

Property characterization and mechanism analysis on organic fouling of structurally different anion exchange membranes in electro dialysis



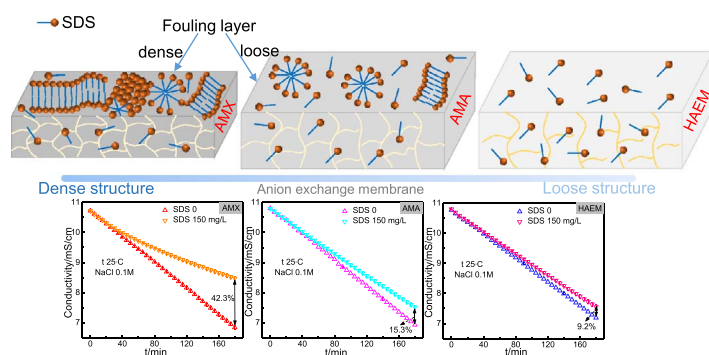
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GRAPHICAL ABSTRACT



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ABSTRACT

The physicochemical and structural properties of the AMX (homogeneous AEM), AMA (semi-homogeneous AEM) and HAEM (heterogeneous AEM) were characterized systematically to examine their effects on membrane fouling from sodium dodecyl sulfate (SDS) in electro dialysis. Results demonstrated that the fouling of three AEMs was closely related to their membrane structure, and the denser membrane structure caused more SDS accumulated on the membrane surface to make the severer membrane fouling. The existence of SDS in the dilute solution caused the desalination rate of AMX, AMA and HAEM decreased by 42.3%, 15.3% and 9.2%, respectively. Different fouling layer was formed on the surface of AMX and AMA except HAEM, which was demonstrated firstly by element mapping of membrane cross-section. Electrochemical impedance spectroscopy (EIS) suggested the ionic migration through the fouled AMX was restricted completely, different from the trans-membrane migration of partial ions through the fouled AMA and HAEM. Compared with the fouled AMA and HAEM, the performance of the fouled AMX caused by SDS was more difficult to be recovered due to its denser membrane structure.

1. Introduction

Electrodialysis (ED) is an electrochemical technology which utilizes

potential difference as the driving force for the separation of charged ions via the ion exchange membranes (IEMs) [1]. Because of the advantages such as high water recovery rate, long operation life of

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membrane and more flexible for various feed water qualities, ED is not only used for desalination of brackish water and seawater, but also can be utilized for the treatment of industrial wastewater and removal of bromate from natural water [2–7]. Nevertheless, the fouling of anion exchange membrane (AEM) is still a limiting factor for the wide application of ED in the treatment of industrial wastewater. It's known that many organics such as humate and anionic surfactants in the wastewater have negative charge, which makes it easier for them to be adsorbed on the AEMs and deteriorate the membrane properties [8–11].

The influence of different organics on membrane fouling in the ED system has been studied widely. Lee et al. [12] investigated the fouling potential of AEMs by different foulants such as humate, bovine serum albumin (BSA) and sodium dodecyl benzene sulfonate (SDBS), and found SDBS caused severer fouling on the AEM than the others. Persico et al. [13] explored the peptide species and nature of peptide/AEMs (AMX-SB) interactions involved in fouling at different pH and demonstrated that the physicochemical and structural characteristics of peptide such as the charge distribution had an influence on the AEM fouling. It was reported that anionic polyacrylamide (APAM) caused the AEM fouling by the formation of a gel layer on the membrane surface and affected the desalination performance of AEM [11,14]. A surfactant could also cause the severe fouling effect on the AEM by its accumulation on the membrane surface and damaged the membrane [12,15,16].

There are two main factors related closely to membrane fouling: electrostatic interaction between organics and the fixed charged groups of AEM, and affinity interactions such as π - π interaction between aromatic compounds and the membrane matrix. Besides, geometrical factors such as the molecular size of foulants and the network structure of membranes also play an important role in fouling [17]. Mitsuru et al. [18,19] found that the chemical composition of AEMs could also affect the formation of membrane fouling, which was proved by comparing the antifouling performance of the aliphatic and aromatic AEMs with various anion exchange groups. However, the study on the influence of the structural difference of AEMs on the fouling remained scarce.

The IEMs are commonly classified into homogeneous, heterogeneous and semi-homogeneous membranes according to their structure difference, which resulted from the different preparation technologies. Many obvious differences existed among the different types of IEMs, such as surface roughness, pore size, area electrical resistance, surface charge distribution and surface hydrophilicity, etc.; moreover, the membrane properties might affect the desalination performance, membrane fouling and service life during the ED process. The heterogeneity degree of IEMs was reported to have an influence on the ion transport through membrane [20–22]. In addition, the differences in the stability and durability of the homogeneous and heterogeneous IEMs were studied by the aging tests in hypochlorite [23]. However, the study on the influence of the structure difference between the homogeneous and heterogeneous AEMs on the membrane fouling during ED hasn't been found up to now.

In this study, the homogeneous, semi-homogeneous and heterogeneous AEMs were utilized to investigate the effects of membrane properties especially structure difference on the AEM fouling, using sodium dodecyl sulfate (SDS) as a model foulant. The physicochemical and structural characterization of the three pristine AEMs were carried out by the measurement of water content, zeta potential, water contact angle and scanning electron microscopy (SEM), etc. The potential difference and desalination performance of the different AEMs with SDS in the feed solution were measured to examine the different fouling degree of the three AEMs under the same conditions. Moreover, the properties of the three fouled AEMs by SDS were characterized by various analytical techniques to further investigate the fouling difference. EIS was utilized to further characterize the electrochemical properties of the three fouled AEMs. The aim of experimental investigation was to reveal the relationship between the membrane structure and the fouling

propensity of AEMs.

2. Experimental

2.1. AEM samples and reagents

The homogeneous AEM (NEOSEPTA AMX) was obtained from Astom in Japan, semi-homogeneous AEM (IONSEP® AM-A) and heterogeneous AEM (3364) were from China, which were labeled as AMX, AMA, HAEM, respectively. All these AEMs were the polymers with quaternary ammonium groups, but they were obtained by different preparation techniques. The fundamental properties of these AEMs provided by the manufacturer were shown in Table S1. Sodium dodecyl sulfate (SDS) was used as a model foulant and purchased from Alfa Aesar. Other reagents such as NaCl (A.R.), Na₂SO₄ (A.R.) were used to prepare the solutions in the experiment. All solutions were prepared using ultra-pure water (18.2 M Ω ·cm⁻¹, Milli-Q, Millipore).

2.2. Membrane fouling experiments

The ED experiments were performed in an ED apparatus with different types of AEMs. One sheet fresh AEM (AMX, AMA or HAEM) and two sheets CEMs from the same manufacturer were contained in an ED stack, which was divided into three compartments, including the dilute, concentrated, and electrode compartments.

In the fouling experiment, the 500 mL feed solution containing 0.1 M NaCl and 150 mg/L SDS in one beaker was flowed through the dilute and concentrated compartments of ED stack, respectively, then into the same beaker to ensure that the electrolyte concentration of the feed solution was unchanged during electro dialysis. The fouling experiment was carried out at a current density of 3 mA·cm⁻². The potential difference (ΔE) of AEM was recorded online using a data logger and the showed ΔE was the average value of the three measurements. The details were reported in the previous study [16]. The AEM after the fouling experiment was dried naturally or stored in ultra-pure water for further characterization.

In the desalination experiment, the initial solution in dilute compartment was 500 mL of 0.1 M NaCl and 150 mg/L SDS, and the feed solution in the concentrated compartment was 500 mL of 0.1 M NaCl, and the solutions in the two different beakers were circulated through the dilute and concentrated compartments, respectively, and then flowed into the two beakers. The membrane stack consisted of one sheet fresh AEM (AMX, AMA or HAEM) and two sheets fresh CEMs. The desalination experiment was performed at a voltage of 4.0 V. The electrical conductivity of the solution in dilute compartment was measured online by a conductivity meter (S230, SevenCompact, Mettler Toledo). Besides, the total organic carbon (TOC) of the solution in the dilute and concentrated compartments was measured by a TOC analyzer (Shimadzu TOC-V_{CPH}, Japan) before and after the desalination experiment. The desalination experiment with the same type of AEM was repeated for three times. The details were reported in the previous study [16].

2.3. Properties characterization of AEMs

The water content of AEM was measured and expressed as the amount of water per gram of a chloride-ion-form AEM (g H₂O/g chloride-ion-form dry AEM). The ion exchange capacity (IEC) of AEM was determined by the titration method and expressed as millimole per gram of AEM (mmol/g chloride-ion-form dry AEM). The area electrical resistance of AEM was measured by LCR Meter (IM3533, HIOKI, Japan) set at 100 kHz, 100 mV in 0.5 M NaCl at 25 °C. The contact angle of AEM was measured by the sessile drop method using contact angle analyzer (OCA20, DataPhysics, Germany). The zeta potential of AEM was measured using a zeta potential measurement analyzer (Anton Paar, Austria) with 1 mM KCl solution as the background electrolyte.

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