Tailored Graphene Oxide Membranes for the Separation of Ions and Molecules

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Experimental

S1. Reagents and Materials

Flaky graphite was purchased from Sigma-Aldrich Co. LLC. The sulfuric acid (H$_2$SO$_4$), nitric acid (HNO$_3$), hydrochloric acid (HCl), phosphorus pentoxide (P$_2$O$_5$), potassium persulfate (K$_2$S$_2$O$_8$), potassium permanganate (KMnO$_4$), hydrogen peroxide solution (H$_2$O$_2$), sodium hydroxide (NaOH), sodium chloride (NaCl), Calcium chloride (CaCl$_2$), potassium chloride (KCl), magnesium chloride hexahydrate (MgCl$_2$·6H$_2$O), were all purchased from Sinopharm Chemical Reagent Co., Ltd. All reagents were used without any purification. 1-allyl-3-vinylimidazolium chloride ([AVIM]Cl) was purchased from Lanzhou Institute of Chemical Physics (LICP) of the Chinese Academy of Sciences (CAS), whose molecular structures are shown as Figure S3. The de-ionized water (DI) used for all the experiment in this paper was taken from Milli-Q purification system (Millipore, USA). Cellulose acetate (CA) membranes (pore size, 0.45 μm) were purchased from Shanghai Xingya Purifying Material Factory.

S2. Preparation of graphene oxide sheets

The graphene oxide sheets were prepared by the modified Hummers’ method. In detail, a certain amount of flaky graphite was poured into the flask, and HNO$_3$ was added for pre-treatment for 24 hours. The graphite powder was then placed in a muffle furnace for high temperature treatment to prepare expanded graphite. The expanded graphite was pre-oxidized with HNO$_3$, H$_2$SO$_4$, K$_2$S$_2$O$_8$, P$_2$O$_5$, and the product was dispersed in water to collect the precipitate. After drying, the pre-oxidized graphene oxide is further oxidized with KMnO$_4$ and H$_2$SO$_4$ and 30% H$_2$O$_2$. Finally, the graphene oxide solution was dialyzed with deionized water until near neutral, and the concentration of the graphene oxide dispersion prepared was 3.7 mg/mL.
Table S1

<table>
<thead>
<tr>
<th>Species</th>
<th>Hydrated dimeter (nm)</th>
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<tbody>
<tr>
<td>K⁺</td>
<td>0.662¹⁻²</td>
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<tr>
<td>Na⁺</td>
<td>0.716¹⁻²</td>
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<tr>
<td>Ca²⁺</td>
<td>0.824¹⁻²</td>
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<tr>
<td>Mg²⁺</td>
<td>0.856¹⁻²</td>
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<tr>
<td>Glycerol</td>
<td>0.93³</td>
</tr>
<tr>
<td>Fe³⁺</td>
<td>0.96¹</td>
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<tr>
<td>MO</td>
<td>0.99⁴</td>
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<tr>
<td>RhB</td>
<td>1.23²⁴</td>
</tr>
</tbody>
</table>

References:


Figure S1. The AFM image of the GO sheets as prepared.
**Figure S2.** The setups of hydrostatic head test (a) and permeation test (b).

![Figure S2](image)

**1-allyl-3-vinylimidazolium chloride ([AVIM]Cl)**

**Figure S3.** The chemical structure of [AVIM]Cl.
**Figure S4.** SEM images of (a) the cross-section of GO membrane, the surface of (b) GO membrane and (c) i-AVG membrane.

**Figure S5.** The wide XPS spectra of the GO membrane and i-AVG membrane.
Figure S6. The XRD patterns of (a) i-AVG membranes (100 kGy, 0.625 kGy s$^{-1}$) prepared from mixing dispersions with different mass ratio of [AVIM]Cl/GO, (b) i-AVG membranes ([AVIM]Cl/GO=2, 0.625 kGy s$^{-1}$) over different absorbed dose at 0.625 kGy s$^{-1}$ of dose rate and (c) i-AVG membranes ([AVIM]Cl/GO=2, 100 kGy) over different dose rate.

Figure S7. The d-spacing of dry and wet (a) i-AVG membranes (100 kGy, 0.625 kGy s$^{-1}$) prepared from mixing dispersions with different mass ratio of [AVIM]Cl/GO, (b) i-AVG membranes ([AVIM]Cl/GO=2, 0.625 kGy s$^{-1}$) over different absorbed dose at 0.625 kGy s$^{-1}$ of dose rate and (c) i-AVG membranes ([AVIM]Cl/GO=2, 100 kGy) over different dose rate.