



Low-resistance anti-fouling ion exchange membranes fouled by organic foulants in electrodialysis



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ABSTRACT

We present an original research on the anion exchange membranes (AEMs) (TWEDAI, TWEDAI, FAS, 3362BW) and the cation exchange membranes (CEMs) (TWEDCI, TWEDCII, FKS, 3361BW) fouled by four typical organic foulants (SDBS, BSA, sodium humate, SA) through electrostatic interaction, affinity interaction and geometric interaction. After fouled by different organic foulants, the unique TWEDAI membrane kept a relatively lower resistance than those of the other ion exchange membranes. It was worth noting that the negligible change of its membrane resistance was mainly attributed to its small original resistance and the structure stability, indicating its excellent anti-fouling characteristic. This new type of low-resistance anti-fouling ion exchange membranes opens an exciting potential for water treatment and separation engineering.

1. Introduction

Water plays an important role in supporting and maintaining human health and sustainable ecosystem development. Population growth, urbanization, industrialization and consumption pattern changes have generated ever-increasing demands for freshwater resources worldwide [1]. Different technologies are employed to produce potable water such as membrane process, multi-stage flash distillation, multieffect distillation, vapour compression desalination [2]. The interest in the membrane process has increased with the development of membrane science and technology [3]. For membrane treatment, two major desalination process are reverse osmosis (RO) and electrodialysis (ED). The ED process is more convenient than RO process because higher brine concentration can be obtained in the former process as there are no osmotic pressure limitations [4]. The desalination can also be controlled by fixing the current and time, reflecting ED is a very flexible process [5]. The advantages of ED over other separation processes includes low energy cost, versatility in term of wide variety of feed streams that can be utilized with minimum requirement of pre-treatment, easier and low cost of maintenance [4]. For meeting the strict environmental regulation in the developed countries, ED is considered as a green method for desalination of complex saline solutions in food, beverage, drug and chemical industries as well as in biotechnology and wastewater treatment [5]. Electrodialysis is an

electrochemical separation process, which employs electrically charged ion exchange membranes with an electrical potential difference as a driving force, resulting in a dilute stream and a concentrate stream formed by ions selectively transport through ion exchange membranes with good permselectivity, low electrical resistance, and acceptable thermal, chemical stability and mechanical strength [6]. However, the fouling of ion exchange membranes is essential issue in the operation of the electrodialysis desalination process.

Membrane fouling, which is usually caused by the chemical and/or physical interactions between membranes and foulants [7], is a very tricky problem in the electrodialysis process [8,9]. The fouling can increase the membrane electric resistance and partly lose the selectivity of the ion exchange membranes [10–14], resulting in a higher power consumption and a lower water quality than those of the original state. The membrane fouling is closely related to the functional groups and surface properties of the membrane [15,16], and foulant species, such as organics [7,10–14], inorganics [9,14,17] and metals [18]. For example, most organic substances are negatively charged in natural waters, while anion exchange membranes are positively charged. The anion exchange membranes can easily repel the cations and preferentially transport the anions through the membranes [19]. As a result, the anion exchange membranes are prone to be fouled due to the electrostatic interaction [5,20]. These organic substances are adsorbed on the surface of the membranes and/or lodge themselves inside the mem-

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branes [21,22]. The transport of sodium dodecyl benzene sulfonate (SDBS) has been measured in four different anion exchange membranes by Korngold's group [23]. The restricted transport of the relatively large DBS ions leads to the increase of the electric resistance. In addition, the accumulation of DBS ions inside the membrane tends to give the anion exchange membrane cation exchange properties. E.J. Kobus and P.M. Heertjes has studied that the DS^- ions exchange stoichiometrically with the original counterions (Cl^-) and the very low membrane mobility of large organic anions. They also presented the excellent demonstration of the reversibility (by current reversal) [24]. Katz found how EDR had achieved a major increase in the stability of membrane process performance and published genial papers. The papers had inspired the development of the antifouling ED reversal which was a breakthrough innovation in ED [25,26]. Lindstrand's group [7] has reported that the structure and size of the fouling molecule can affect the fouling degree of the ion exchange membranes. In detail, the longer chain or the larger size of the foulants can lead to the more severe fouling of the ion exchange membranes. Therefore, it is very important to choose appropriate foulants for fouling the specific ion exchange membranes to study the fouling mechanism based on the interactions between the foulants and the membranes [27,28].

Herein, we present different fouling mechanisms of four types of cation exchange membranes (CEMs) and other four types of anion exchange membranes (AEMs) fouled by different organic compounds. The compaction and surface hydrophilicity of the membrane matrix are obtained by testing water permeability coefficient and contact angle, respectively. Furthermore, the membrane electric resistances before and after fouling have been measured to analyze the fouling mechanisms based on electrostatic interaction, affinity interaction and geometrical factors.

2. Experimental

2.1. Organic foulants

The organic foulants used in the experiments were sodium dodecyl benzene sulfonate (SDBS), bovine serum albumin (BSA), sodium humate and sodium alginate (SA). All these were purchased from Qingdao Zhengye Reagent Instrument Co. Ltd.. SDBS is a kind of anion surfactant with the molar mass of 348.48 g/mol [11]. The critical micelle concentration (CMC) of SDBS is 415 mg/L, and the effect of micelle forming on membrane fouling is negligible [20]. It dissociates into DBS anion and sodium cation in aqueous solution. BSA is a globular protein with an average molecular weight of 67 kDa. It is amphoteric with the isoelectric point of 4.7 [29]. Sodium humate consists of both aromatic and aliphatic components with phenolic and carboxylic functional groups, respectively. Commercial sodium alginate (SA) extracted from brown algae (with the reported molecular weight of 12–80 kDa) is employed in the experiment [30].

2.2. Ion exchange membranes

Four kinds of AEMs (FAS, TWEDAI, TWEDAI and 3362BW) and four kinds of CEMs (FKS, TWEDCI, TWEDCII and 3361BW) are used in this work. The chemical and physical properties of all the ion exchange membranes are shown in Table 1 and Table 2. FAS and FKS are homogeneous ion exchange membranes manufactured by FuMa-Tech. FAS and FKS reinforced by polymeric net are based on aminated poly (phenylene oxide) and sulfonated polyether sulfone, respectively. TWEDAI, TWEDCI, TWEDAI and TWEDCII are homogeneous membranes manufactured by Shandong Tianwei Membrane Technology co. Ltd. The former two membranes are made by impregnation and polymerization of aliphatic monomers in a non-woven fabric, and the last two membranes are from aromatic monomers in a non-woven fabric. 3362BW and 3361BW are heterogeneous ion exchange membranes manufactured by Shanghai Shanghai Water Treatment Material

Table 1

The properties of FAS, TWEDAI, TWEDAI and 3362BW anion exchange membranes.

Membrane property	FAS	TWEDAI	TWEDAI	3362BW
IEC (mmol/g dry)	1.0	1.0	1.0	1.8
Water content (%)	19	15	15	40
Transport number	≥ 0.95	≥ 0.97	≥ 0.93	≥ 0.89
Electric resistance ($\Omega\text{-cm}^2$)	≤ 6	≤ 3	≤ 6	≤ 16
Thickness (mm)	0.13	0.05	0.09	0.50
Membrane structure	Aromatic (PPO)	Aliphatic	Aromatic	Aromatic (PSt)
Manufacturer	FuMA-Tech	Shandong Tianwei	Shandong Tianwei	Shanghai Shanghaihua

Table 2

The properties of FKS, TWEDCI, TWEDCII and 3361BW cation exchange membranes.

Membrane property	FKS	TWEDCI	TWEDCII	3361BW
IEC (mmol/g dry)	1.0	1.0	1.0	2
Water content (%)	19	15	17	40
Transport number	≥ 0.96	≥ 0.96	≥ 0.93	≥ 0.90
Electric resistance ($\Omega\text{-cm}^2$)	≤ 8	≤ 3	≤ 6	≤ 16
Thickness (mm)	0.13	0.05	0.09	0.4
Membrane structure	Aromatic (PES)	Aliphatic	Aromatic	Aliphatic (PE)
Manufacturer	FuMA-Tech	Shandong Tianwei	Shandong Tianwei	Shanghai Shanghaihua

Co. Ltd., which are made of polystyrene resin bonded with polyethylene and reinforced by polymeric net.

2.3. Testing module and method

Fig. 1 shows a diagram of the experimental system, which includes AEM module and CEM module, a DC power supply device from GW INSTEK, magnetically driving pumps from Nanjing's Rick Micro-Pump Co. Ltd. and flow meters from Changzhou Chengfeng Instrument Co. Ltd. As shown in Fig. 2, the modules had the cross-sectional area of 7 cm². One module was assembled with AEMs, and the other with CEMs. Magenta is special CEM, blue is compartment, yellow is a kind of AEM and red is another kind of AEM in the AEM module. While CEM module has a same design, yellow represents a kind of CEM and red represents another kind of CEM in the CEM module. Each kind of all the membranes was tested for three times before and after fouling.

In the experiment, the voltage output of the DC power supply was set to the constant 14.5 V and the electric current was recorded. There are 21 AEM membranes in the AEM module, with single membrane under 0.5 V. In addition, 2 V was individually set on special membrane near the cathode and anode chamber. In all, the constant 14.5 V is obtained according to the following equation: $21 \times 0.5 \text{ V} + 2 \times 2 \text{ V}$. The CEM module is set as the same as that of the AEM module. 10 L of 1% Na₂SO₄ was used as the electrode solution. As a blank reference experiment, 10 L of 3000 mg/L NaCl solution was respectively fed into the chambers next to the membranes in the AEM and CEM modules. The solutions were kept in a water bath at 25 °C and replaced with fresh solutions every day. When the blank reference experiment was stable, the NaCl solution was added with 500 mg/L organic foulants for the fouling experiment. During the long time of fouling tests, the salt concentration was retained the same, attributed to the cycle use of the salt concentration.

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