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Hygienic water production in an innovative air lift bioreactor followed by high antifouling ultrafiltration membranes modified by layer-by-layer assembly

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ABSTRACT

The possibility of continuous hygienic water production from wastewater in a combined system comprising an innovative air lift bioreactor and a membrane module was investigated. To improve the performance of a commercial poly acrylonitrile ultra-filtration membrane, layer by layer method was used. The effect of bilayer number (1, 3 and 5) and the type of polyanions on the membrane surface characteristics was investigated. Poly (4-styrenesulfonic acid-*co*-maleic acid) and poly (methacrylic acid) with different molecular weights were considered as polyanions and poly diallyl dimethyl ammonium as a polycation was used for surface modification. The polyelectrolyte multilayer fabrication was verified via attenuated total reflection, scanning electron microscope, atomic force microscopy, and contact angle characterization. According to the results, poly (4-styrenesulfonic acid-*co*-maleic acid) with higher molecular weight and charge density showed a better performance in terms of antifouling performance and flux recovery ratio compared to poly (methacrylic acid). An increase in bilayer numbers caused an increase in hydrophilicity and antifouling property. A membrane with 5 bilayers of Poly (4-styrenesulfonic acid-*co*-maleic acid) and poly diallyl dimethyl ammonium presented the highest hydrophilicity (the lowest contact angle) and flux recovery ratio (100%), and showed a gel-like layer on the surface. The production of hygienic water was confirmed by microbial test results.

1. Introduction

The increased population and demand due to higher living standard create challenges for clean water access. Many researches have reported that water reuse is an effective alternative to supply the required water of drinking, manufacturing, washing and etc (Omenn, 2006). Membrane technology has been of interest for water reuse to gain high quality water from wastewaters over last decades. For instance, membrane technology was coupled with photocatalytic bioreactor for water purification (Athanasiou et al., 2016), and a membrane was used for micro-pollutant removal from surface water (Manda et al., 2014).

Membrane technology also can be coupled with other technologies to improve the separation efficiency. Cost-effective biological processes as preliminary treatment have been coupled with membrane technology to reduce organic pollutants and nutrients from wastewater. This combination is well-known as a membrane bioreactor (MBR) which has been developed to treat different wastewaters. For example, MBR was applied for treating sewage sludge (Collivignarelli et al., 2017), industrial wastewater (Deowan et al., 2016), municipal wastewater (Tan et al., 2015), azo dye (You and Teng, 2009), and soft drink industrial wastewater (Sheldon and Erdogan, 2016).

The main category of fouling phenomenon in MBR is biofouling which is so problematic through creating biofilm as a result of attaching the microorganisms to membrane surfaces. An acceptable approach to prevent MBR fouling is to restrict the adhesion of bacteria on membrane surface. Thus, the top layer of membrane plays an essential role for membrane separation performance. Surface modification methodology has been recognized as a favored method to reduce fouling phenomena such as plasma modification (Moraczewski et al., 2016), interfacial polymerization (Zhang et al., 2016) polyelectrolyte adsorption (Xu et al., 2017),

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grafting polymerization (Kochameshki et al., 2017), and layer by layer (LBL) assembly (Chen et al., 2014).

LBL assembly is a technique to generate thin film composite (TFC) membrane due to depositing polyanion and polycation on the membrane surface consecutively, which increases the hydrophilicity of membrane surface (Das and Pal, 2002). Electrostatics (predominant), hydrogen bonding, and van der Waals forces are the main driving forces to perform LBL-assembly (Kochkodan and Hilal, 2015). LBL-assembly was recently investigated to modify membrane surface for water purification and wastewater treatment. Multilayered membranes of polyvinylamine (PVA) and polyvinylsulfate (PVS) on a porous polymer (polyacrylonitrile/polyethylene terephthalate) support were used for desalination of seawater (Toutianoush et al., 2005). In another study, poly (styrene sulfonic acid) sodium salt (PSSS) was utilized for preparation of PSSS/polypiperazine-amide nanofiltration (NF) membrane for the softening of highly concentrated seawater (Hu et al., 2014). Zhu et al. (2014) applied dual-layer polybenzimidazole/polyethersulfone (PBI/PES) NF hollow fiber membranes for heavy metals removal from wastewater (Zhu et al., 2014).

Another important aspect of MBR system is the type of bioreactor used. Different bioreactors with batch and continuous flow patterns were coupled with membrane module to treat various wastewaters (Zinatizadeh and Ghaytooli, 2015). Recently, continuous feed and intermittent discharge (CFID) regime, which presents both features of batch and continuous regimes, has been attracted by researchers (Asadi et al., 2012b). Bioreactors with CFID regime has been investigated for removing carbon and nutrients from industrial estate and milk processing wastewaters (Asadi et al., 2012a; Rezaee et al., 2015). As a finding, in CFID regime, microorganisms are separated from treated wastewater at the end of reaction phase inside the bioreactor, so a secondary sedimentation tank is needless. By applying membrane module inside the bioreactor and passing the effluent from membrane module, an efficient submerged MBR system with low biofouling could be achieved.

In the previous study, a novel continuous feed and intermittent discharge air lift bioreactor (CFIDAB) was operated for simultaneous carbon and nutrients removal from soft drink wastewater (Asadi et al., 2016). Anaerobic, anoxic, and aerobic zones are separated physically in the CFIDAB, so that three different zones are provided in a single bioreactor under continuous aeration condition. Moreover, the sedimentation process is occurred in the bioreactor as the CFIDAB has both features of batch and continuous regimes. This system showed a good ability to remove carbon and nutrients from wastewater with 99%, 75% and 50% of carbon, nitrogen, and phosphorus removal efficiencies at hydraulic retention time (HRT) of 12 h. It was concluded from this study that the CFIDAB can be coupled with membrane technology to gain a system for hygienic water production with low biofouling potential.

In the present study, poly acrylonitrile (PAN) ultra-filtration membrane was modified by LBL-assembly to enhance its antifouling potential and performance. The CFIDAB as a single bioreactor was operated at the optimum condition achieved from our previous study to treat soft drink wastewater biologically followed by a high performance ultrafiltration membrane set-up. The type of polyanion (different molecular weights) and the number of bilayers (1, 3, and 5 bilayers) were selected as variables. Poly (4-styrenesulfonic acid-*co*-maleic acid) (PSSMA) and poly (methacrylic acid) (PMA) as polyanions and poly (diallyl dimethyl ammonium) (PDADM) as polycation were employed. Moreover, microbial test to prove the production of hygienic water was performed. It should be mentioned that hygienic water without the presence of microorganisms and carbon pollutants is used for a wide range of activities except for drinking uses.

2. Materials and methods

2.1. Wastewater characteristics

The system was operated to treat industrial soft drink wastewater, collected from a soft drink processing production plant (Zamzam Co., Kermanshah, Iran). The characteristics of the used wastewater is presented in Table 1. In order to avoid any changes in the wastewater characteristics, the samples were stored at 4 °C. NH_4Cl and KH_2PO_4 were used to adjust C/N/P ratio around 100/15/5.

2.2. Polymer and membrane characteristics

PAN ultra-filtration membrane was used as a support membrane purchased from STERLITECH (USA). Also, PSSMA (Aldrich; Mw = 20,000), PMA (Aldrich; Mw = 4000–6000), and PDADM (Aldrich; Mw = 400,000–500,000) were purchased from Sigma-Aldrich.

2.3. Surface modification method

In order to deposit polyanion and polycation layers on the PAN ultra-filtration membrane, the commercial membrane was pretreated by 1.5 M NaOH solution for 1.5 h. After that, it was washed with de-ionized water until reaching a neutral pH. The concentration of PSSMA, PDADM, and PMA was 1.66, 0.2, and 0.2 wt%, respectively. $CaCl_2$ was added into the PMA and PSSMA solutions to provide 0.5 M of $CaCl_2$. Also, NaCl was added to PDADM solution (0.5 M of NaCl). $CaCl_2$ and NaCl were used as supporting electrolytes. The pH of PSSMA, PDADM, and PMA solutions was adjusted around 2.5, 2.5, and 5, respectively, to obtain ionic solutions. Polycation and polyanion layers were deposited on the membrane surface alternately through dipping steps in the polymer solutions which is a well-known procedure as a static self-assembly method. A dipping step in PDADM and PMA solutions lasted for 5 min, whereas it was 2 min for PSSMA solution. The membrane was inserted into distillated water for 1 min after each step.

2.4. Flux decline and membrane fouling

For filtration test, a dead-end stirred cell was used as shown in Fig. 1. Pressurized nitrogen gas was utilized to supply the required operating pressure (4 bar). In order to provide a good mixed-solution in the set-up, the existing mixer was rotated at 400 rpm. At first, the membrane was hydraulically compressed at a pressure of 3 bar for about 15 min with pure water to avoid the compaction effect of the membrane. Then, 100 ml of the bioreactor effluent was passed from the dead-end set-up.

Permeate samples were collected over a certain time and weighed. The permeation flux was calculated based on the following equation (1):

Table 1
Characteristics of soft drink industrial wastewater.

Parameters	Unit	Amount
Soluble COD	(mg/l)	945–1200
Soluble BOD_5	(mg/l)	756–960
N-NH_4^+	(mg/l)	140–150
N-NO_3^-	(mg/l)	19–21
TN	(mg/l)	159–171
TP	(mg/l)	46–50
pH	–	7.5–8.5