Supporting Information

Efficient separation of Mg^{2+}/Li^{+} using reduced GO membranes modified by positively charged arginine

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1. Experimental procedures and Characterizations

1-1. Preparation of graphene oxide suspensions.

Graphene oxide nanosheets were prepared using a modified Hummers method. Briefly, 5 g of 8000 mesh graphite powder was added to 12 mL of mixed solution (12 mL of H_2SO_4, 2.5 g of K_2SO_4, and 2.5 g of P_2O_5) and mixed homogeneously in a water bath at 80 °C for 5 h to obtain the raw material of pre-oxidized graphite. Then dried under vacuum at 60 °C overnight. The resulting pre-oxidized graphite powder was gradually added to a mixed solution of 120 mL concentrated sulfuric acid and 15.0 g potassium permanganate, and the mixture was stirred at 60 ℃. The stirring was continued at room temperature for an additional 2 hours. The solution was then slowly diluted with 250 mL deionized water in an ice-water bath, ensuring the temperature remained around 80 °C throughout the process. After stirring for another 2 hours, 20 mL of 30% hydrogen peroxide was added. The mixture was left to stand overnight. The precipitate was subsequently dissolved in ultrapure water, and the resulting solution was washed sequentially with a 1:10 aqueous hydrochloric acid solution and deionized water. Finally, the solution was sonicated and stirred for 0.5 hours each and allowed to stand for 7 days.

1-2. Characterizations

The surface and cross-sectional morphology of Arg-rGO membrane were observed by scanning electron microscopy (SEM) (Nova Nano SEM 450, USA) at an accelerating voltage of 3 kV; Raman scattering analysis was performed using a laser with a wavelength of 532 nm (Jobin Yvon HR800); The distribution of membrane elements and functional groups was analyzed by XPS (Thermo Fisher ESCALAB 250Xi spectrometer); The variation of layer spacing was analyzed by XRD using a Rigaku D/max 2550 VB/PC powder diffractometer. The radiation source used to determine the interlayer spacing of graphene oxide (GO) membranes is a copper target (Cu Kα) with a wavelength of $\lambda \approx 1.54$ Å. The scanning angle (2θ) range is 5°–40°.

The pretreatment steps of the samples in different characterization are as followings:

XRD: The membranes were completely dried in a vacuum oven at 60 °C and then uniformly spread on the sample holder for testing.

SEM: The membranes were fully dried and adhered to the copper substrate using carbon tape (surface morphology). The membranes were quenched in liquid nitrogen, fractured, and then fixed on the copper substrate (cross-sectional morphology). Both samples were sputter-coated with a 5 nm gold layer to enhance conductivity.

XPS: The samples were pressed into pellets and pre-cleaned with argon plasma to remove surface contaminants.

1-3. Economic and Environmental Assessment of Arg-rGO Membranes

Below is a table of the economic cost composition of raw materials for the preparation of arginine-crosslinked graphene oxide (Arg-rGO) filtration membranes. The data is based on laboratory-scale production and assumes current market prices (in Chinese Yuan, RMB):

Raw Material/Reagent	**Function**	**Unit Price (RMB/g)**	**Typical Usage (m^2)**	**Total Cost (RMB/m^2)**

Graphene Oxide (GO)	Membrane matrix material	80–150	1–2 g	80–300
Arginine	positive charge supplier	0.5–1.0	0.5–1.0 g	0.25–1.00
EDC	Carboxyl activator (crosslinking reaction)	20–30	0.3–0.5 g	6–15
NHS	Enhances crosslinking stability	15–25	0.2–0.4 g	3–10
Deionized Water/Buffer (PBS)	Reaction solvent	0.01–0.05	500–1000 mL	0.5–5.0
Organic Solvent (e.g., Ethanol)	Washing, post-processing	0.5–1.0	100–200 mL	0.5–2.0
Support Membrane (MCE)	Filtration substrate	10–20	1 piece (0.1 m²)	100–200

Total Cost Estimate: At laboratory scale, the raw material cost per square meter of Arg-rGO membrane is approximately 190–532 RMB (excluding equipment, energy, and labor costs). GO and EDC are the primary cost drivers (60%–80% of total cost).

Reducing their usage or finding alternatives is a key optimization strategy.

Scalability Potential: Industrial-grade GO prices can drop to 30–50 RMB/g (through bulk production); EDC/NHS consumption can be reduced via recycling or continuous-flow reactions; Support membranes can be replaced with cheaper porous materials (e.g., cellulose membranes). The high flux and reusable cleaning performance of Arg-rGO membranes reduce pumping energy consumption (saving 20%–30%), resulting in lower long-term operational costs compared to traditional membranes.

In addition, the Arg-rGO membrane has a high selectivity for Mg^{2+}/Li^{+} separation, which can effectively improve the recovery rate of lithium, reduce resource waste, and typically operates at a lower pressure, resulting in lower energy consumption compared to traditional separation techniques such as evaporation crystallization. However, the preparation of GO involves the use of chemical reagents such as concentrated sulfuric acid and potassium permanganate, which need to be properly treated to avoid environmental pollution.

Conducting a combined Life Cycle Assessment (LCA) and Techno-Economic Assessment (TEA) to evaluate the environmental and economic performance of the membrane, it can be concluded that the Arg-rGO membrane demonstrates significant environmental and economic potential in separation technologies.

2. Support Information Pictures

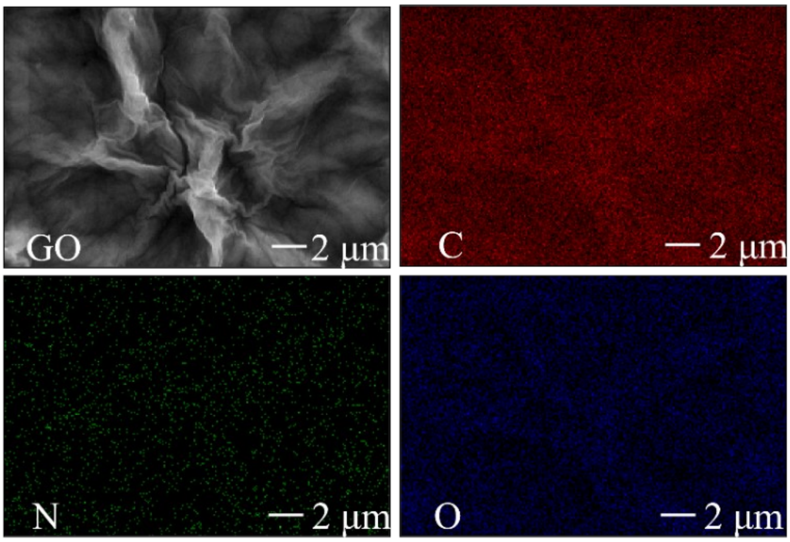

Figure S1. Elemental mapping of the GO membrane

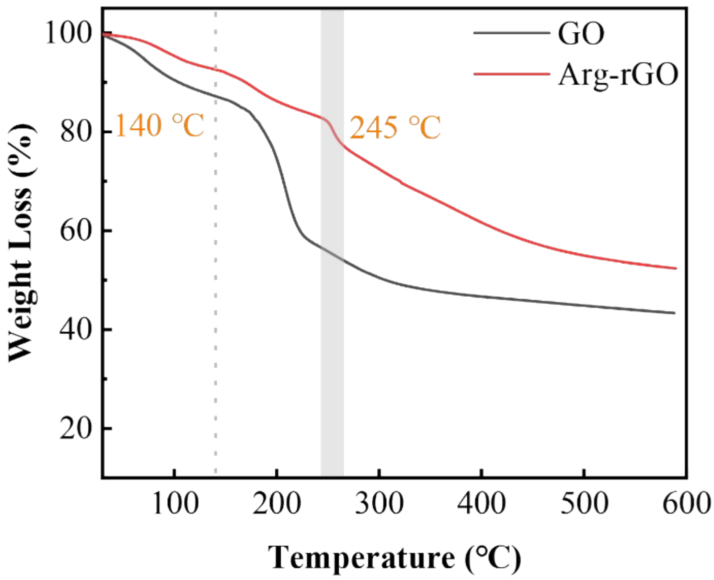

Figure S2. TGA of the GO and Arg-rGO membrane

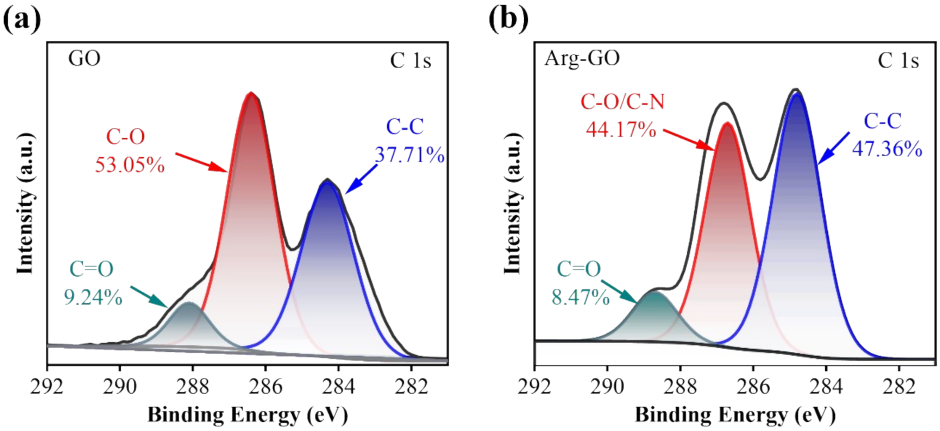

Figure S3. (a) The XPS C 1s spectra of the GO membrane; (b) The XPS C 1s spectra of the Arg-GO membrane

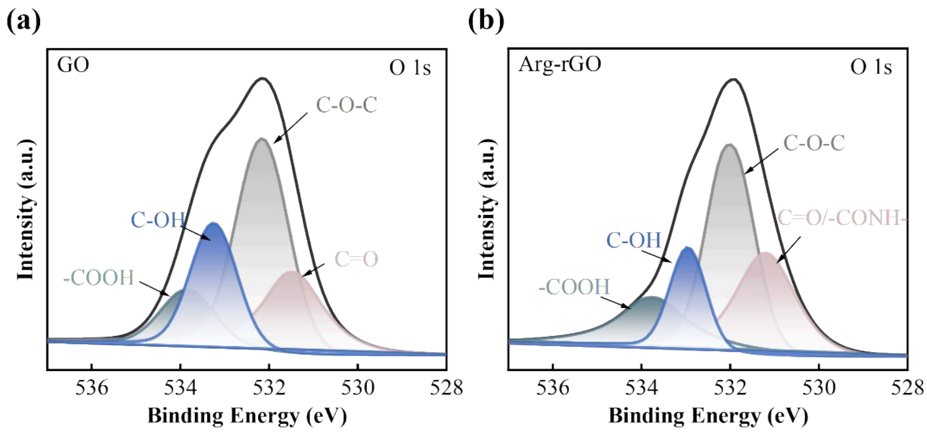

Figure S4. (a) The XPS O 1s spectra of the GO membrane; (b) The XPS O 1s spectra of the Arg-rGO membrane

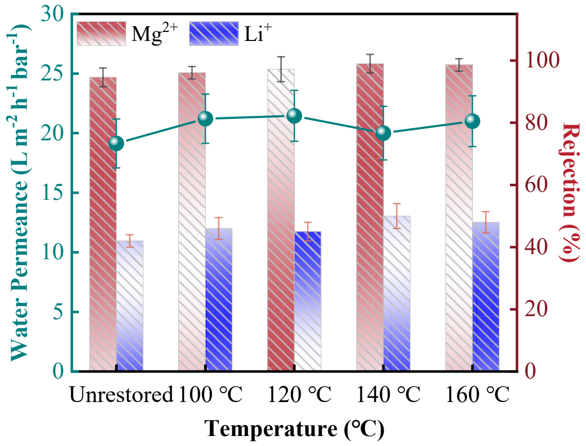

Figure S5. Rejection rate of the Arg-rGO membranes for lithium-magnesium mixed solutions with different reduction temperatures

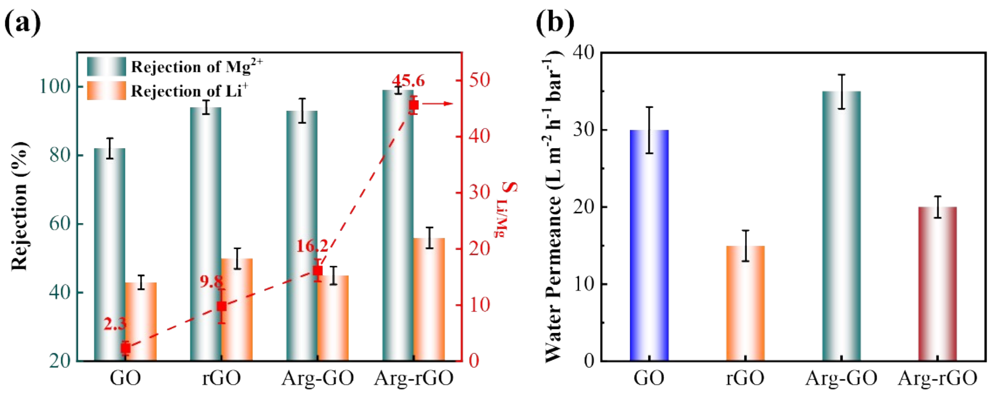

Figure S6. Comparation of the separation performance of the GO, rGO, Arg-GO, and Arg-rGO membranes

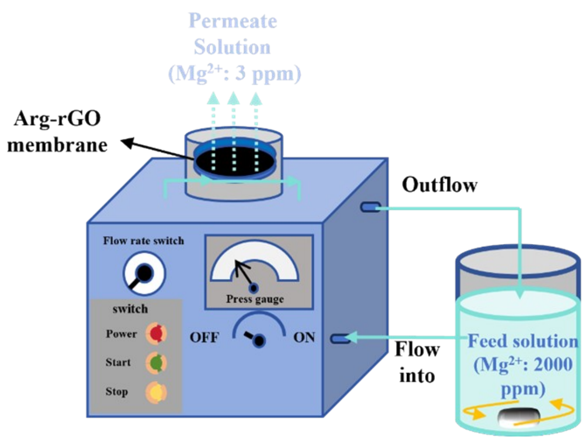

Figure S7. Cross-flow experimental setup

3. Table S1. Reported separation performance of different nanofiltration membranes

Membranes	Feed concentration (mg L^{-1})	$S_{Li/Mg}$	Permeance ($L\ m^{-2}\ h^{-1}\ bar^{-1}$)	Mg^{2+}/Li^{+} mass ratio^{+}	Ref
Nanofiltration membrane with RIP	2000	9.2	13.0	20	S1 [1]
PEI-4A-B15C5-TMC	1000	17.4	7.7	/	S2 [2]
QSPIP-TMC freestanding membrane	1000	12.0	23.0	/	S3 [3]
PEI@15C5-TMC	1500	14.0	8.0	50	S4 [4]
PA-TA-Cu	2000	26.5	4.87	20	S5 [5]

$(GO_{-0.002\%})$ /PIP/TMC	/	29	3.6	35	S6 [6]
GEM-TMC	1000	15.4	19.2	100	S7 [7]
PEI/TMC nanofiltration membrane	2000	13.0	6.2	20	S8 [8]
PSF-UF	2000	12.4	9.2	150	S9 [9]
PEI-GO/MXene	1000	5.7	2	/	S10 [10]
Arg-rGO membrane	**300**	**45.6**	**21.3**	**20**	**This Work**

4. References

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