

Supporting Information

Highly Efficient and Ultrafast Gaseous Iodide Removal by Controlling the Oxidation Degree and Size of Graphene Oxide Membranes

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EXPERIMENTAL SECTION

Chemicals and Materials.

Concentrated sulfuric acid (H₂SO₄, 98%), phosphorus pentoxide (P₂O₅, AR ≥ 98.0%), hydrochloric acid (HCl, 36.0%), potassium nitrate (KNO₃, 99.9%), borate buffer (50 mM, pH=8) and hydrogen peroxide (H₂O₂, 30%) were purchased from Beijing Chemical Works. Graphite powders (≤ 30 μm, 99.8%), potassium permanganate (KMnO₄, ≥ 99.5%), potassium persulfate (K₂S₂O₈, ≥ 99.5%), ammonium hydroxide solution (NH₄OH) were purchased from Adamas-beta Chemical Co., Ltd. Iodomethane (CH₃I, ≥ 99.5%), sodium iodate (NaIO₃, ≥ 99.8%), sodium iodide (NaI, ≥ 99.0%) and sodium hydroxide (NaOH, ≥ 96.0%) were purchased from China National Medicines Corporation Co., Ltd. All chemical reagents were used as purchased without further purification. The alumina ceramic substrates were obtained from Guizhou New Material of Membranes Co. Ltd (with a diameter of 1 cm and an average pore size of 0.22 μm). He (99.999% pure) was purchased from Shanghai Special. Gas Factory Co., Ltd, China Deionized (DI) water with 18.2 MΩ cm⁻¹ resistivity was obtained by reverse osmosis followed by ion exchange and filtration.

Preparation of different sizes of GO suspensions

The graphene oxide (GO) film was fabricated on the inner wall of the ceramic tube using the pneumatic filtration method. A GO solution with a concentration of 5 mg/mL was employed as the starting material. First, the original solution was diluted to prepare 100 mL of working solution at the desired concentration. Subsequently, the ceramic tube was inserted into a stainless steel housing, and a gas tightness test was performed to ensure system integrity. The prepared GO solution was then introduced into the ceramic tube, and the gas valve was opened to apply controlled pressure. Under the pressure differential within the cavity, the GO nanosheets were uniformly deposited onto the inner wall of the ceramic tube within 10 minutes. The thickness of the resulting film was modulated by adjusting the concentration of the GO solution. After deposition, the obtained GO film was dried in an oven at 60 °C for 1 hour. The preparation of the reduced graphene oxide (rGO) film followed an identical procedure, except that a

highly reduced GO (rGO) solution was used as the precursor.

The previously obtained GO suspension was first diluted by a factor of 50 and then combined with 30 mL of NH_4OH solution to initiate the reduction process, yielding a rGO suspension. Subsequently, the resulting mixture was subjected to ultrasonication followed by continuous stirring at 80 °C for 4 h. After that, the temperature was raised to 90 °C to eliminate the residual NH_4OH . Once cooled to room temperature, the final volume of the rGO suspension was adjusted to 100 mL.

Characterization Methods for As-prepared GOMs.

As prepared GO membranes were characterized by Scanning electron microscopy (SEM, ZEISS Gemini 300), X-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha), and X-ray diffraction analysis (XRD, Rigaku, Smartlab) spectra were utilized to analyze the functional groups of the samples and chemical properties of the membrane. Atomic Force Microscope (AFM, Bruker Dimension Icon) was used to measure GO flakes. The separated products were detected by using a penetration curve analyzer (Merck KGaA), ion chromatography (Metrohm) and gas mass spectrometry (BSD-mass).

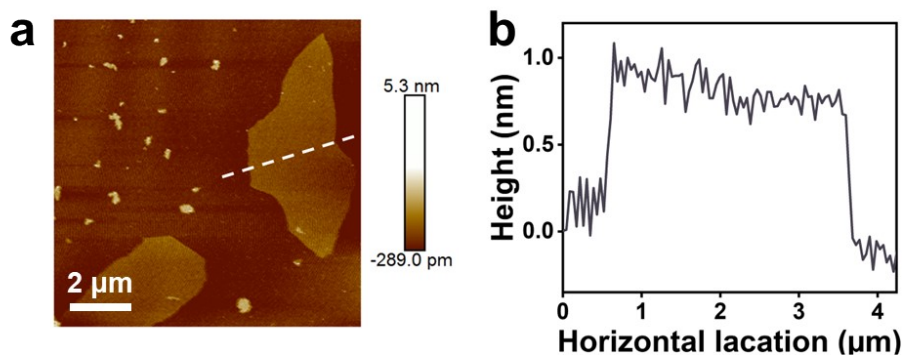


Fig. S1 (a) AFM image of the rGO and the corresponding height profile of pieces.

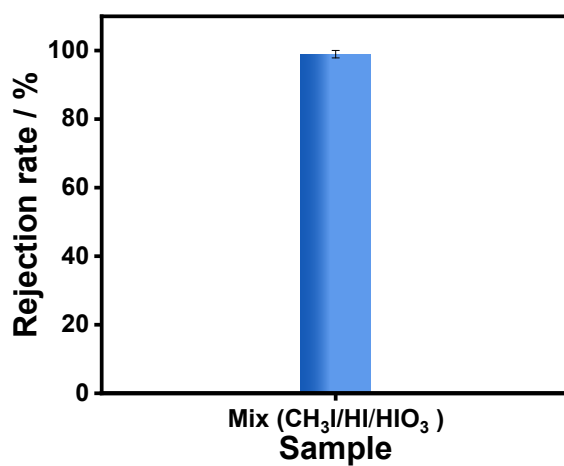


Fig. S2. Retention rates of the rGO-280 membrane in the gas of CH₃I, HI, and HIO₃.

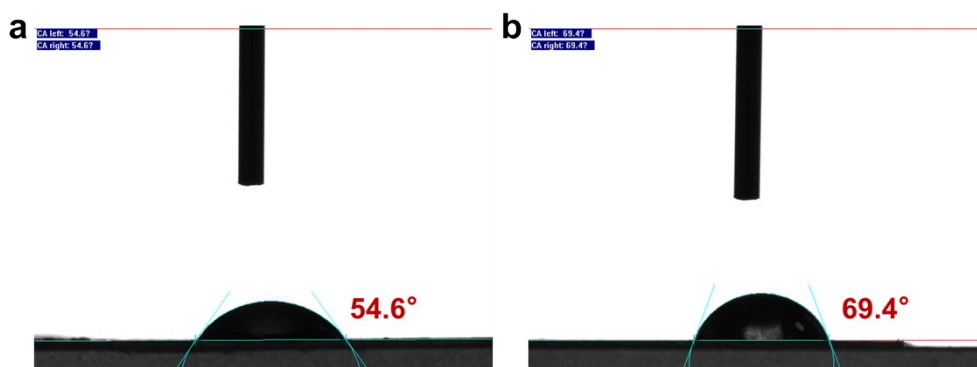


Fig. S3 (a) The water contact angle measurements for GO membranes. (b) The water contact angle measurements for rGO membranes.

Table S1. Comparison the gas permeance of different membranes in the CH₃I/He gas.

Sample	Permeance ($\times 10^{-6}$ mol m ⁻² s ⁻¹ Pa ⁻¹)
BCT	11.1
GO-280	6.50
rGO-220	6.21
rGO-280	5.29
rGO-300	4.26

Table S2. The molecular dimensions of different types of gaseous iodides.

Sample	Molecular dimensions
CH ₃ I	elongated shape, with I-C bond length ~ 2.14 Å
HI	linear, H-I bond ~ 1.61 Å
HIO ₃	tetrahedral-like, with O-I bonds ~ 1.78 Å