

# Surface-Tension-Governed Nanoconfinement for Tunable Hydrogen-Isotope Transport

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Interfacial forces and nanoconfined environments play crucial roles in transport phenomena within low-dimensional materials. In this study, we propose a photothermally regulated surface-tension mechanism for hydrogen-isotope fractionation, in which light-induced changes in the surface tension of gallium (Ga) dynamically modulate the confined environment between graphene oxide (GO) layers. Under near-infrared irradiation, the surface tension of Ga decreases, altering its wettability within the interlayer galleries and inducing a reversible contraction of the GO interlayer spacing. *In situ* temperature-dependent transmission electron microscopy confirmed this dynamic modulation, showing that confined Ga nanoparticles undergo fully reversible expansion-contraction and display surface-energy-driven extension behavior analogous to macroscopic particles. Such confinement-induced structural responsiveness regulates the local arrangement of water molecules, leading to distinct molecular aggregation states for H<sub>2</sub>O and D<sub>2</sub>O. Due to stronger intermolecular attraction, D<sub>2</sub>O diffusion is more restricted in confinement, leading to pronounced isotope selectivity. Experimentally, the membrane achieved a separation factor of 70.53 for H<sub>2</sub>O:D<sub>2</sub>O and demonstrated selective removal of tritium water (T<sub>2</sub>O). Our study elucidates the interplay between photothermally driven interfacial energy modulation and confined molecular transport, offering new insights into tunable separation processes and interfacial physics.

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## I. INTRODUCTION

Efficient separation of hydrogen isotopes (protium H, deuterium D, and tritium T) plays a critical role in nuclear-fusion fuel cycles [1–3]. Tritium, although extremely scarce in nature, can be effectively obtained through specific pathways in nuclear fuel cycles, such as fission reactions and nuclear-waste recycling [4–7]. Therefore, how

to efficiently and energy-efficiently separate tritium, deuterium, and hydrogen is a key factor in the development of nuclear-fusion energy technology. The separation of hydrogen isotopes is not only crucial for the feasibility of nuclear-fusion reactions but also forms the basis for advancing clean-energy transformation [8]. However, existing hydrogen-isotope separation technologies, such as cryogenic distillation and electrolysis, face challenges such as high energy consumption, low selectivity, and poor scalability, which limit their application in nuclear-fusion energy and related fields [7,9–14].

To address these challenges, the development of hydrogen-isotope separation technologies with low energy consumption and high selectivity is particularly important. In recent years, membrane separation technologies based on two-dimensional (2D) materials, such as graphene oxide (GO), have emerged as promising

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solutions to the isotope separation problem due to their unique nanochannels and tunability [15–23]. The layered structure of 2D materials provides narrow molecular channels, where molecular transport is strictly controlled by size exclusion and intermolecular interactions, effectively amplifying the diffusion-rate differences between isotopes such as deuterium and hydrogen, thus achieving efficient separation [15,24–29]. This advancement offers the potential for low-energy sustainable hydrogen-isotope separation, paving the way for the widespread application of future nuclear-fusion energy and supporting global clean-energy transformation.

However, a significant challenge in existing 2D membrane technologies remains: how to adjust the size of these nanochannels precisely and dynamically to further enhance the selective separation efficiency of hydrogen isotopes [30–32]. In this study, we propose a novel hydrogen-isotope separation mechanism based on photothermal effects. By combining graphene oxide with liquid gallium (Ga) to form a photothermally responsive composite membrane (PRCM) system, the surface tension of liquid gallium undergoes reversible changes under near-infrared (NIR) irradiation, thereby adjusting the interlayer spacing of graphene oxide [33–35]. This dynamic modulation driven by photothermal effects allows us to precisely control the structure of the nanoconfined channels, thus regulating the molecular-transport behavior within these confined channels. Notably, due to the differences in hydrogen bonding strength, deuterium oxide ( $D_2O$ ) and protium oxide ( $H_2O$ ) exhibit distinct diffusion behaviors in these confined channels, enabling efficient isotope-selective separation.

This work provides a new controllable physical framework for hydrogen-isotope separation, integrating photothermal-modulation, nanoconfined-transport, and isotope-fractionation mechanisms, with broad application prospects. This technology offers a low-energy efficient solution for isotope separation in nuclear-fusion fuel cycles, while also enabling new approaches to nuclear-wastewater treatment and clean-energy development, thereby contributing to global energy transformation and climate-change mitigation.

## II. METHODS

### A. Ga metal dispersion

First, we placed the metal Ga in an oven (at about  $50^\circ C$ ) to melt it into a fluid state, and then we took a certain amount of metal Ga and transferred it to a centrifuge tube. Then, a certain volume of mixed solutions with ratios of ethanol:acetic acid = 1:2, acetic acid:water = 1:1, hydrochloric acid:water = 1:1, and ammonia:water = 1:1 was used to perform ultrasonic disruption and dispersion on metallic gallium until the solution had changed color

and become uniformly dispersed. The supernatant after ultrasound was taken out and set to volume for later use.

### B. Preparation of PRCMs

First, freeze-dried GO powder and liquid-metal Ga were individually dispersed in solvent to obtain stock suspensions at a concentration of  $0.5\text{ g} \cdot \text{L}^{-1}$ . Using these stock suspensions, a pure GO dispersion (GOS), a pure Ga dispersion (GaS), and a series of photothermal-responsive composite solutions (PRCSs) were prepared. The PRCSs were formulated with a fixed GO content while varying the mass ratios of GO to Ga ( $m_{GO}:m_{Ga} = 2:1, 1:1, \text{ and } 1:2$ ), designated as PRCS-1, PRCS-2, and PRCS-3, respectively. All dispersions were subsequently subjected to vacuum-assisted filtration to fabricate the corresponding photothermal-responsive composite membranes (PRCM-1, PRCM-2, and PRCM-3). For comparison, a pure GO membrane (GOM) and a pure Ga membrane (GaM) were also prepared. Membranes with different thicknesses were obtained by adjusting the filtration volumes of the GO dispersion to 1, 3, 5, 7.5, and 10 mL.

### C. Prototype-simulation experiment

We prepared two rectangular GOMs. First, one GOM was fixed on a glass slide and Ga was dropped in liquid state on the GOM. Then, another piece of GOM was placed on the Ga droplet and fixed on the left side to the contact point of the underlying GOM. Under NIR light, different temperatures, different Ga droplet sizes, and different upper GOM qualities, images were taken to observe the changes in the angles of the droplets and the upper GOM.

### D. *In situ* variable-temperature transmission electron microscopy

*In situ* transmission electron microscopy (TEM) heating experiments were performed using the Talos 200 $\times$  TEM operating at 200 kV, equipped with a dedicated Protochips Fusion 350 *in situ* heating holder. The sample was prepared by ultrasonically dispersing Ga along with GO nanosheets in an ethanol–acetic acid mixture to minimize oxidation. A small amount of the dispersion was dropped onto a heatable TEM grid, and Ga nanoparticles encapsulated by GO were identified on the grid to best simulate the state of Ga between GO layers. During the test, the temperature was raised to 25, 40, 60, and  $80^\circ C$ , and images were recorded at each stable temperature. This heating-cooling cycle was repeated 4 times. The projected particle area was quantified from TEM images based on threshold segmentation (excluding areas  $<500\text{ nm}^2$ ) using the ImageJ software package. For each temperature point, four independent measurements (corresponding to four cycles) were obtained, and additional measurements at  $25^\circ C$  were recorded after cycling to confirm reversibility. The raw

data and statistical analysis are provided in Table S2 in the Supplemental Material [98].

### E. Ion-filtration experiment

First,  $\text{LiCl}$ ,  $\text{MgCl}_2 \cdot 2\text{H}_2\text{O}$ ,  $\text{CaCl}_2$ , and  $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$  were prepared into 5 ml  $5 \times 10^{-4}$  M single-ion solutions using deionized water. Then, through negative-pressure filtration, the PRCM was used at 0.8 bar to carry out a retention experiment on 5 ml of single-ion solution. The filtrate was taken for concentration testing using an atomic absorption spectrometer, and then the permeability was calculated according to [36,37]

$$P_{ct}(\%) = \frac{C}{C_0}, \quad (1)$$

where  $P_{ct}$  is the transmittance (in percent),  $C$  is the concentration of single ions in the filtrate, (in  $\text{mol l}^{-1}$ ), and  $C_0$  is the initial concentration of the single-ion solution (in  $\text{mol l}^{-1}$ ).

### F. Ion-permeation kinetics experiment

We prepared 50 ml of  $5 \times 10^{-4}$  M  $\text{LiCl}$  ion solution. Using a separation-osmosis device, the original ion solution was placed in the left separation tank, the deionized water was placed in the right separation tank, and the separation membrane was sandwiched between the two separation tanks. The infiltration was carried out under different temperature conditions (5, 22, 30, 50, and 80 °C). We took 1-ml amounts of the solutions on the left and right sides at 0.5, 1, 2, 3, 6, 9, and 12 h, respectively, and the concentration of the solution on the right was tested using inductively coupled plasma optical emission spectrometry (ICP-OES). The test results were entered into the following equation [38,39] to calculate the dynamic permeation:

$$P = \frac{C(V_0 - V_{n-1})}{A}, \quad (2)$$

where  $P$  is the permeability per unit area (in  $\text{mol m}^{-2}$ ),  $C$  is the solution test concentration (in  $\text{mol l}^{-1}$ ),  $V_0$  is the initial volume of the separation liquid (in liters),  $V_{n-1}$  is the volume of the removed solution (in liters) and  $A$  is the effective separation area (in  $\text{m}^2$ ).

### G. Water-flux test

Using PRCMs, a certain volume (0.5 ml, 1 ml, and 1.5 ml) of  $\text{H}_2\text{O}$  solution or  $\text{D}_2\text{O}$  solution was filtered at a suction pressure of 0.8 bar, and the filtration time was recorded. The temperature was adjusted to approximately 5 °C using an ice bath, and NIR light was used to change the membrane temperature and adjust the temperature on

the membrane to about 50 °C. Then, the water flux was calculated according to [36]

$$WF = \frac{V}{A \times t \times \Delta P}, \quad (3)$$

where  $WF$  is the water flux (in  $\text{l m}^{-2} \text{h}^{-1} \text{bar}^{-1}$ ),  $V$  is the volume of water filtered (in liters),  $A$  is the effective filtering area (in  $\text{m}^2$ ), and  $\Delta P$  is the filtration pressure value (in bar).

## III. RESULTS AND DISCUSSION

### A. Preparation and characterization of PRCMs

Since metallic Ga is highly prone to oxidation, we first optimized the dispersion conditions of metallic Ga (Figs. S1 and S2 in the Supplemental Material [98]). Finally, a mixed dispersion with a volume ratio of ethanol to acetic acid of 1:2 was selected for the dispersion of metallic Ga, and ethanol was used as the dispersant to disperse GO before membrane preparation (Scheme S1 in the Supplemental Material [98]). By fixing the content of GO and varying the Ga doping amount, we prepared PRCMs with different ratios (PRCM-1, PRCM-2, PRCM-3). For comparison, a pure GO membrane (GOM) and a pure Ga membrane (GaM) were also prepared. First, dispersions of GO and metallic Ga mixed in different ratios were prepared, and it was found that the introduction of metallic Ga could weaken the Tyndall effect of the GO solution [Fig. 1(a)]. The zeta-potential results show that the dispersion containing GO has a certain electronegativity, and the dispersion stability of colloidal particles deteriorates with the increase of metallic Ga [Fig. 1(b)]. After being fabricated into membranes, as shown in Fig. 1(d), the color of the membrane gradually darkens with the increase in the content of metallic Ga. The SEM surface images [Figs. S3(a), S3(d), and S3(e) in the Supplemental Material [98]] show that with the introduction of metallic Ga, a large number of Ga particles are also distributed on the membrane surface, with sizes ranging from approximately 0.25 to 4  $\mu\text{m}$ . Compared with the cross-section image of the GOM [Fig. 1(f)], the cross-section SEM characterization of PRCM-2 [Fig. 1(g) and Fig. S3(g) in the Supplemental Material [98]] shows that flat metallic Ga particles are dispersed between the GO layers. Pure GaM is deposited into a blocklike membrane [Fig. S3(f) in the Supplemental Material [98]]. Elemental distribution analysis using SEM revealed a relatively low oxygen content in the Ga distribution areas [Fig. 1(g) and Fig. S3(g) in the Supplemental Material [98]], indicating that the metal Ga dispersed with acetic acid and that ethanol was resistant to oxidation. However, the relatively large metallic Ga particles between the layers may affect the flatness of the interlayer channels. Due to the high surface tension of metallic Ga, the difference in binding force between internal and external atoms, and the presence of a dense oxide

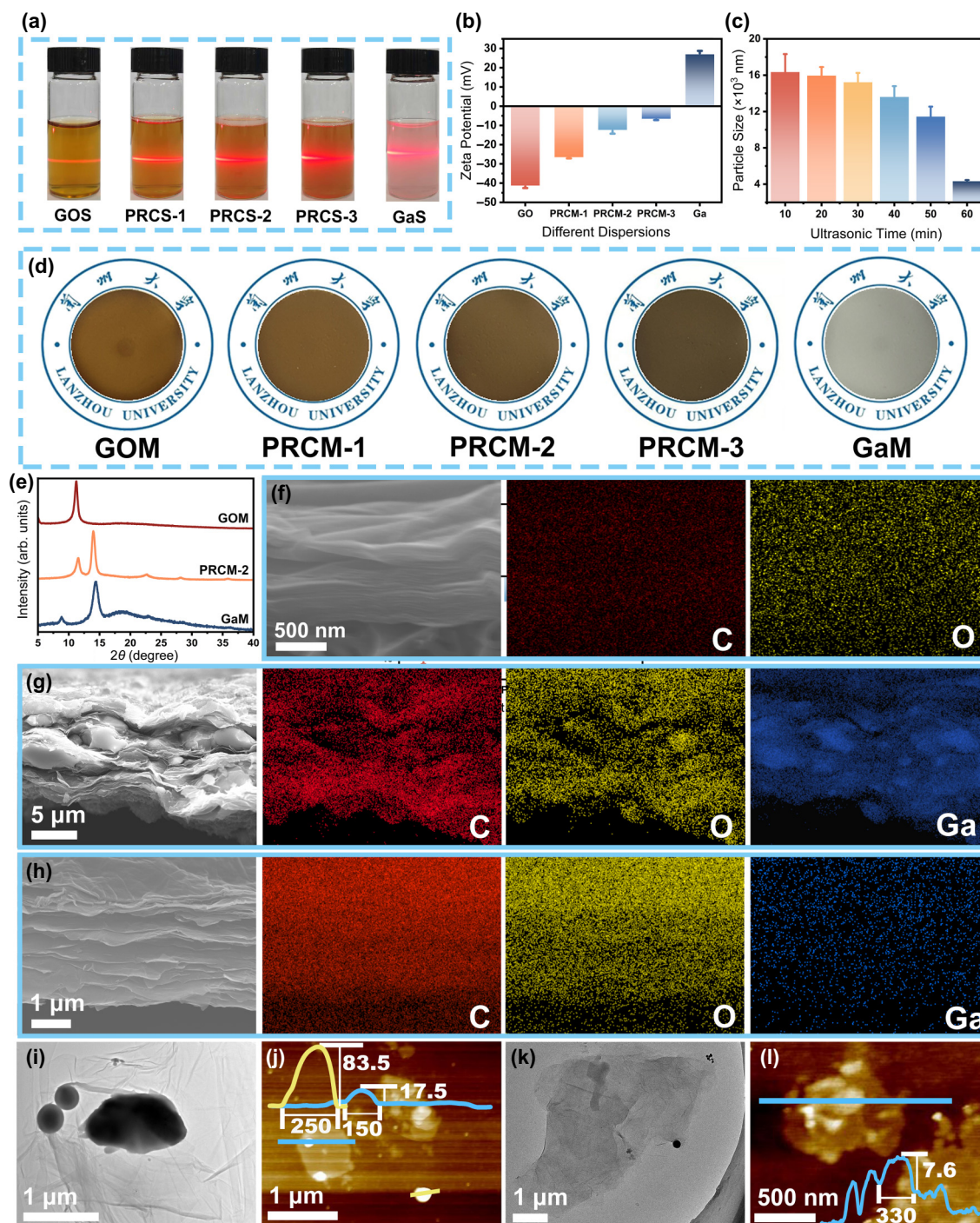


FIG. 1. The characterization of photothermally responsive composite membrane (PRCM) morphology. (a) The Tyndall effect of dispersions with different ratios: GOS is the GO dispersion, PRCs-1 is the dispersion with  $m_{GO}:m_{Ga} = 2:1$ , PRCs-2 is the dispersion with  $m_{GO}:m_{Ga} = 1:1$ , PRCs-3 is the dispersion with  $m_{GO}:m_{Ga} = 1:2$ , and GaS is the Ga dispersion. (b) The zeta-potential diagram of dispersions with different ratios. (c) The particle size of metallic Ga dispersion with different ultrasonic times. (d) Digital images of membranes with different ratios: fixed  $m_{GO} \sim 2.5$  mg. (e) XRD of GOM, PRCM-2 with improved ultrasonic dispersion of metallic Ga, and GaM. (f) The SEM cross-section characterization and energy-dispersive spectroscopy (EDS) element distribution of C and O of GOM. (g) The SEM cross-section characterization and EDS element distribution of C, O, and Ga of PRCM-2. (h) The SEM cross-section characterization and EDS element distribution of C, O, and Ga of PRCM-2 with improved ultrasonic dispersion of metallic Ga. (i) The transmission electron microscopy (TEM) characterization of PRCM-2. (j) The atomic-force-microscopy (AFM) characterization of PRCM-2: the curves in the figure are the cross-section height diagrams at different positions. (k) The TEM characterization of PRCM-2 after heating. (l) The AFM characterization of PRCM-2 after heating: the curve in the figure is a cross-section height diagram.

layer on its surface, dispersing it into smaller particles requires the involvement of greater energy [40,41]. For this reason, we further processed the metallic Ga dispersion by increasing the power and duration of ultrasonic disruption, thereby making the metallic Ga particles smaller during the disruption process [Fig. 1(c)]. Then, it was mixed with GO and filtered into a membrane. From the SEM cross section, the cross-section morphology of the improved PRCM-2 is consistent with that of pure GOM [Figs. 1(f) and 1(h)], showing a “booklike” stacking. Additionally, the distributed Ga can also be observed in the energy-dispersive spectroscopy (EDS) elemental distribution. Meanwhile, by comparing the XRD characterizations of GOM, PRCM-2, and GaM [Fig. 1(e)], it can also be confirmed that metallic Ga exists between the membrane layers. Due to the improvement in the dispersion method, Ga was inserted into the GO interlayers in a tinier or flakelike form.

We then characterized the structure of the membrane. The XRD results [Fig. S4(a) in the Supplemental Material [98]] show that the characteristic peak of the GO (001) crystal plane is located at  $10.28^\circ$ , and the characteristic peaks of metallic Ga are located at  $8.78^\circ$ ,  $14.24^\circ$ ,  $22.88^\circ$ ,  $28.41^\circ$ ,  $36.05^\circ$ , and  $43.39^\circ$ . With the increase of the content of metallic Ga particles, the intensity of its GO characteristic peak gradually weakened, and the peak related to metallic Ga became more obvious; in particular, when  $m_{\text{GO}}:m_{\text{Ga}} = 1:2$  (PRCM-3), the characteristic peak of metallic Ga appeared. Raman spectroscopy [Fig. S4(b) in the Supplemental Material [98]] indicates that increasing Ga content has a minimal effect on GO defects, as evidenced by the weak variation in the  $I_D:I_G$  ratio. Fourier-transform infrared (FT-IR) spectroscopy [Fig. S4(c) in the Supplemental Material [98]] reveals more pronounced characteristic peaks for free O—H groups at  $3660\text{ cm}^{-1}$  and Ga—O bonds at  $674$  and  $632\text{ cm}^{-1}$ . Due to the ethanol solution used to disperse metallic Ga, as the amount of metallic Ga increases, the amount of free O—H in the ethanol solution that is not affected by C—H increases, so the characteristic peaks of O—H and Ga—O are enhanced. XPS characterization [Fig. S4(d) in the Supplemental Material [98]] confirms Ga doping, with the appearance of Ga  $2p$  peaks at  $1145$  and  $1118\text{ eV}$  and Ga  $3d$  peaks at  $20$  and  $26\text{ eV}$  in the spectra. Thermogravimetric (TG) characterization [Fig. S4(e) in the Supplemental Material [98]] shows greater mass loss with lower Ga content below  $300^\circ\text{C}$ . The stress-strain curves [Fig. S4(f) in the Supplemental Material [98]] demonstrate that the addition of metallic Ga has little effect on the tensile strength and toughness of the membrane. Finally, adsorption-desorption curves [Figs. S4(g)—S4(l) in the Supplemental Material [98]] reveal that compared to GOM, PRCM has a smaller specific surface area and lacks a regular pore distribution.

## B. Intelligent control of PRCMs

Since metal Ga is plastic under temperature control [33,41–54], it was introduced into the interlayer of GO to leverage this property for regulating the interlayer spacing. We performed XRD characterization of the membrane at various temperatures ( $5$ ,  $25$ ,  $40$ ,  $60$ , and  $80^\circ\text{C}$ ). In XRD characterization, the characteristic peak of the GO (001) crystal plane can be used to calculate the change in interlayer spacing. The results show that as the temperature increases, the GO (001) characteristic peak shifts from  $9.44^\circ$  to  $12.06^\circ$  [Fig. 2(a)]. As shown in Fig. S5 in the Supplemental Material [98], heating the PRCM from room temperature ( $25^\circ\text{C}$ ) to  $80^\circ\text{C}$  causes the characteristic peak to shift rightward, indicating a decrease in interlayer spacing. After the membrane was allowed to stand at approximately  $20^\circ\text{C}$  for  $20\text{ min}$ , the characteristic peak partially recovered to  $10.76^\circ$ , demonstrating the elastic recovery of the interlayer spacing to its original state upon cooling. Based on the XRD characterization data in Fig. 2(a), we calculated the interlayer spacing of the GO membrane at different temperatures and plotted the interlayer spacing against temperature [37,55]. A power-function curve was fitted to the data, yielding the equation  $y = 10.66x^{-0.08}$  [ $R^2 = 0.9924$ , Eq. (4)] [Fig. 2(b)]. This function allows accurate prediction of the temperature needed to achieve a specific interlayer spacing, enabling precise control of size-selective screening channels.

For PRCMs with different ratios, we studied the changes in interlayer spacing at different temperatures (Fig. S6 in the Supplemental Material [98]). Although GOM exhibits a slight shift, GO is reduced when the temperature reaches  $80^\circ\text{C}$ , and its characteristic peak of the (001) crystal plane disappears—the characteristic peak does not recover after cooling [Fig. S6(a) in the Supplemental Material [98]]. When a small amount of Ga is introduced (PRCM-1), the (001) characteristic peak still disappears at  $80^\circ\text{C}$ , but recovers after cooling for a period of time [Fig. S6(b) in the Supplemental Material [98]]. When the Ga and GO contents are consistent (PRCM-2), the (001) characteristic peak shifts with temperature and returns to the initial state after cooling for a period of time [Fig. 2(a) and Fig. S5 in the Supplemental Material [98]]. With the increase of Ga content (PRCM-3), the characteristic peak of GO (001) shifted without reduction at  $80^\circ\text{C}$  and gradually recovered after cooling [Fig. S6(c) in the Supplemental Material [98]]. According to the variation trend of the interlayer spacing and the characteristic peaks, the introduction of metallic Ga reduces the reduction degree of GO and improves the range of interlayer regulation. When the content of metallic Ga and GO is consistent (PRCM-2), the interlayer-regulation range and reversible performance are optimal [Fig. S6(d) in the Supplemental Material [98]]. To explore the impact of oxygen content on interlayer

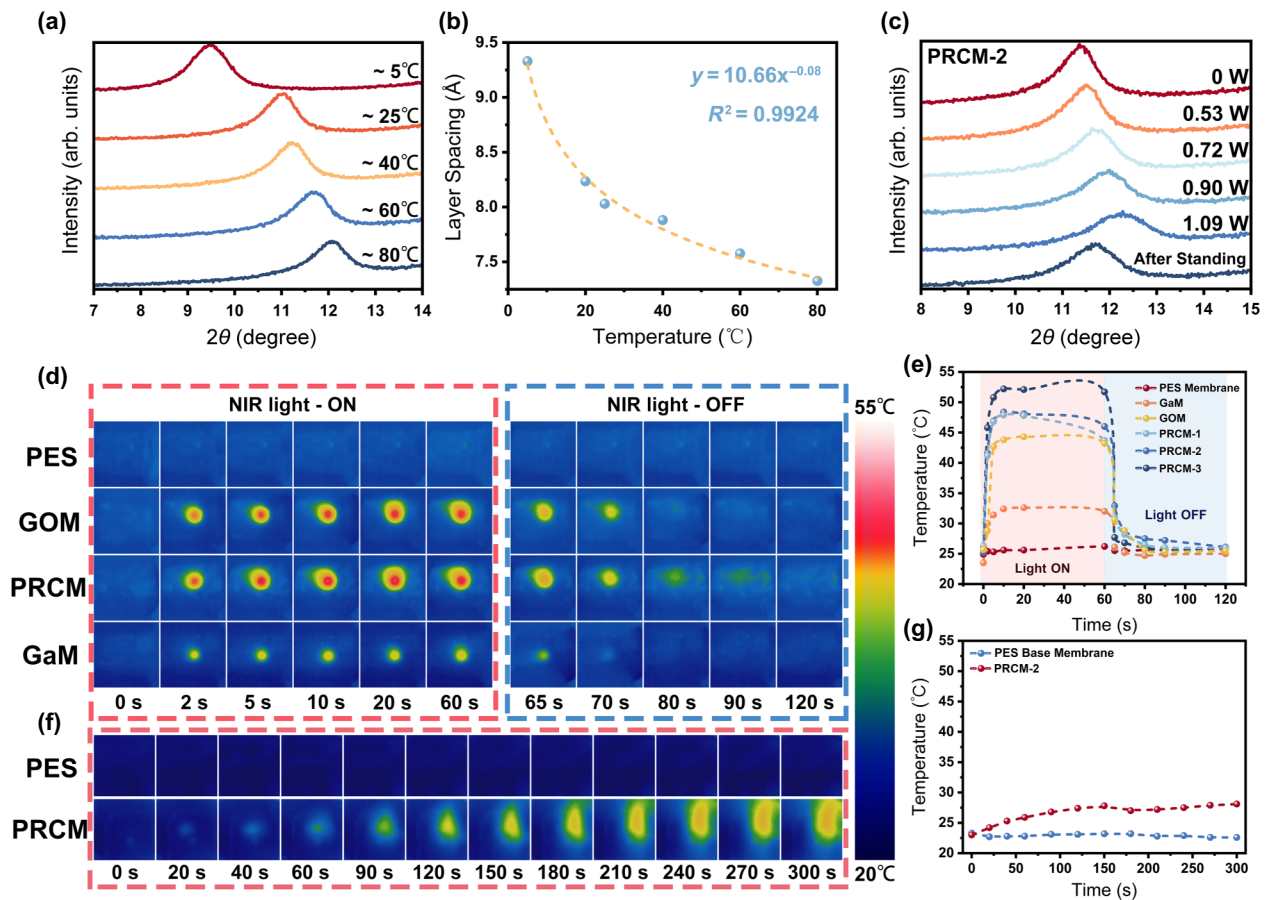
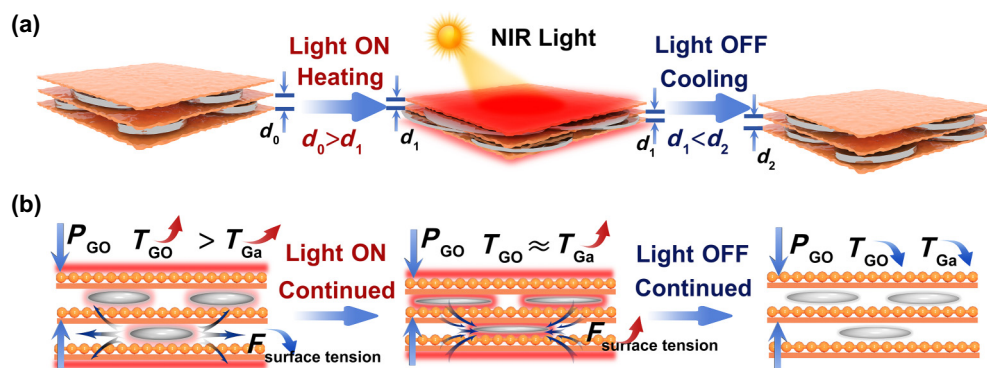


FIG. 2. PRCM interlayer-spacing regulation. (a) The XRD characterization of PRCM-2 at different temperatures. (b) The layer-spacing–temperature-fitting curve. (c) The XRD characterization of PRCM-2 under different power illuminations. (d) The thermal imaging of different membranes under 808-nm near-infrared (NIR) light: 0–60 s, light ON; 60–120 s, light OFF; power approximately  $0.88 \text{ W cm}^{-2}$ . (e) The temperature variation of different membranes under 808-nm NIR light over time: 0–60 s, light ON; 60–120 s, light OFF; power approximately  $0.88 \text{ W cm}^{-2}$ . (f) The thermal imaging of different membranes in water under 808-nm NIR light: power approximately  $2.70 \text{ W cm}^{-2}$ . (g) The temperature-variation curves of different membranes in water under 808-nm NIR light: power approximately  $2.70 \text{ W cm}^{-2}$ .

spacing, we reduced the GO to prepare PRCMs with different oxygen contents: approximately 52.14%, 51.59%, and 50.91%, respectively (the reduction temperatures and the corresponding oxygen contents are listed in Table S1 in the Supplemental Material [98]). The XRD characterization and calculated interlayer-spacing results showed that although reduced oxygen content decreases interlayer spacing, the overall trend remains unchanged [Fig. S7 in the Supplemental Material [98]]. These findings demonstrate that Ga plays a crucial role in the intelligent regulation of interlayer spacing, enabling greater flexibility in tuning the membrane properties.

GO is an excellent photothermal material, and is widely used in medicine, energy storage, and intelligent control [34,56,57]. To address energy-consumption concerns, we utilized the photothermal properties of GO to regulate interlayer spacing through temperature changes under light exposure [Scheme 1(a)]. We conducted illumination

experiments on different membranes (PES-based membrane, GOM, PRCM-1, PRCM-2, PRCM-3, and GaM) using 808-nm NIR light to observe the temperature changes over time [as shown in Figs. 2(d) and 2(e) and Fig. S8 in the Supplemental Material [98]]. The PES-based membrane showed no photothermal effect, while GOM, PRCM, and GaM exhibited excellent photothermal performance. As the Ga content in PRCM increased, higher temperatures were generated. The temperature stabilized after 10 s of illumination and returned to the initial state quickly after the light was turned off. For membranes with a fixed Ga content (2.5 mg) and varying GO content (1.25, 2.5, and 5 mg), a higher GO content resulted in increased temperatures, which also stabilized after 10 s and quickly returned to the initial state upon light removal (Figs. S9 and S10 in the Supplemental Material [98]). Thermal-imaging experiments were conducted on various membranes under different power



SCHEME 1. (a) A schematic diagram of the PRCM interlayer structure changes with light illumination. (b) A schematic diagram of the interlayer Ga particle structure of PRCMs changing with light illumination.

levels (Figs. S11–S16 in the Supplemental Material [98]). The PES-based membrane showed no temperature change [Figs. S11 and S17(a) in the Supplemental Material [98]], while GOM, PRCMs, and GaM displayed excellent photothermal performance, with temperatures increasing as the power increased [Figs. S12–S16 and S17(b)–S17(f) in the Supplemental Material [98]]. The temperature-power curves showed that higher total GO and Ga content resulted in steeper slopes, indicating enhanced photothermal effects (Fig. S18 in the Supplemental Material [98]). Thermal-stability tests demonstrated that all membranes maintained good photothermal stability across different power levels (Fig. S19 in the Supplemental Material [98]). To assess photothermal performance in solution, we placed PRCM-2 in an aqueous solution and irradiated it with a laser. PRCM-2 generated more heat than the base membrane under the same power, and heat transfer became more pronounced with increasing power [Figs. 2(f)–2(g) and Figs. S20 and S21 in the Supplemental Material [98]]. These results confirm that PRCM retains its photothermal effect in solution.

Next, we used a prototype-simulation method to place Ga droplets between two GOM layers and observed changes in the upper GOM under 808-nm NIR light. Since the light irradiated the back side of the upper GOM, we reexamined the temperature change on the back side of PRCM-2 under different power levels. Compared to front-side irradiation, the temperature increased more slowly but could still reach the desired level at sufficient power (Figs. S22 and S23 in the Supplemental Material [98]). In the prototype simulation, the length and width of the Ga droplet were fixed while its height was varied. Shortly after turning on the light, the angle of the upper GOM changed significantly, recovering gradually after the light was turned off. Greater droplet height resulted in more pronounced angle changes (Figs. S24 and S25 in the Supplemental Material [98]). This confirms that PRCM can achieve intelligent interlayer regulation through photothermal response. Further, we performed

XRD characterization, which showed significant shifts in the characteristic peak of the GO (001) crystal plane, indicating notable changes in interlayer spacing under different power levels [Fig. 2(c)]. Comparing the interlayer-spacing variation trends of different membranes under light-controlled conditions, PRCMs with Ga intercalation demonstrated a broader and more controllable range of interlayer spacing, offering distinct advantages, and the interlayer-spacing adjustment is reversible (Fig. S26 in the Supplemental Material [98]).

### C. PRCM interlayer intelligent control mechanism and size screening function

The prototype simulation and XRD results confirmed that PRCMs can regulate interlayer spacing under photothermal control. Due to the thermal sensitivity of Ga, we investigated whether Ga in different membranes undergoes phase transitions. Differential scanning calorimetry (DSC) results indicated potential solidification at high Ga content (PRCM-3) [Fig. 3(c)]. XRD analysis revealed distinct peaks for solid and liquid Ga [Fig. S27(a) in the Supplemental Material [98]], but when Ga was in the liquid state, its peaks remained stable across temperatures [Fig. S27(b) in the Supplemental Material [98]]. To explore this further, we conducted heating and cooling cycles on the PRCM-3 membrane. The XRD results showed that while the characteristic peak of GO shifted, the peak of Ga remained unchanged, confirming that Ga between PRCM layers remained in a liquid state with no phase transitions [Fig. 3(d)].

The morphological changes occur based on the dependence of the Ga surface tension on temperature and the effect of surface tension on wettability [58–61]. After small metallic Ga particles are introduced between the GO layers, they are flattened under the action of interlayer pressure [62]. As the temperature rises, the surface tension of Ga decreases and its wettability increases, causing it to spread and deform between the GO layers, thereby

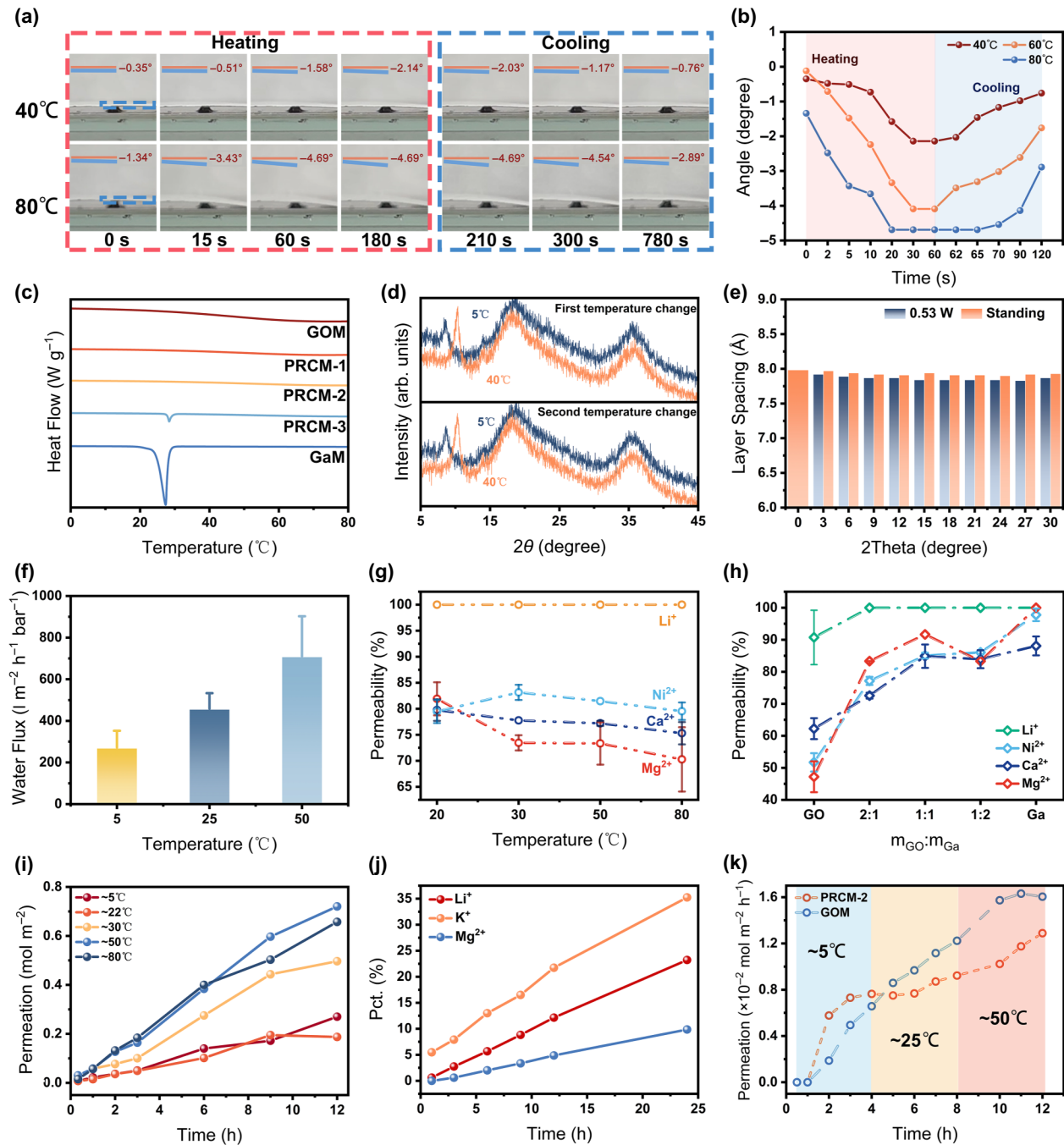


FIG. 3. The intelligent control mechanism and investigation of the performance of interlayer spacing. (a) Digital images of the prototype simulation of interlayer-spacing control by Ga droplets and GOMs at different temperatures: 0–180 s, heating; 180–600 s, cooling; the angle measurement in the figure is to fix the glass of the lower GOM as the horizontal line (red line) and measure the angle of the upper GOM (blue line); the angle test range is marked with a blue dotted box. (b) The angle of the upper GOM changes with time during the prototype simulation of regulating the interlayer spacing: 0–180 s, heating; 180–600 s, cooling. (c) The differential scanning calorimetry (DSC) characterization of different membranes. (d) The XRD characterization of PRCM-3 after two cycles of heating and cooling. (e) A study on the interlayer-spacing cycle of the photothermal response of PRCM-2. (f) The water flux of PRCMs at different temperatures: PRCM-2,  $m_{GO} \sim 0.5$  mg,  $n = 3$ . (g) The ion permeability of PRCMs at different temperatures under filtration conditions: PRCM-2,  $m_{GO} \sim 3.75$  mg,  $n = 3$ . (h) The ion permeability of PRCMs with different ratios under filtration conditions:  $m_{GO} \sim 3.75$  mg,  $n = 3$ . (i) The kinetics of Li<sup>+</sup> at different temperatures under permeation conditions. (j) The kinetics of different ion permeabilities under normal-temperature permeation conditions. (k) The kinetics of Li<sup>+</sup> with GOM and PRCM-2 under variable-temperature permeation conditions.

reducing the interlayer spacing; the surface tension of Ga increases and its wettability decreases, causing it to shrink and restore its original shape, thereby increasing the interlayer spacing [Scheme 1(b)] [35,63]. This regulation process relies on the surface-tension–temperature relationship described by Eötvös’s law [64], providing a reversible and controllable mechanism for the dynamic regulation of interlayer spacing. The role of the surface tension of metallic Ga was confirmed with the help of macroscopic simulations, and macroscopic prototype simulations were carried out under different conditions (temperature, metallic Ga size, and GOM quality) [Figs. 3(a) and 3(b) and Figs. S28–S31 in the Supplemental Material [98]]. The metallic Ga droplets were flattened under pressure between two pieces of GOM. Temperature and droplet size significantly affect the surface tension of Ga, resulting in a significant change in the angle of the upper GOM. When the surface tension of Ga is constant, its GOM quality is less affected. Larger droplets exhibit higher surface tension and weaker angle changes, while smaller droplets (lower surface tension) show more pronounced changes. Especially at higher temperatures, the surface tension is smaller and the angle change is more obvious. We also performed TEM-morphology and atomic-force-microscopy (AFM) morphology characterization on the membranes after the heating treatment. The TEM-morphology and EDS-element-distribution characterization before heat treatment showed that, compared with GO [Fig. S3(b) in the Supplemental Material [98]], there was flattened and deformed metallic Ga between the GO nanosheets [Fig. 1(i) and Fig. S34(a) in the Supplemental Material [98]]. AFM characterization revealed that the width of the metallic Ga particles between the layers of PRCM-2 was approximately 150 nm, with a thickness of about 17.5 nm, while the exposed metallic Ga particles had a width of about 250 nm and a thickness of around 83.5 nm [Fig. 1(j)]. The differences in width and thickness between exposed Ga particles and interlayer Ga particles confirmed that the interlayer Ga particles were flattened. After photothermal-response treatment, the TEM-morphology and EDS-element-distribution characterization of PRCM-2 showed that the GO nanosheets were covered with thin metallic Ga flakes [Fig. 1(k) and Fig. S34(b) in the Supplemental Material [98]]. In the AFM characterization, the GO nanosheets were at the micrometer scale, with a thickness of approximately 3.14 nm [Fig. S3(c) in the Supplemental Material [98]]. After photothermal-response treatment, metallic Ga flakes with a width of about 300 nm and a thickness of around 7.6 nm appeared on PRCM-2 [Fig. 1(l)].

To directly verify the reversible expansion of Ga within confined GO interlayers, we conducted *in situ* temperature-dependent TEM experiments on Ga nanoparticles encapsulated by GO nanosheets. EDS spectra [Figs. 4(a) and 4(c)] confirmed the elemental composition of the encapsulated nanoparticles. Representative TEM images [Figs. 4(b)

and 4(d)] clearly show a systematic increase in projected particle area upon heating ( $25 \rightarrow 40 \rightarrow 60 \rightarrow 80^\circ\text{C}$ ), followed by almost complete recovery upon cooling to  $25^\circ\text{C}$ . Quantitative analysis of two representative particles demonstrated consistent and reproducible behavior over four heating-cooling cycles: particle 1 expanded from  $838.35 \pm 5.54 \text{ nm}^2$  at  $25^\circ\text{C}$  to  $929.04 \pm 19.47 \text{ nm}^2$  at  $80^\circ\text{C}$  (+10.82%) and recovered to 100.16% of its initial area upon cooling, while particle 2 expanded from  $663.51 \pm 7.10 \text{ nm}^2$  to  $742.95 \pm 7.80 \text{ nm}^2$  (+11.97%) and recovered to 98.13% of its initial area [Figs. 4(e) and 4(f) and Table S2 and Figs. S32 and S33 in the Supplemental Material [98]]. These *in situ* observations provide direct microscopic evidence that Ga nanoparticles undergo fully reversible expansion and contraction driven by temperature-dependent surface tension. Importantly, this microscopic reversibility is fully consistent with ensemble-level results, including reversible interlayer-spacing shifts detected by XRD [Figs. 2(a) and 3(e)] and reversible modulation observed in macroscopic prototype simulations [Figs. 3(a) and 3(b) and Figs. S28–S31 in the Supplemental Material [98]]. Together, these multiscale characterizations provide strong support for the proposed surface-tension-governed reversible nanoconfinement mechanism. After photothermal excitation, the interlayer Ga in PRCM-2 exhibits temperature-dependent surface-tension variations, leading to surface-energy–dominated dynamic behavior that reduces the interlayer spacing. Finally, cyclic XRD measurements confirmed the stability of this reversible modulation over 30 heating-cooling cycles [Fig. 3(e) and Fig. S35 and Movie S1 in the Supplemental Material [98]].

Interlayer channels significantly affect the transport properties of ions and molecules. Therefore, we conducted water-flux and ion-permeation experiments at different temperatures. Since the experimental environment is affected by the swelling of water molecules, here we first investigated the interlayer-regulation performance of the membrane in the swollen state. After intercalation of ions of different sizes ( $\text{Li}^+$ ,  $\text{Ni}^{2+}$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$ ), different temperatures are regulated. The results show that the swollen membrane can still achieve effective regulation of the interlayer spacing under photothermal response (Fig. S36 in the Supplemental Material [98]). As shown in Fig. 3(f), the water fluxes at 5, 20, and  $50^\circ\text{C}$  are approximately 271.76, 458.69, and  $710.80 \text{ l m}^{-2} \text{ h}^{-1} \text{ bar}^{-1}$ , respectively, exhibiting a clear increase with rising temperature. This trend appears inconsistent with the observation that the interlayer spacing of the PRCMs decreases as temperature increases. The apparent contradiction can be resolved by considering the ordering of the interlayer channels. It has been reported that in 2D layered membranes, greater interlayer order correlates with enhanced mass transport capacity [65]. The order of GO-based 2D layered membranes can be assessed by the full width at

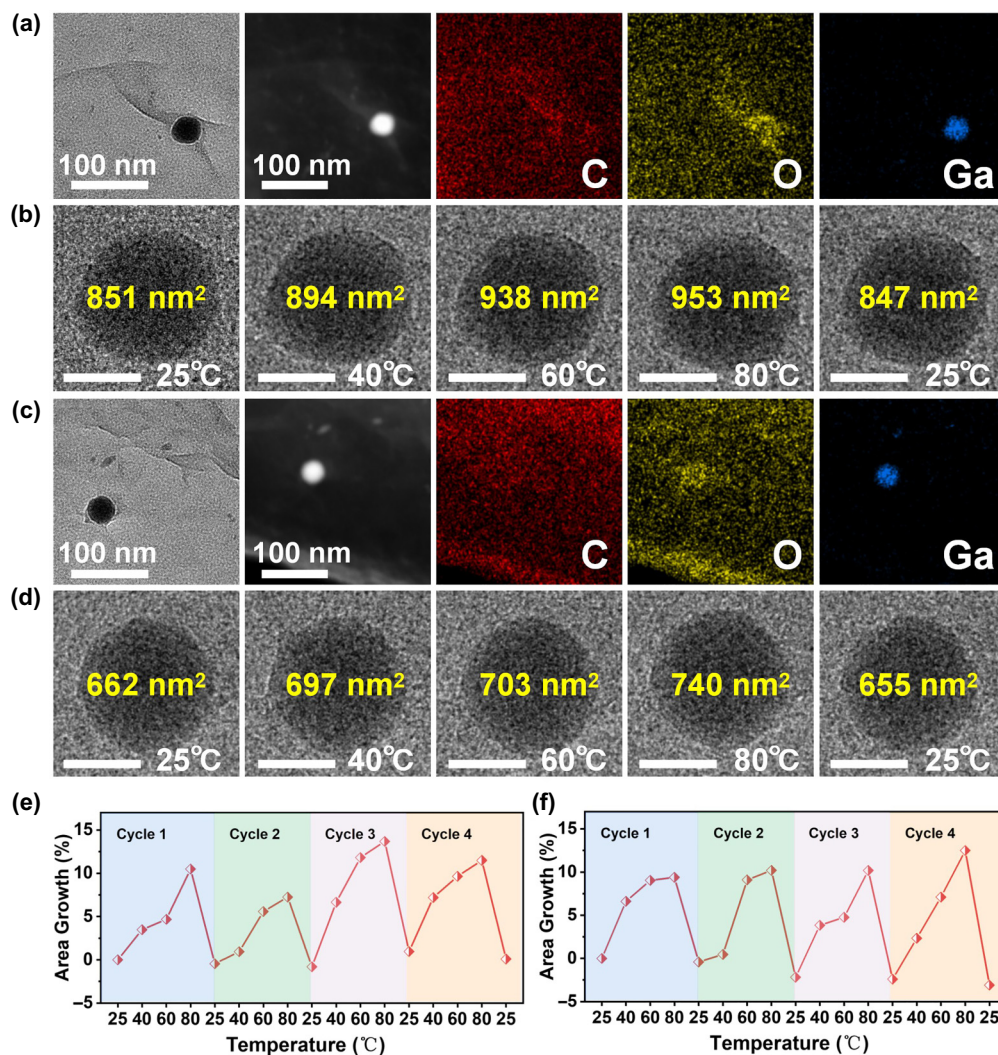


FIG. 4. (a)–(d) *In situ* temperature-dependent TEM analysis of Ga nanoparticles encapsulated by graphene oxide nanosheets. The EDS elemental mapping of representative Ga nanoparticles (a) 1 and (c) 2. Representative TEM images showing the reversible change in the projected area of nanoparticles (b) 1 and (d) 2 during a single heating-cooling cycle (25 → 40 → 60 → 80 → 25 °C). The scale bar represents 20 nm. (e),(f) The percentage increase in the projected area of the representative Ga nanoparticles (e) 1 and (f) 2 during four consecutive heating-cooling cycles (25 → 40 → 60 → 80 → 25 °C).

half maximum (FWHM) of the GO (001) XRD diffraction peak. XRD analysis shows that the FWHM of the GO (001) peak in PRCM systematically narrows with increasing temperature, from 0.93790 (5 °C), 0.76549 (20 °C), 0.77139 (40 °C), and 0.77694 (60 °C) to 0.66746 [80 °C, Fig. 2(a)]. This progressive narrowing indicates that the nanochannels become increasingly uniform and smooth upon heating. These findings suggest that while the introduction of metallic Ga particles enlarges the interlayer spacing, it also distorts the interlayer channels, particularly at low temperatures, where this effect is more pronounced. Such irregularity severely impedes water transport between the layers, thereby reducing the overall flux [66,67], while low temperatures themselves further

suppress molecular dynamics [68]. Upon heating, however, the interlayer spacing decreases due to Ga diffusion between GO nanosheets, driven by the temperature-dependent reduction in Ga surface tension. This surface-energy-driven dynamic behavior improves the smoothness and continuity of the interlayer channels, reducing bottlenecks and promoting more efficient water transport [36]. Thus, even though the average interlayer spacing contracts, the channel-smoothing effect enhances the effective permeability of the water molecules. In addition, elevated temperatures increase the mobility and diffusion coefficient of the water molecules, further accelerating their passage through the membrane [68]. For comparison, pristine GOMs exhibited far lower fluxes under identical

conditions, with only approximately 10.52, 12.93, and 15.11  $\text{l m}^{-2} \text{h}^{-1} \text{bar}^{-1}$  at 5, 20, and 50 °C, respectively (Fig. S37 in the Supplemental Material [98]). Although the flux of both GOMs and PRCM-2 increased with temperature, the enhancement was much more pronounced in PRCM-2, confirming the critical role of Ga in dynamically regulating interlayer channels. Taken together, these synergistic effects reconcile the apparent paradox between reduced interlayer spacing and increased water flux and highlight the key role of Ga-mediated surface-tension modulation in governing the dynamic interlayer behavior.

Next, we investigated the relationship between interlayer spacing and ionic radius under filtration conditions using metal ions of different hydration diameters:  $\text{Li}^+$ ,  $\text{Ni}^{2+}$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$ . Based on Eq. (4), the temperatures corresponding to the hydration diameters of  $\text{Li}^+$  (7.6 Å),  $\text{Ni}^{2+}$  (8.1 Å),  $\text{Ca}^{2+}$  (8.2 Å), and  $\text{Mg}^{2+}$  (8.6 Å) were calculated as 68.67, 33.63, 26.56, and 14.65 °C, respectively [69]. Negative-pressure filtration experiments were then conducted at 20, 30, 50, and 80 °C [device shown in Fig. S38(b) in the Supplemental Material [98]]. The results showed that  $\text{Li}^+$  transmittance remained at 100%, while the transmittance of  $\text{Ni}^{2+}$ ,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$  gradually decreased with increasing temperature. In addition, the ion permeability followed the order  $P(\text{Li}^+) > P(\text{Ni}^{2+}) > P(\text{Ca}^{2+}) > P(\text{Mg}^{2+})$ , consistent with their increasing hydration radii. These results suggest that the membrane intercepts ions larger than the interlayer spacing through temperature-regulated size sieving [Fig. 3(g)]. We further confirmed this mechanism by cross-section SEM and elemental mapping of membranes after mixed-ion filtration at different temperatures (Fig. S39 in the Supplemental Material [98]). Mg exhibited the lowest elemental signal, supporting the size-sieving effect. Moreover, ion transmittance increased with higher Ga loading [Fig. 3(h)], indicating that Ga incorporation loosens the interlayer channels, thereby enhancing permeability. In permeation experiments [device shown in Fig. S38(a) in the Supplemental Material [98]],  $\text{Li}^+$  permeability increased with temperature [Fig. 3(i)]. At room temperature, the permeability of different ions ( $\text{K}^+$ ,  $\text{Li}^+$ , and  $\text{Mg}^{2+}$ ) was inversely proportional to their hydration radius [Fig. 3(j)], further confirming size-selective separation. Finally, variable-temperature  $\text{Li}^+$  permeation experiments (5, 25, and 50 °C) showed distinct kinetics for PRCM-2 compared to pristine GO membranes [Fig. 3(k)]. For PRCM-2, permeability was highest at 5 °C, attributed to the larger interlayer spacing at low temperature. At 25 °C, the reduced interlayer channels slowed ion transport, while at 50 °C, continued channel contraction was offset by enhanced thermal mobility, again accelerating ion transport. These results demonstrate that temperature effectively regulates interlayer channels and thereby modulates ion transport.

#### D. PRCM performance inspection

Stability and selectivity are two of the most important properties of membrane materials in the separation process, so we investigated the stability and selectivity of the membrane at room temperature. The membranes with different ratios were immersed in aqueous solution for 14 days, and XRD structure characterization and SEM morphology characterization were performed before and after immersion (Fig. S40 in the Supplemental Material [98]). Comparison of XRD in Fig. S40(a) in the Supplemental Material [98] and the digital photographs before and after immersion in the inset shows that a large amount of GO falls off from the membrane before and after immersion, causing its interlayer spacing to change. The intensity of the XRD GO (001) crystal-plane characteristic peak was significantly weakened after shedding, indicating that the structural stability of the GOM was poor. On the other hand, as the Ga content increases, the shedding phenomenon disappears, and the characteristic peaks of GO in XRD shift slightly, which may be because the interlayer spacing is slightly expanded due to immersion in water, and the introduction of metallic Ga makes the color of the membrane gradually deepen in water. However, compared with GOM, its structure becomes more stable after adding metallic Ga. And in an environment with approximately 40% relative humidity, the interlayer spacing changes slightly and the structure is stable (Fig. S41 in the Supplemental Material [98]). However, the SEM surface-morphology and element-distribution results showed that compared with the membrane before immersion [Fig. S40(b) in the Supplemental Material [98]], the metal Ga on the surface of the membrane was oxidized after immersion [Fig. S40(c) in the Supplemental Material [98]], and the XRD characterization also showed that the increase in metal Ga led to the appearance of Ga-oxide-related peaks after immersion [Fig. S40(a) in the Supplemental Material [98]]. In addition, we investigated the stability of Ga between membrane layers and tested the Ga content in the filtered-water solution. The leaked Ga content did not exceed one ten-thousandth of the total content and its impact was relatively low (Fig. S42 in the Supplemental Material [98]). Then, we used PRCM to separate simulated salt-lake water at room temperature. The results show that the membranes with different ratios have little difference in ion permeability, and all have a certain separation effect on ions in a short time, but the separation factor decreases with time. The best separation effect in the experimental results was the PRCM with PRCM-3, where the separation factors for  $\text{K}^+:\text{Mg}^{2+}$ ,  $\text{K}^+:\text{Li}^+$ ,  $\text{K}^+:\text{Na}^+$ , and  $\text{Li}^+:\text{Mg}^{2+}$  reached 49.21, 9.07, 5.07, and 5.43, respectively, at 3 h (Fig. S43 in the Supplemental Material [98]). This shows that the membrane has a certain stability and has excellent separation performance for ions in a short time.

### E. PRCM hydrogen-isotope separation

Finally, we applied PRCMs to hydrogen-isotope separation, using the slight differences between hydrogen and deuterium water to separate them. In the water-flux test, we found that the membrane had a difference in flux between ordinary water and 10% deuterated water [Fig. 5(a)] and the separation performance was investigated using the water-flux ratio. We investigated the ratio of water flux for different thicknesses, different proportions, different abundances, and different volumes. The results show that at different thicknesses, the water flux decreases with increasing thickness, while the water-flux ratio is not much different. When using membrane separation with 1 ml of GO solution for filtration, the  $\text{H}_2\text{O}:\text{D}_2\text{O}$  water-flux ratio reached approximately 4.66 [Fig. 5(a)]. For membranes with different ratios, as the amount of metallic Ga increases, the water-flux ratio increases and the separation effect is enhanced. When the Ga content is the highest (PRCM-3), the water-flux ratio reaches approximately 3.02 [Fig. S44(a) in the Supplemental Material [98]]. This may be due to the excellent catalytic properties of metallic Ga, which improves the selectivity of the membrane during the separation process. We selected 1%, 5%, and 10% abundances of  $\text{D}_2\text{O}$  for water-flux testing. As the abundance increased, the difference in the water-flux ratio became greater. When the  $\text{D}_2\text{O}$  abundance was 10%, the water-flux ratio reached 5.36 [Fig. S44(b) in the Supplemental Material [98]]. When the abundance is greater, the difference between  $\text{H}_2\text{O}$  and  $\text{D}_2\text{O}$  is magnified, so the difference in water flux increases. When filtering different volumes of water, we found that the larger the volume of filtered water, the greater was the water-flux ratio. When the filtration volume was 1.5 ml, the water-flux ratio reached 5.66 [Fig. S44(c) in the Supplemental Material [98]]. This indicates that when the filtration volume increases, the filtration time increases, which effectively improves the ability of the interlayer channel to identify  $\text{H}_2\text{O}$  and  $\text{D}_2\text{O}$ , and can separate  $\text{H}_2\text{O}$  and  $\text{D}_2\text{O}$  with slight differences. At the same time, we also tested the water flux of 30%  $\text{D}_2\text{O}$  at different temperatures. The results were consistent with the conclusion of  $\text{H}_2\text{O}$ , and the water flux increased with increasing temperature [Fig. 5(b)]. We then used a membrane with a ratio of PRCM-2 to filter and separate 30%  $\text{D}_2\text{O}$  at different temperatures, at 5, 25, and 50 °C, its concentration dropped to approximately 20.50% (rejection rate approximately 31.67%), approximately 10.78% (rejection rate approximately 64.07%), and approximately 17.27% (rejection rate approximately 42.43%), respectively, and its separation factor [ $\text{SF}(\text{H}_2\text{O}:\text{D}_2\text{O})$ ] reached 3.55 at room temperature [Figs. 5(c) and 5(d)]. The conclusion shows that when the interlayer spacing increases, although the flux slows down, the slight difference between water and heavy water makes it difficult to achieve efficient separation. Under high temperature conditions, the

interlayer spacing decreases but the flux increases, and its selectivity decreases. A similar trend was observed for GOM: while both membranes exhibited temperature-dependent  $\text{H}_2\text{O}:\text{D}_2\text{O}$  transport [Figs. 5(b)–5(d)], the separation factor of PRCM-2 (3.55 at 25 °C) far exceeded that of GOMs (1.17 under the same conditions; Fig. S45 in the Supplemental Material [98]). Therefore, we separated  $\text{D}_2\text{O}$  with different abundances at room temperature. The results showed that the separation effect of  $\text{D}_2\text{O}$  at lower abundances of 5% (down to approximately 4.62%, rejection rate approximately 0.76%) and 15% (down to approximately 13.95%, rejection rate approximately 0.70%) was poor. It had a good separation effect for 60% and its abundance was reduced to approximately 17.53% ( $\text{SF} \approx 70.53$ ) [Figs. 5(e) and 5(f)]. As the concentration increased, the difference in water flux became more obvious, and it had excellent separation performance for high-concentration  $\text{D}_2\text{O}$  [Fig. 5(f) and Fig. S44(b) in the Supplemental Material [98]]. Moreover, GOMs failed to achieve comparable selectivity (optimal  $\text{SF} \approx 1.88$ ; Fig. S46 in the Supplemental Material [98]). To further examine the influence of Ga particle size on membrane performance, PRCMs were fabricated using Ga nanoparticles of varying diameters obtained by adjusting the ultrasonic power and duration during nanoparticle preparation [Fig. 1(c)]. Flux and isotope separation tests revealed a clear size-dependent trend: membranes prepared with smaller Ga nanoparticles exhibited lower overall water flux but significantly enhanced H:D separation factors. Notably, the membrane containing the smallest Ga nanoparticles achieved the highest SF of 3.45 (Fig. S47 in the Supplemental Material [98]). These results confirm that Ga particle size plays an important role in regulating the trade-off between flux and selectivity in PRCMs.

Afterward, we applied the membrane to the separation of tritium, and the results showed [Fig. 5(g)] that there was a certain removal rate for wastewater with different initial counts. The count dropped from 2.244 and 3.877 counts/min to 1.797 and 3.303 counts/min, respectively, and the removal rates reached 19.92% and 14.81%, respectively. We then conducted a kinetic study on the tritium-removal experiment and the removal rate was above 11% within 4 h [Fig. 5(h)]. This result proved that the membrane can separate hydrogen and deuterium while also removing tritium water. At the same time, we investigated the stability of the membrane in retaining tritium. Within 20 cycles, the rejection rate remained at around 13% [Fig. 5(i)], indicating that the membrane has excellent separation stability within a certain range. In the experiment, tritium water came from wastewater discharged from nuclear power plants. In real samples, the membrane showed excellent tritium-separation performance and maintained stable performance in multiple cycles, proving its application potential in reducing radioactive wastewater pollution and improving tritium recovery efficiency. This not

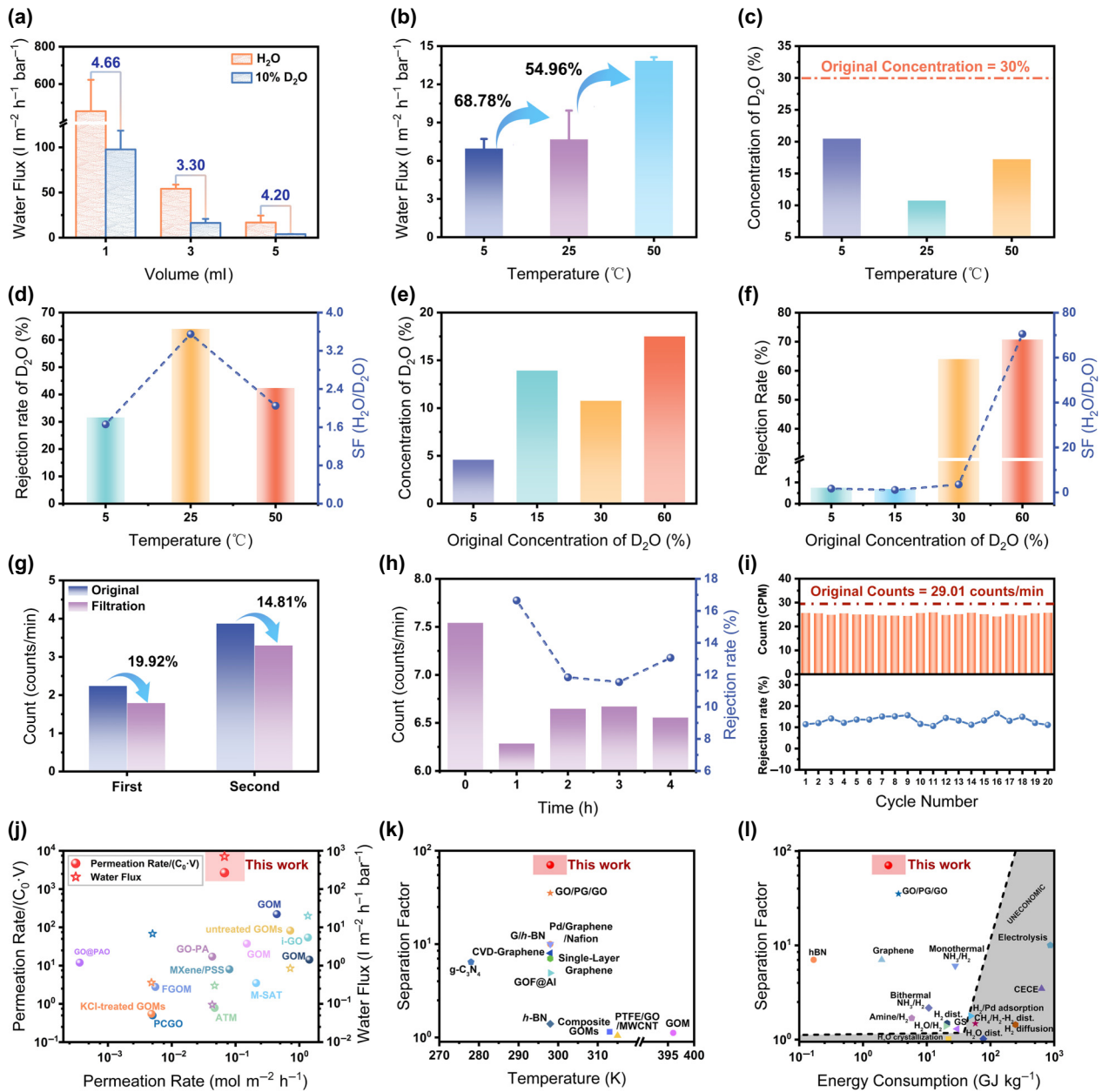


FIG. 5. Investigation of the performance of the PRCM separation of hydrogen, deuterium, and tritium water. (a) The water-flux test of  $\text{H}_2\text{O}$  and  $\text{D}_2\text{O}$  and the ratio of  $\text{H}_2\text{O}$  and  $\text{D}_2\text{O}$  water flux for membranes of different thicknesses: PRCM-2, the horizontal axis is the volume of  $0.5 \text{ g l}^{-1}$  GO dispersion taken when preparing the membrane,  $n = 3$ . (b) The water flux of 30%  $\text{D}_2\text{O}$  at different temperatures: PRCM-2,  $m_{\text{GO}} \sim 3.75 \text{ mg}$ ,  $n = 3$ . (c) The separation results of 30%  $\text{D}_2\text{O}$  at different temperatures using a suction filtration device: PRCM-2,  $m_{\text{GO}} \sim 3.75 \text{ mg}$ . (d) The rejection rate and separation factor of 30%  $\text{D}_2\text{O}$  at different temperatures. (e) The separation results of  $\text{D}_2\text{O}$  with different abundances at room temperature using a suction filtration device: PRCM-2,  $m_{\text{GO}} \sim 3.75 \text{ mg}$ . (f) The  $\text{D}_2\text{O}$  rejection rate and separation factor at different abundances. (g) Tritium-rejection experiments at different concentrations: PRCM-2,  $m_{\text{GO}} \sim 3.75 \text{ mg}$ . (h) The kinetic count and rejection rate of tritium removal by PRCM-2. (i) The counts and rejection rates of the tritium-rejection cycles: PRCM-2,  $m_{\text{GO}} \sim 3.75 \text{ mg}$ . (j) A comparison of the water flux and ion permeation rate of 2D membranes with other literature [70–80]. (k) A comparison of the separation performance of 2D membranes for hydrogen isotopes with other literature [15,19,24,81–88]. (l) A comparison of the performance and energy consumption of common hydrogen-isotope separation methods [19].

only helps to reduce the environmental impact of nuclear energy facilities, but also provides technical support for the efficient management of hydrogen isotopes in future fusion reactors.

We compared this work with similar work and found that the membrane has excellent water flux and ion flux [Fig. 5(j)] [70–80]. Compared with representative hydrogen-isotope separation membranes, the separation

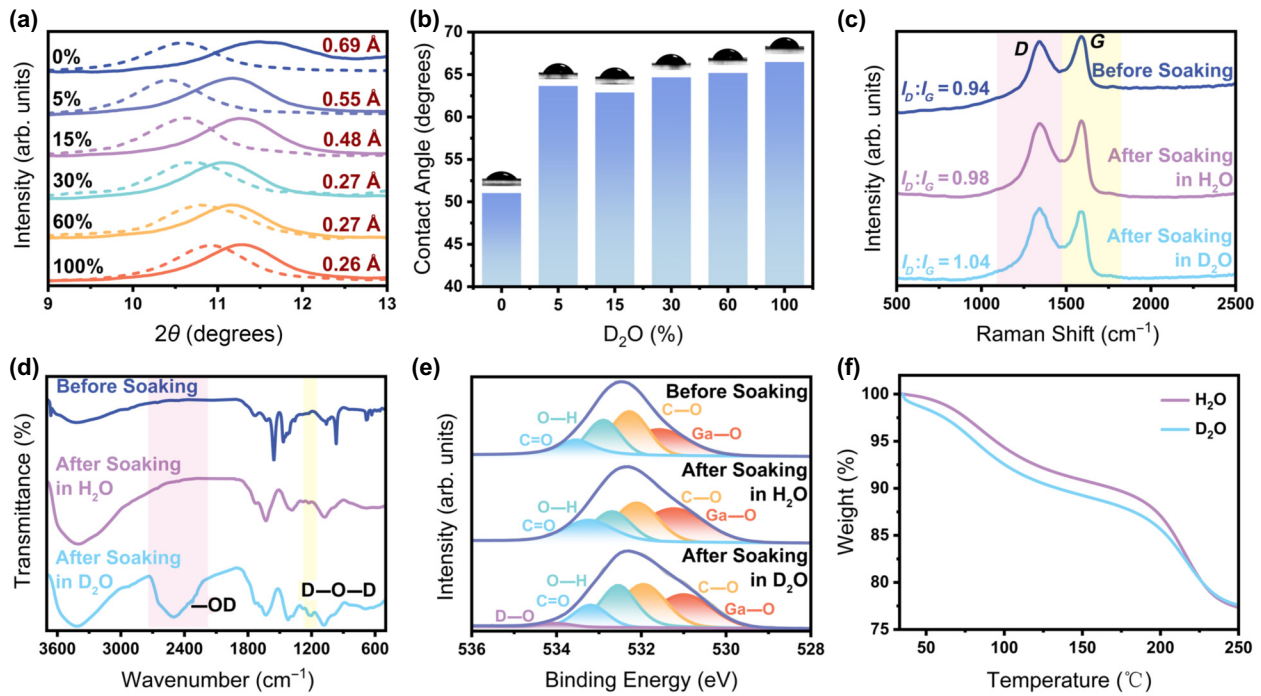


FIG. 6. A study on the mechanism of separation of hydrogen and deuterated water by PRCM. (a) An XRD comparison of PRCMs before and after immersion in  $D_2O$  with different abundances. (b) The contact angle of PRCM for  $D_2O$  with different abundances. (c) Raman spectra of the PRCM before and after immersion in  $H_2O$  and  $D_2O$ . (d) Fourier-transform infrared (FT-IR) spectroscopy of the PRCM before and after immersion in  $H_2O$  and  $D_2O$ . (e) The XPS O 1s fine spectrum of the PRCM before and after immersion in  $H_2O$  and  $D_2O$ . (f) The thermogravimetric (TG) change curve of the PRCM after immersion in  $H_2O$  and  $D_2O$ .

factor obtained, in the range of 270–400 K, demonstrates a clear advantage, based on comparisons with the best-performing values reported in similar studies [Fig. 5(k)] [15,19,24,81–88]. Additionally, we compared the energy consumption of our method with that of conventional hydrogen-isotope separation techniques and found that, while achieving acceptable separation selectivity, our approach exhibits significantly lower energy consumption [Fig. 5(l)] [19]. Compared with traditional separation technologies, this membrane provides a separation method with lower energy consumption while achieving high flux and high selectivity, thereby reducing the energy demand and environmental impact of the separation process. These characteristics make it an efficient, low-carbon, and sustainable separation technology in the nuclear-fusion fuel cycle, providing key support for the development of future clean energy.

The separation of  $H_2O$  and  $D_2O$  is inherently governed by differences in their physical properties. To investigate this mechanism, membranes were immersed in  $D_2O$  solutions of varying concentrations (0%, 5%, 15%, 30%, 60%, and 100%) for 2 h, followed by XRD analysis. The results showed a systematic decrease in interlayer spacing with increasing  $D_2O$  content [Fig. 6(a)]. Correspondingly, flux measurements revealed that  $H_2O$  permeated faster than  $D_2O$ , and the total flux decreased with increasing  $D_2O$

fraction (Fig. S48 in the Supplemental Material [98]), consistent with the XRD observations. This phenomenon can be explained by hydrogen-bond-mediated water clustering. Previous studies have reported that  $D_2O$  forms stronger hydrogen bonds than  $H_2O$ , resulting in more compact molecular association, smaller average cluster size, and stronger intermolecular cohesion [89]. Consequently,  $H_2O$  tends to form more extended water layers within the PRCM nanochannels, thereby facilitating faster mass transfer. Furthermore, stronger hydrogen bonding also increases the viscosity of  $D_2O$  relative to  $H_2O$ . Contact-angle measurements confirmed this effect: membranes exhibited larger contact angles at higher  $D_2O$  concentrations, reflecting higher viscosity [Fig. 6(b) and Fig. S49 in the Supplemental Material [98]]. Spectroscopic characterizations further substantiated these isotope effects. Raman spectroscopy showed that the change in the  $I_D/I_G$  ratio after  $H_2O$  immersion was weaker than after  $D_2O$  immersion, indicating a stronger structural influence of  $D_2O$  [Fig. 6(c)]. FTIR spectra of  $D_2O$ -immersed membranes displayed characteristic  $-OD$  and  $D-O-D$  stretching bands at 2505 and 1209  $cm^{-1}$ , respectively [Fig. 6(d)] [90]. XPS analysis revealed a D-O-related peak at 534 eV [Fig. 6(e)] [91]. In addition, thermogravimetric analysis demonstrated that below 250  $^{\circ}C$ ,  $D_2O$ -immersed membranes exhibited greater mass

loss than H<sub>2</sub>O-immersed membranes, confirming stronger retention of D<sub>2</sub>O [Fig. 6(f)]. Together with the detection of D—O bonds by XPS, these results indicate that the membrane undergoes partial isotope exchange. Tritiated-water-adsorption experiments further confirmed this effect [Figs. S50(a) and S50(b) in the Supplemental Material [98]]. The adsorption capacity decreased with decreasing oxygen-containing functional groups in GO [Fig. S50(c) in the Supplemental Material [98]], highlighting the role of these groups in isotope retention. Taken together, these findings demonstrate that the membrane achieves efficient H:D isotope separation through the combined effects of interlayer-spacing regulation, intrinsic physical property differences between H<sub>2</sub>O and D<sub>2</sub>O (mass, hydrogen bonding strength, clustering behavior, and viscosity), and isotope exchange. This mechanism not only accounts for the observed flux differences but also underscores the potential of this approach for optimizing deuterium and tritium recovery in nuclear-fusion fuel cycles.

In summary, due to the differences in physical properties such as mass, hydrogen bond strength, molecular aggregation behavior, and viscosity between H<sub>2</sub>O and D<sub>2</sub>O, the membrane can achieve efficient hydrogen-deuterium separation by utilizing interlayer-spacing regulation and permeation-rate differences. The membrane confirmed that as the D<sub>2</sub>O concentration increased, the membrane interlayer spacing decreased, making the transmission rate of H<sub>2</sub>O significantly higher than that of D<sub>2</sub>O. In addition, the membrane exhibits a certain isotope-exchange effect, which further enhances the separation ability of hydrogen isotopes through the formation of D—O bonds and the retention of D<sub>2</sub>O in the membrane. Tritium-water-separation experiments also show that the oxygen functional groups on the membrane have a certain adsorption effect on tritium water, and that the reduction of oxygen content will weaken this rejection ability. The revelation of this mechanism confirms that this method is of great significance for optimizing the recycling of deuterium and tritium in the nuclear-fusion fuel cycle.

#### IV. CONCLUSIONS

In this paper, a GO interlayer-spacing intelligent control membrane with photothermal response was prepared and synthesized by a simple physical-intercalation method. Furthermore, according to the characterization results of XRD at different temperatures, the interlayer-spacing–temperature-change curve is fitted so as to achieve precise control of the interlayer spacing during the separation process. Furthermore, *in situ* variable-temperature TEM directly revealed that Ga nanoparticles encapsulated between GO layers undergo fully reversible expansion and contraction during heating-cooling cycles, providing microscopic evidence for the proposed surface-tension-driven modulation of interlayer spacing. The

membrane had excellent stability and has a certain separation effect on metal ions in simulated salt lakes in a short period of time. At the same time, the membrane used temperature to control the interlayer spacing to change its water flux, and combined the slight physical differences between hydrogen and deuterium water and isotope exchange to achieve the separation of H<sub>2</sub>O and D<sub>2</sub>O. For 10% D<sub>2</sub>O, the water-flux ratio reaches 5.66, and for 60% D<sub>2</sub>O, SF  $\approx$  70.53. In addition, and most importantly, the membrane has a removal rate of approximately 19.92% for tritium water in nuclear-power-plant wastewater. This work has not only proposed a new design idea for membrane-layer regulation but, more importantly, the membrane has provided an energy-saving and efficient solution for hydrogen-isotope separation by utilizing precise interlayer-spacing control and the isotope-exchange effect. Its effective removal of tritiated water not only reduces the environmental risks associated with nuclear energy, but also improves the sustainability of fusion fuel management, providing a promising path for future fusion-reactor applications.

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#### DATA AVAILABILITY

The data supporting the findings of this study are available within the paper [98].

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