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Diffusion behaviour of multivalent ions at low pH through a MFI-type zeolite membrane

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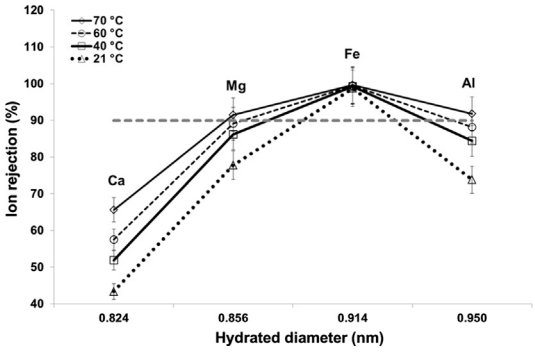
GRAPHICAL ABSTRACT



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ABSTRACT

Zeolite membranes have been widely examined for desalination. Work to date has shown effective performance in monovalent rich solutions, but the understanding of the unique ion-zeolite interactions for multivalent ion rich solutions, such as those found in acidic mining wastewaters, has not yet been studied. Filtration performance of MFI-type zeolite membrane was evaluated on a model multivalent ion, Fe^{3+}, Al^{3+}, Ca^{2+} and Mg^{2+}, solution with total dissolved solids (TDS) of 97,000 mg L^{-1} and pH 2.03 at between 3 MPa and 7 MPa and 21 °C to 70 °C. At 7 MPa and 21 °C, rejection for Fe^{3+} was 97%, 80% for Al^{3+} and Mg^{2+}, and 50% for Ca^{2+}. This behaviour followed the rejection of ions with larger hydrated diameter, except for Al^{3+} which was attributed to its unique strong interaction with zeolites. However, an unusual trend of increasing rejection with increasing temperature was observed. Instead of activated transport which occurs with monovalent or dilute solutions, temperature accelerated the infiltration of multivalent ions into the zeolite structure to further block ions and synergistically increased rejection. Zeolite membranes exhibited unique effects in multivalent ion rich solution that could be further utilised in niche desalination applications or benefit other applications such as sensors.

1. Introduction

Ceramic membranes made from zeolites are good candidates for desalination as they possess the required small pore properties to reject ions while diffusing water [1,2]. Zeolite membranes, especially MFI-type, have been widely examined for desalination [2–11], and may

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offer an alternative option for desalination of acidic, metal- and sulphate containing mining wastewaters, where they may have extended service life and increased cleaning ability compared to current polymer membrane technology. The MFI-type zeolite has orthorhombic crystal symmetry with nearly cylindrical, 10-member ring channels. The aperture size of the MFI-type zeolite is around 0.56 nm [4,12–14], which is smaller than the sizes of hydrated ions (e.g. Ca^{2+} 0.824 nm, Mg^{2+} 0.856 nm, Fe^{3+} 0.914 nm, Al^{3+} 0.950 nm, [15]) but larger than the kinetic diameter of water (0.276 nm, [15]).

MFI-type zeolite membranes have demonstrated high ion rejections, even for the smallest ions (e.g. Na^+) when working as a reverse osmosis (RO) membrane [8,10], and also showed great ability to separate dissolved organics from aqueous solution [10,16]. Our recent work [11] also demonstrated the possibility of avoiding the pre-treatment needed for polymeric RO membranes by using MFI-type zeolite membranes for desalination of saline recycled wastewater. The zeolite membrane also showed no deterioration after extreme chlorine or acid cleaning, enabling simplified cleaning and biofouling control techniques.

The performance of MFI-type zeolite membranes, however, was found not always constant. For example, high rejection (> 93%) was achieved for Ca^{2+}, Mg^{2+} and Na^+ from 0.3 wt% seawater solution [8], and Na^+ rejection of 99.4% was reported for 0.1 M NaCl solution [10]. Recent work carried out by Garofalo and co-workers also demonstrated high rejections of Na^+ (99.8% and 99.6% for 0.2 M and 0.9 M NaCl feed solutions, respectively) on scaled-up MFI-type zeolite membranes (30 cm long) by vacuum membrane distillation [17]. However, low ion rejection (e.g. 30% for Na^+) was also observed in our previous study [18]. Although significant fluxes (13.8 L m^{-2} h^{-1} and 8.2 L m^{-2} h^{-1} for 0.2 M and 0.9 M NaCl feed solutions, respectively) have been achieved on MFI-type zeolite membranes by vacuum membrane distillation [17], fluxes (e.g. 0.5–1.4 L m^{-2} h^{-1} at a pressure of 2.76 MPa [19]) of MFI-type zeolite membranes when working as a RO membrane are lower than polymer RO membranes possibly due to their greater active layer thickness. Membrane water transport resistance is proportional to the dense surface layer thickness and hydrophilic properties. Zeolite membranes (3–10 μm) [2,3,11] are much thicker than commercial RO membranes (0.2 μm) [20,21]. For MFI-type zeolite membranes to be used commercially, further study is needed to understand the cause of the inconsistent ion rejections and fluxes need to be significantly increased.

Apart from inconsistent ion rejections and low flux of MFI-type zeolite membranes, recent studies also showed that MFI-type zeolites exhibited dynamic behaviour associated with ion interactions in different ion complexes (e.g. seawater, or multivalent ion solution) [18,22–24]. For example, monovalent cations (e.g. K^+, Na^+) can enter the zeolite lattices, while divalent cations (e.g. Ca^{2+}, Mg^{2+}) can be absorbed into the grain boundaries of zeolites [22,23]. The strong uptake of Fe^{3+} and Al^{3+} from a multivalent ion solution containing Fe^{3+}, Al^{3+}, Ca^{2+} and Mg^{2+} into MFI-type zeolite powders resulted in changes to crystal structure and the porous properties [18]. These changes in structure and porosity could impact on diffusion properties of ions in this material when used as a membrane [18,22,23]. Temperature was also found to have an impact on membrane performance. Increasing the testing temperature resulted in an increase in permeate flux, but a decrease in ion rejection [7,8]. Despite ion diffusion through MFI-type zeolite membranes following the hydrated size of ions, more work is needed to correlate ion diffusion with their size and potentially other factors, such as dynamic ion interactions in different ion complexes and operating temperature. In exploring the unique behaviour of ions within MFI zeolite, we recently reported on the beneficial function of multivalent ions to block defects [18]. However no work has been conducted to date exploring the behaviour of these ions in continuous filtration, in particular, with mining wastewaters which contain solutions rich in multivalent ions. Considering the strong interaction between multivalent ions and MFI-type zeolites to block defects and improve salt rejection of NaCl-rich solutions, it remains unknown how these ions would influence membrane performance benefiting these more specialist applications, or even wider applications involving zeolites exposed to multivalent ion rich solutions such as sensors.

In this study, we deposited MFI-type zeolite seeds on the outer surface of a tubular α-alumina substrate using the rubbing method [7,11,25,26], and then used the secondary hydrothermal growth to prepare the MFI-type zeolite membrane [7,11]. The prepared membrane underwent filtration performance testing on a model multivalent ion solution containing Fe^{3+}, Al^{3+}, Ca^{2+} and Mg^{2+} with TDS of 97,000 mg L^{-1} and pH 2.03. The effects of applied pressure, role of operating temperature and influence of acid cleaning on membrane performance were investigated. The dependence of osmotic pressure on the impact of ion diffusion is discussed. Scanning electron microscopy (SEM) was also used to develop a better understanding of the effect of multivalent ion solution exposure on membrane structure and surface morphology.

2. Experimental and methods

2.1. Preparation of MFI-type membrane

The MFI-type membrane was prepared by the seeded secondary growth method [7,11], which involved depositing MFI-type zeolite seeds on the outer surface of a α-Al_2O_3 support using a rubbing method [25,26] followed by growth of the membrane under hydrothermal conditions. This seeded secondary growth method has recently attracted attention as it decouples the nucleation and growth from seed growth, while also reducing the demand for the costly structure directing agents [6,27,28]. The porous α-Al_2O_3 tubular support (95.7% Al_2O_3, apparent porosity 34.9%, external diameter 15 mm, internal diameter 10 mm, length 25 mm, mean pore size ~12.2 μm) used in this study was supplied by Chosun Refractories Co. Ltd., Korea. The MFI-type zeolite seeds (ZSM-5, SiO_2/Al_2O_3 = 360) were purchased from ACS Material, USA. The particle size distribution of the MFI-type zeolite seeds was measured to be between 1000 nm and 3000 nm (peaking at ~1800 nm) by Zetasizer (Malvern Instruments-nano-series) [7]. The secondary growth was conducted by placing the seeded support into a Teflon lined stainless steel autoclave and adding the desired growth solution of 2 mL of 1 M tetra-propyl ammonium hydroxide (TPAOH) (Aldrich), 2 mL of tetraethyl orthosilicate (TEOS) (98%, Aldrich) and 36 mL deionised water (DI water). Secondary growth was carried out at 180 °C for 16 h. After growth, the membrane tube was washed in DI water to remove loose precipitate and was then calcined at 500 °C for 4 h.

2.2. Filtration test

Filtration performance of the prepared MFI-type zeolite membrane was analysed for the ion complex model of mining wastewater, using a high pressure test system used in our previous work [7]. The membrane was installed into the stainless steel membrane housing, and model multivalent ion solution was fed at a flow rate of 5 mL min^{-1} by a high pressure piston pump (Series 1, LabAlliance, USA) with an applied gauge pressure of up to 7 MPa. The ion complex used in this study was prepared by adding 138 g $Al_2(SO_4)_3 \cdot 18H_2O$ (AJAX Chemicals, Australia), 205 g $MgSO_4 \cdot 7H_2O$ (Merck, Australia), 58 g $Fe_2(SO_4)_3 \cdot 9H_2O$ (AJAX Chemicals, Australia) and 1.7 g $CaCl_2 \cdot 2H_2O$ (Merck, Australia) into 2 L DI water. The solution was filtered by a 0.45 μm membrane filter to remove undissolved solids prior to filtration test. The ion concentrations of the pre-filtered solution were determined by inductively coupled plasma-optical emission spectrometry (ICP-OES) (Shimadzu ICPE-9000) and are shown in Table 1. Cl^- was not measured as it is assumed to be minor (estimated to be ~400 mg L^{-1} based on the mount of $CaCl_2 \cdot 2H_2O$ added) in comparison to other ions (e.g. Fe^{3+}, Al^{3+}, Mg^{2+} and SO_4^{2-}) present in the multivalent ion solution. The pH of the pre-filtered multivalent ion solution was measured to be

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