showed that addition of zeolite to activated sludge would increase permeability and decrease membrane resistance. He et al. compared nutrient removal, filtration characteristic, microorganism activity, and permeability of membranes between control MBR and the MBR with zeolite [12].

Although zeolite is well known in wastewater industry and many researchers have studied clinoptilolite in activated sludge, fewer pieces of research have studied MBR in combination with zeolite comprehensively, bearing in mind that clinoptilolite effects on membrane fouling in MBR are not clear. Thus, the main objective of this work is to investigate a hybrid MBR system which reduces membrane fouling substantially and to determine the effect of clinoptilolite, which is very cheap and available all over the world, as a bio-fouling reducer in the MBR. For further reduction of bio-fouling, different amounts of zeolite (4 g L^{-1}, 8 g L^{-1} and 12 g L^{-1}) were used as a bio-fouling reducer to be examined on membrane fouling in MBR. Then, COD, MLSS, EPS, soluble microbial products (SMP), trans-membrane pressure (TMP), SVI, zeta potential and relative hydrophobicity (RH), particle size distribution (PSD), scanning electronic microscopy (SEM), zone settling velocity (ZSV) were assessed as well in both the control membrane bioreactor (CMBR) and zeolite membrane bioreactor (ZMBR), which operated under similar conditions.

2. Materials and methods

2.1. Experimental setup

The laboratory MBR set-up (Fig. 1) consisted of a Plexiglas reactor divided into two similar bioreactors working in parallel. In both sides, a membrane module was submerged in the middle. The bioreactors were cube shaped with a total volume of 16 L and working volume of 15 L. The membrane modules used in this study were Kubota polyethylene flat microfiltration type with a pore size of

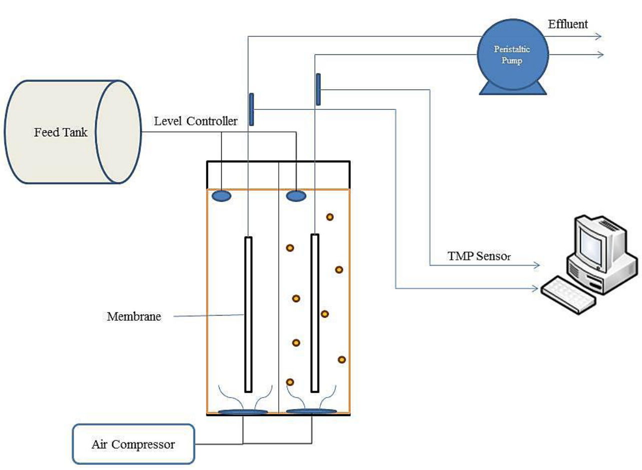

Fig. 1. Schematic diagram of the membrane bioreactor system.

0.4 μm and filtration area of 0.11 m^2. A peristaltic pump was used for both of the permeate flows. In case of any TMP augmentation more than the maximum allowable TMP, the membranes were cleaned physically. The permeated effluent had a constant flow rate of 31.25 (mL/ min) to provide 8 h hydraulic retention time (HRT) in both reactors. An air diffuser was installed in the bottom of each reactor to supply air and mix the microorganisms. The temperature was the same as the laboratory temperature (17–25°C).

2.2. Feed medium

The feed tank was installed above the bioreactors with a volume of 100 L. The influent content was fed continuously into the bioreactors as shown in Table 1. The feed flow rate was controlled by a level controller. Activated sludge was fed for 60d with COD: 500–1000 for adaptation of the microorganisms before starting the runs.

2.3. Analysis

The composition of COD, MLSS, TMP, SVI, PSD (CA-SP3 analyzer, Japan), relative hydrophobicity, and zeta potential (Brookhaven, USA) contents were determined according to Standard Methods [13]. In general, a two-step extraction method was adopted to extract SMP and EPS from the sludge sample. The method was inspired to the heating EPS extraction procedures [14]. In order to measure the protein and carbohydrate contents, the extracted bound EPS and SMP were analyzed using Lowry's and Antrone's methods [15], respectively. Surface morphology of the zeolite was analyzed by SEM (Hitachi, Model: s4160, Japan). DO concentration, pH and temperature were measured using a multimeter sensor (WTW-Multi 340i, Germany).

2.4. Preparation and characterization of zeolite

Zeolite samples used in this study were taken from Semnan mines, Iran. The samples were ground and sieved to a size of 75–90 μm and then washed with distilled water several times to remove any non-adhesive impurities and small particles. Then, it was shaken with distilled water in a shaker for 24 h to remove any remaining fine impurities, and thereafter was dried at 105°C in an oven for 24 h. The chemical composition of zeolite samples was determined using Philips PW1730 X-ray, as presented in Table 2.

Table 1
Composition of the synthetic wastewater

Parameter	Amount
$C_6H_{12}O_6$, mg L^{-1}	469.045
COD, mg L^{-1}	500
$(NH_4)_2HPO_4$, mg L^{-1}	21.29
NH_4NO_3, mg L^{-1}	58.525
Total nitrogen(TN), mg L^{-1}	25
Total phosphor (TP), mg L^{-1}	5

Table 2
Zeolite composition and physical characteristic

Sample composition	% (wt)
SiO_2	68.95
Al_2O_3	11.14
Fe_2O_3	0.97
CaO	4.83
Na_2O	0.95
K_2O	0.9
MgO	0.79
TiO_2	0.201
MnO	0.011
P_2O_5	0.012
SO_3	0.068
Loss on ignition	11.178
Physical characteristic	
Density, kg/m^3	2.2
Surface area, m^2/g	40
Pore volume cm^3/g	0.1456
Mean pore diameter, nm	22.033

3. Results and discussion

3.1. The effect of zeolite on COD

The wastewater COD ''hardness'' is based on biodegradable and non-biodegradable elements contained in the wastewater. High wastewater COD ratio indicates rapid biodegradability, while low COD ratio indicates slow biodegradability or existence of non-biodegradable elements. Slowbiodegradability is caused by diverse unknown constituents in industrial wastewater leading to fouling problems [16,17]. Fig. 2 shows COD concentration in MBR and Z4-MBR: both CMBR (91.68%) and Z4-MBR (90.96%) also presented the same performance in terms of COD removal. Therefore, zeolite does not affect COD removal significantly. Other researchers have confirmed that zeolite does not have any potential to enhance COD elimination [10].

3.2. The effect of zeolite on MLSS

Mixed liquor suspended solids (MLSS) represent the concentration of solids (cells and solid particles) in the mixture of sludge. Higher MLSS results in better performance especially for strong wastewater. One of the treatment problems with MBR is that the increased MLSS accelerates the membrane fouling due to high suspended solids [18,19]. Long biomass adaptation is needed to degrade complex pollutants in high strength industrial wastewater and achieve high quality effluent [17]. Fig. 2 demonstrates MLSS during the experiment. Both CMBR and ZMBR have an average initial MLSS of 7.62 g L^{-1} in startup with no sludge withdrawn during the experiment (SRT = ∞). As shown in Fig. 2, MLSS in Z4-MBR, Z8-MBR and Z12-MBR, has increased by 4.8%, 15.8% and 17%, respectively in comparison with CMBR at the end of experiments. There are two main reasons resulting in elevation of MLSS in ZMBR.

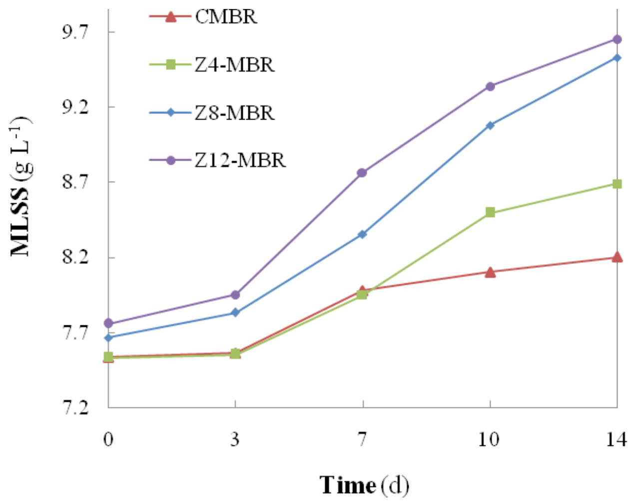

Fig. 2. MLSS concentration versus time during experiments. (Membrane bioreactor without zeolite called CMBR and the hybrid MBR with zeolite, namely 4 g L^{-1} zeolite (Z4-MBR), 8 g L^{-1} zeolite (Z8-MBR) and 12 g L^{-1} zeolite (Z12-MBR)).

Firstly, the cation exchange of zeolite with activated sludge results in ammonium adsorption, where consequently zeolite changes into a rich source of substrate for microorganisms to feed on and grow. Secondly, with the increase in the population of nitrifiers (due to easier digestion of ammonium in the fluid's bulk),the biofilm preference to develop on zeolite particles grows, thereby increasing the MLSS. So the presence of zeolite particles with respect to the density of ammonium causes increased nitrifiers population [11].

3.3. The effect of zeolite on EPS/SMP

EPS is a complex mixture of macromolecular polyelectrolytes (i.e. polysaccharides, protein, nucleic acids, and humic compounds) contributing to the formation of microbial aggregates, consisting of insoluble materials (sheaths, capsular polymers, condensed gel, loosely-bound polymers, and attached organic material). On the other hand, SMP is considered as a soluble part of EPS (i.e. soluble macro-molecules, colloids, and slimes) released into the solution from substrate metabolism and biomass decay [20,21]. The influence of microorganism metabolites, such as SMP and EPS, on membrane fouling is essential to understanding and controlling membrane fouling [22]. Higher SMP concentrations in the solution lead to higher membrane fouling rates, due to higher pore blocking. It also reduces the cake porosity by filling the void spaces between the cell particles in the cake layer [23]. Higher EPS concentration in the sludge enhances bioflocculation, resulting in the development of larger and more permeable flocs, which tends to reduce membrane fouling [22]. Fig. 3 illustrates the average and the details of carbohydrate and protein concentrations of SMP and EPS, respectively in the bioreactor over the experiment period. These results confirm that the concentration of SMP (both SMP_C and SMP_P) in bulk solution, which is regarded as soluble cellular components released during cell lysis, decreased in ZMBR compared to CMBR, with reduction of

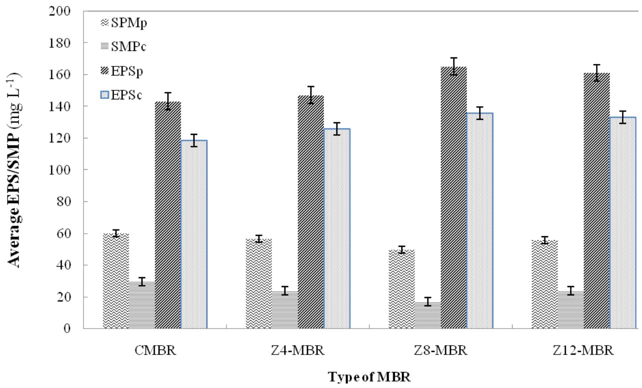

Fig. 3. Concentration of supernatant SMP and EPS in different MBRs. (Membrane bioreactor without zeolite called CMBR and the hybrid MBR with zeolite, namely 4 g L^{-1} zeolite (Z4-MBR), 8 g L^{-1} zeolite (Z8-MBR) and 12 g L^{-1} zeolite (Z12-MBR)).

SMP_C being more significant. This could be due to adsorption process [24], and/or attachment onto the membrane surface as a bio-layer [25]. The average concentration of SMP_C in the bulk solution after the addition of zeolite has decreased by 18.96%, 42.11% and 19.44%, in Z4-MBR, Z8-MBR and Z12-MBR respectively when compared to SMP_C in CMBR. The Z4-MBR, Z8-MBR, Z12-MBR show a potential to reduce the protein portion of SMP by 5.68%, 17.45% and 6.94%, respectively. The increase in EPS (both EPS_C and EPS_P) on ZMBR, compared to CMBR, indicates that systems with zeolite produce flocs that have better structure, and/or formation. Typical SEM images of zeolite obtained before and after experiments are presented in Fig. 4. As mentioned previously, EPS is a complex mixture of macromolecular polyelectrolytes participating in the formation of microbial aggregates. Formation of aggregate is confirmed by lower rise of TMP in Fig. 5. The concentration of EPS_C in Z8-MBR (which has increased by 14.57%)

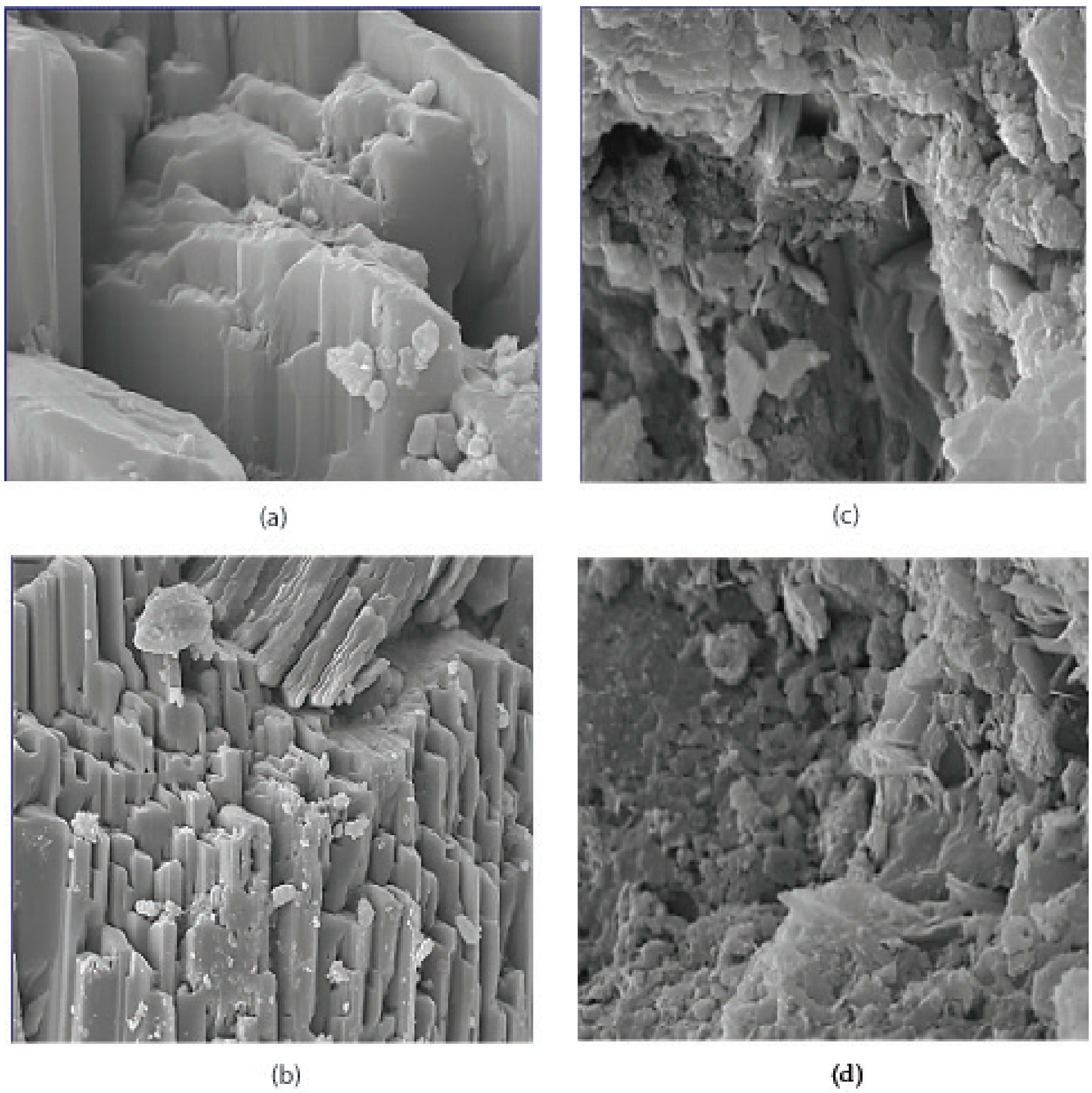

Fig. 4. The SEM images from surface of zeolite before (a,b) and after (c,d) filtration experiments a) 3 µm b) 5 µm c) 3 µm d) 5 µm.

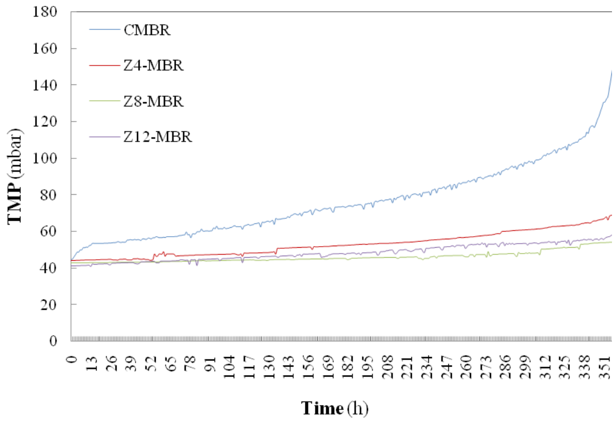

Fig. 5. Trans-membrane pressure versus time during experiment in MBRs. (Membrane bioreactor without zeolite called CMBR and the hybrid MBR with zeolite, namely 4 g L^{-1} zeolite (Z4-MBR), 8 g L^{-1} zeolite (Z8-MBR) and 12 g L^{-1} zeolite (Z12-MBR)).

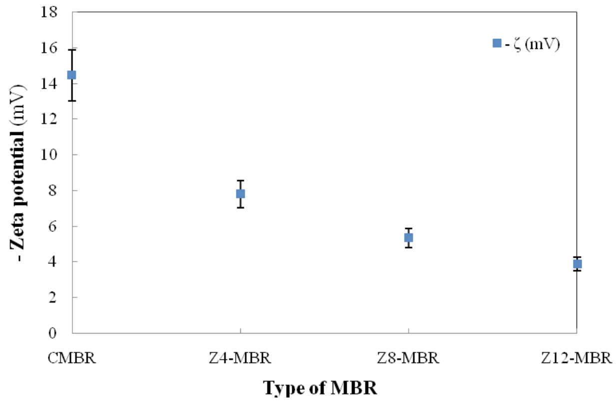

Fig. 6. Zeta potential with different doses of zeolite. (Membrane bioreactor without zeolite called CMBR and the hybrid MBR with zeolite, namely 4 g L^{-1} zeolite (Z4-MBR), 8 g L^{-1} zeolite (Z8-MBR) and 12 g L^{-1} zeolite (Z12-MBR)).

indicates that this system has a higher possibility to form bigger and better floc structures. The addition of zeolite with adsorption capacity to the MBR system changes the biomass flocs size and its behavior to attract more soluble organic matters to attach or stay in the bulk solution [26,27]. Due to the ion exchange property of zeolite and increasing the ion strength mixture by adding zeolite in ZMBRs, the intercellular water would apparently decrease and this cause reducing in chemical potential of cake layer and finally lead to decreasing in membrane fouling [28].

3.4. The effect of zeolite on zeta potential

Zeta potential is the charge that develops at the interface between a solid surface and its liquid medium. This potential may arise by the dissociation of ion-genic groups in the particle surface and the differential adsorption of solution ions into the surface region. Increasing the concentration of polymeric compounds and SMP which causes growth of the filamentous bacteria, results in elevated zeta potential of the sludge and then increased membrane fouling. As shown in Fig. 6, zeta potential declines by adding zeolite, causing diminished membrane fouling. The main reason is that when zeta potential approaches zero, particles tend to aggregate and the resistance grows thus playing a significant role in membrane fouling. Another study has also indicated this state previously [29].

3.5. The effect ofzeolite on TMP

TMP is regarded as trans-membrane pressure which is the pressure difference between the two sides of the membrane. It is used as a criterion to represent the amount of fouling in a membrane. As shown in Fig. 5 the mean TMP within 360 h in CMBR, Z4-MBR, Z8-MBR, and Z12-MBR is 77.36, 53.08, 45.99 and 48.63 mbar respectively indicating 31.38%, 40.54%, 37.14% TMP reduction (compared to CMBR) respectively. One of the most important reasons behind TMP reduction and in turn membrane fouling reduction in ZMBR is SMP drop especially for polysaccharide type of SMP [10]. Particle size growth, discussed in Section 3.7, can be regarded as another reason behind TMP reduction and membrane fouling due to bio-film growth on zeolite particles as carriers. Development of particle size may reduce the probability of the particle to pass through membrane pores and also enhanced particle movement from the membrane surface to the bulk solution. On the other hand, SVI enhancement (as in Section 3.6) diminishes TMP and fouling and also improves the performance of MBR (owing to the modification of sludge properties).

3.6. The effect of zeolite on SVI

Sludge volume index (SVI) shows the tendency of sedimentation and flocculation inactivated sludge. It was calculated after 30-min sludge settlement. Settlement of the sludge is generally caused by two main factors: growth of filamentous organisms and bounding water to bacterial cells that absorb water. Non-filamentous microorganisms orviscous bulking is most likely related to the morphological characteristics of the flocs and the presence of large amounts of extracellular slime. EPS has also been considered as an essential contributor to bio-flocculation and thus SVI value. The main parameters influencing SVI are EPS and the floc size. As shown in Fig. 7, the value of average SVI (between the 10^{th} and 15^{th} day) in the bulk solution has decreased by 14.63%, 58.14% and 48.73%, in Z4-MBR, Z8-MBR and Z12-MBR respectively in comparison to CMBR after the addition of zeolite. Zeolite addition to activated sludge reduces SMP, increases biofilm growth on particles and results in greater tendency to settling. The main reason for the reduction of SVI by adding zeolite is entrapment of zeolite in the middle of microorganisms especially nitrifiers, which prefer attached growth, after which sludge granules become heavy leading to enhanced settlement. Other studies have also reported SVI enhancement with addition of zeolite [10].

3.7. The effect of Zeolite on PSD

Fig. 9 demonstrates PSD in the15^{th} day of the experiment. With regard to zeolite size in the range of 75–90 µm

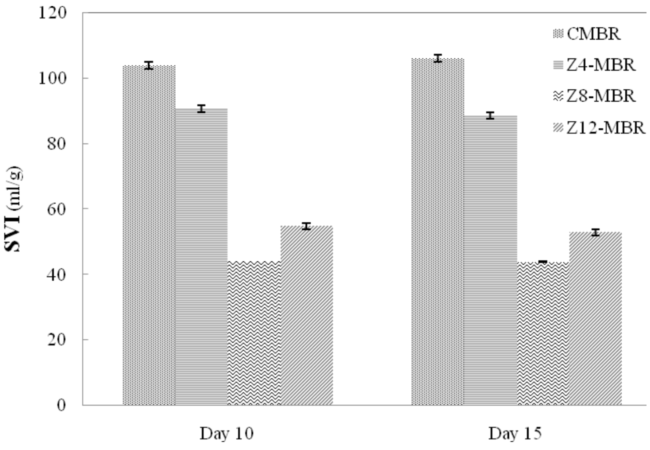

Fig. 7. The sludge volume index during the complete experiment. (Membrane bioreactor without zeolite called CMBR and the hybrid MBR with zeolite, namely 4 g L^{-1} zeolite (Z4-MBR), 8 g L^{-1} zeolite (Z8-MBR) and 12 g L^{-1} zeolite (Z12-MBR)).

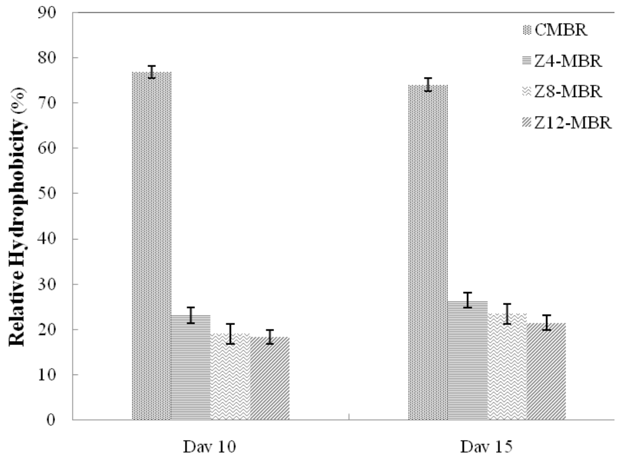

Fig. 8. Relative Hydrophobicity in 10th and 15th day of experiment in different MBRs. (Membrane bioreactor without zeolite called CMBR and the hybrid MBR with zeolite, namely 4 g L^{-1} zeolite (Z4-MBR), 8 g L^{-1} zeolite (Z8-MBR) and 12 g L^{-1} zeolite (Z12-MBR)).

and the mean diameter of flocs in 15th day of experiment, Z8-MBR has the maximum absorption in the solution. In CMBR, Z4-MBR, Z8-MBR and Z12-MBR, the mean particles sizes are 43.44 µm, 91.27 µm, 108.24 µm and 100.43 µm, respectively. As zeolite particles are sized 75–90 µm and ZMBRs particles have an average size of 100 µm (or more), it can be concluded that biofilm thickness is more than 20 µm. Other researchers have also reported the growth of floc size due to biofilm formation on zeolite [27].

3.8. The effect of Zeolite on RH

Hydrophilicity/hydrophobicity of organic matters plays a key role in membrane fouling. Due to the hydrophobicity of sludge and hydrophilicity of membrane (Polyethylene: the surface of the polyethylene can be altered to form a hydrophilic layer without altering the bulk properties of the polymer) the degree of membrane fouling in CMBR has increased. As revealed in Fig. 8, adding zeolite in bioreactor leads to reduced hydrophobicity and increased hydrophilicity. The amount of average RH (between the 10th and 15th day) in the bulk solution after the addition of zeolite has increased by 67.19%, 71.89% and 73.69%, in Z4-MBR, Z8-MBR and Z12-MBR respectively in comparison with CMBR. Elevated hydrophilicity is due to zeolite cations binding (ion-exchange mechanism especially with ammonium of the solution) to the cell wall. Song et al. have reported that increased hydrophilicity is due to environmental shocks in a system, i.e. adding zeolite can be somehow considered as an environmental shock [30]. Over time, saturation of zeolite surface causes increased hydrophobicity of the new bacteria.

3.9. The effect of Zeolite on ZSV

The interface height is recorded at regular time intervals, and the linear portion of the settlement curve (interface level vs. time) is used to calculate ZSVs, occurring after an initial lag phase and before a final compression phase. According to Table 3, Z8-MBR has the best ZSV rate. In Z12-MBR, the tension between zeolite and cell wall causes cell disruption and results in diminished ZSV and elevated SMP at higher doses of zeolite (as in Section 3.3).

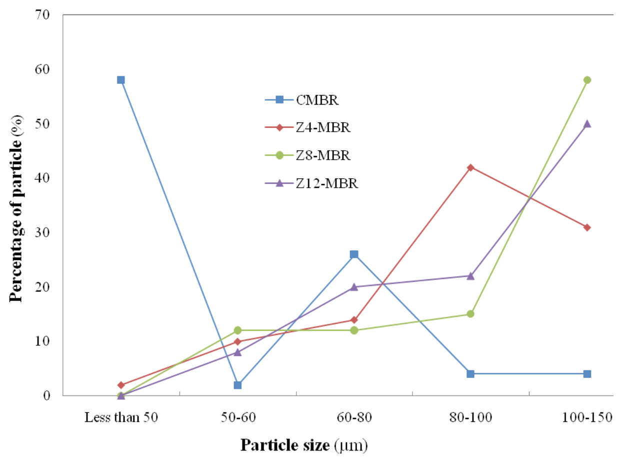

Fig. 9. Particle size distribution (PSD) after 15 days of operation. (Membrane bioreactor without zeolite called CMBR and the hybrid MBR with zeolite, namely 4 g L^{-1} zeolite (Z4-MBR), 8 g L^{-1} zeolite (Z8-MBR) and 12 g L^{-1} zeolite (Z12-MBR)).

Table 3
ZSV measurement

Type of MBR	10th day		15th day	
	ZSV (cm/min)	R^{2*}	ZSV (cm/min)	R^2
CMBR	0.35	0.9891	0.3	0.98
Z4-MBR	0.6	0.9979	0.5	0.972
Z8-MBR	2.14	0.998	2.06	0.994
Z12-MBR	1.17	0.9967	1.597	0.9985

* Using linear model of settling curve for calculating R^2

4. Conclusion

This study has examined the effectiveness of zeolite for controlling the fouling of membrane in a hybrid membrane bioreactor. The ZMBR systems reduced membrane fouling, in terms of lower TMP and lower SMP. After 15 days of operation, in the bulk solution, SMP_C has decreased by 18.96%, 42.11% and 19.44%, after addition of zeolite in Z4-MBR, Z8-MBR and Z12-MBR respectively compared to CMBR due to adsorption process and/or also attachment onto the membrane surface as a bio-layer. ZMBR systems enhanced zeta potential –7.8 mV, –5.35 mV, –3.89 mV due to reduction of the concentration of SMP in Z4-MBR, Z8-MBR and Z12-MBR, respectively. However, these additions increased the concentration of both EPS_C and EPS_P.

Soluble organic compounds (especially SMP_C) was absorbed by zeolite powder or sludge attached on the zeolite powder, lowering the chance of direct interaction between colloids/soluble and the membrane. Thus, ZMBRs generate significantly lower TMP in long-term operations; 31.38%, 40.45% and 37.14% in Z4-MBR, Z8-MBR and Z12-MBR respectively compared to CMBR. It increased PSD by 91.27 μm, 108.24 μm, 100.43 μm in Z4-MBR, Z8-MBR and Z12-MBR, respectively. Increasing dosage of zeolite is not always helpful to alleviate membrane fouling; thus, the optimized zeolite dose for reducing membrane fouling in MBR is reported as 8 g L^{-1} because of its performance. ZMBRs, as compared to the CMBR system, significantly reduced the membrane fouling.

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