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Stable and efficient composite anion-exchange membranes based on silica modified poly(ethyleneimine)–poly(vinyl alcohol) for electrodialysis



Ravi P. Pandey^{a,b}, Amit K. Thakur^a, Vinod K. Shahi^{a,b,*}

^a Electro-Membrane Processes Division, CSIR-Central Salt and Marine Chemicals Research Institute (CSIR-CSMCRI), Council of Scientific & Industrial Research (CSIR), Gijubhai Badheka Marg, Bhavnagar 364002, Gujarat, India

^b Academy of Scientific and Innovative Research, CSIR-Central Salt and Marine Chemicals Research Institute (CSIR-CSMCRI), Council of Scientific & Industrial Research (CSIR), Bhavnagar 364002, Gujarat, India

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ABSTRACT

Anion exchange membranes (AEMs) have found numerous electrochemical applications because of their good conductivity and permselectivity. Herein, we are reporting a method to prepare silica modified poly (ethyleneimine) (SMPEI) with 3-Glycidoxypropyl-trimethoxysilan (GPTMS) by epoxide ring opening reaction. Stable AEMs of different compositions were prepared with SMPEI and a plasticizer poly(vinyl alcohol) (PVA) by acid catalyzed sol–gel followed by formal cross-linking. The reported method is simple and a green alternative for the preparation of AEM without the use of hazardous chemicals. Suitability of prepared AEMs for electrodialytic application was assessed by analyzing their physicochemical properties, stabilities under operating conditions, conductivity, electro-osmotic and chronopotentiometry studies. A highly suitable membrane, SMPEI/PVA-40, exhibited 55.32 mS cm⁻¹ conductivity (in equilibration with 0.04 N NaCl solution at 30 °C), ion–exchange capacity (1.31 meq g⁻¹) and permselectivity (0.79) with good electrodialytic performance. Preparation protocols and properties of the reported composite AEM represent a promising starting point for architecting highly conducting and stable AEMs, but we have to study the trade-off between properties, stabilities and electro-osmotic mass drag before its commercial exploitation.

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1. Introduction

Positively charged quaternary ammonium tethered polymers have found numerous successful applications in fuel cells, flow batteries, water electrolyzers, electrodialysis (ED), and reverse electrodialysis [1–8]. Current state of knowledge, concerning structure–morphology– performance relationships of anion exchange membranes (AEMs),

* Corresponding author at: Electro-Membrane Processes Division, CSIR-Central Salt and Marine Chemicals Research Institute (CSIR-CSMCRI), Council of Scientific & Industrial Research (CSIR), Gijubhai Badheka Marg, Bhavnagar 364002, Gujarat, India. Fax: +91 0278 2566970.

E-mail addresses: vkshahi@csmcri.org, vinodshahi1@yahoo.com (V.K. Shahi).

is much less in comparison with that about proton exchange membranes (PEMs) [9,10]. Several AEMs based on different polymers, such as poly(2,6-dimethyl-1,4-phenylene oxide) (PPO), copolymer of chloromethylstyrene and divinylbenzene, PVDF-vinylbenzyl chloride, and poly(vinyl alcohol)-poly(1,3-diethyl-1-1-vinyl imidazolium bromide), are reported in the literature [11-15]. These AEMs were prepared via chloromethylation of polysulfone, poly(ether imide), and Cardo poly(ether sulfone)-based polymers using chloromethyl methyl ether (CMME) followed by amination with a tertiary amine [16,17]. In addition, different cationic groups such as imidazolium [18], phosphonium [19], guanidinium [20], sulfonium [21], and pyridinium moieties [22] were also screened for developing stable and efficient AEM. In general, preparation procedures of AEMs, especially chloromethylation and quaternary amination, are complicated and require potentially harmful chemicals such as chloromethyl styrene, different amines and pyridine, etc. [7,23,24] Thus, usage of an expensive and hazardous chemical dramatically increases the manufacturing cost of the AEM. However, the promising strategies for preparing efficient AEM, effect of hydrophobic and ionic clusters on membrane conductivity, and different experimental protocols to assess the suitability of AEM for electro-driven separation technologies are not yet clear.

Abbreviations: AEM, anion exchange membrane; IEM, ion exchange membrane; SMPEI, silica modified poly(ethyleneimine); ED, electrodialysis; CMME, chloromethyl methyl ether; DC, diluted comportment; CC, concentrated comportment; PEM, proton exchange membrane; IEC, ion exchange capacity; CEM, cation exchange membrane; PVA, poly(vinyl alcohol); TGA, thermo-gravimetrical analysis; φ_w , volume fraction of water in the membrane matrix; χ^m , surface charge concentration; I, applied current density across the IEM; τ , transition time; P_s , membrane permselectivity; κ^m , ionic conductivity; A, surface area of membrane; DSC, differential scanning calorimetry; DMA, dynamic mechanical analyzer; SEM, scanning electron microscopy; E, energy consumption; CE, current efficiency

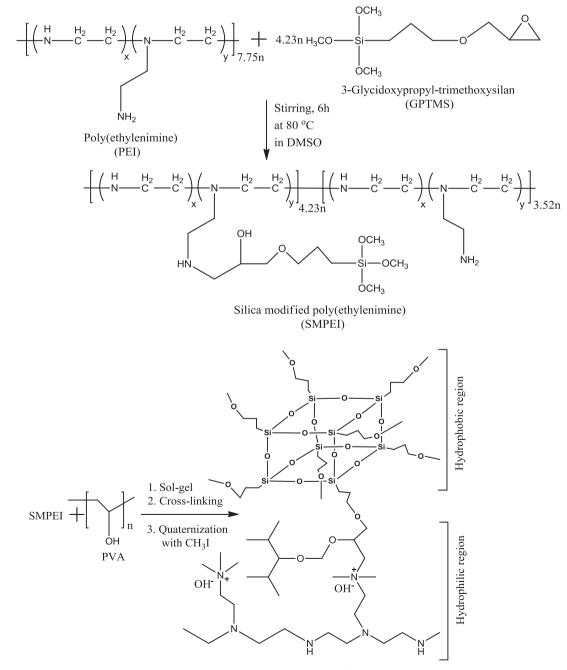
Hydrophobic and ionic clustering in membrane morphology is a key factor for enhanced ion conductivity, which was validated in the case of cation-exchange membranes such as Nafion and sulphonated polymers [25]. Ionic clustering contributes to the formation of hydrophilic channels for the fast diffusion of water and conduction of ions through the membrane, while hydrophobic clusters contribute toward phase separation and stabilities of ion-exchange membranes [10,26]. To facilitate the formation of ionic and hydrophobic clusters, different strategies such as grafting of polymer chain, usages functionalized monomer or block copolymers were adopted [3,7,27]. To solve these problems, attention was rendered for developing organic-inorganic composite membrane forming materials because they combine attractive properties or organics and inorganics [7,27–29]. Therefore, to design stable and efficient membranes by eco-friendly preparation protocols with suitable hydrophobic and ionic clustering responsible for good conductivity is extremely important for next-generation materials.

For developing high performance AEMs with superior morphology, conductivity, stability and scalable synthetic steps, herein we disclose a novel route for producing silica-modified poly (ethylenimine) (SMPEI) and membrane preparation methodology by acid catalyzed sol-gel using poly(vinyl alcohol) (PVA) as a plasticizer. Detailed physicochemical and electrochemical properties of SMPEI/PVA membranes were analyzed to assess their suitability for ED.

2. Experimental

2.1. Materials

Poly(ethyleneimine) (PEI, 50 wt% solution in water), and 3-Glycidoxypropyl-trimethoxysilan (GPTMS) were obtained from Sigma Aldrich Chemicals. Poly(vinyl alcohol) (PVA, Mw: 125,000; degree of polymerization: 1700, degree of hydrolysis: 88%), methyl



Scheme 1. Schematic reaction route for the preparation of SMPEI/PVA composite AEMs.

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