Electronic Supplementary Material (ESI) for Nanoscale.

Influence of the Presence of the Cations on the Water and Salt Dynamics Inside Layered Graphene Oxide (GO) Membranes

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Interpretation of the parameter D

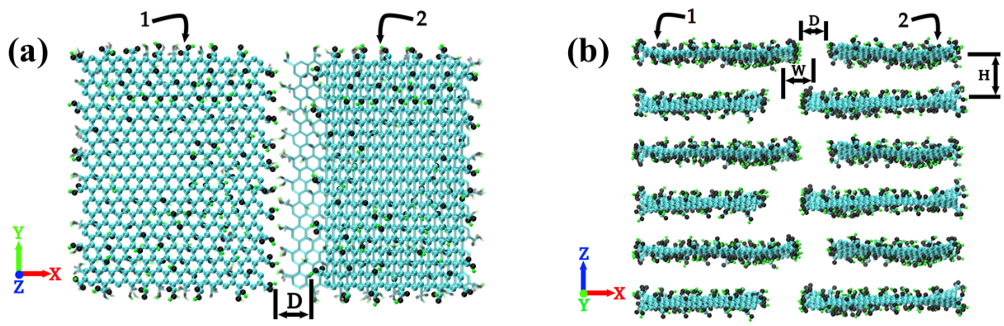


Figure S1: GO membrane. (a) Top view. (b) Side view. Green color is for hydrogen atoms, black color is for oxygen atoms, cyan color is for carbon atoms.

The parameter **D** is called pore width of the membrane. Here a detailed interpretation of this parameter is provided. As shown in Figure S1 , at the top layer of the membrane, the GO sheet on the left is marked as 1 and the GO sheet on the right is marked as 2. Suppose, the maximum value of x coordinate of sheet 1 is $X1_{max}$ and the minimum value of x coordinate of sheet 2 is $X2_{min}$. Then the parameter **D** is defined as **D**$= X2_{min} - X1_{max}$. This definition of **D** holds good for other layers of the GO membrane and will have the same value.

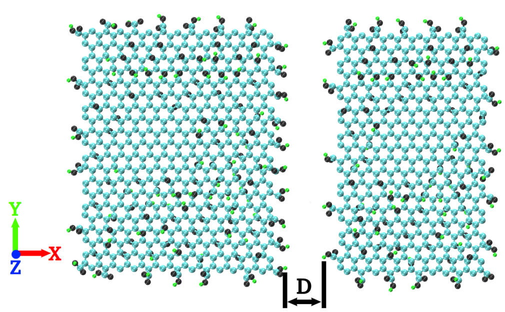


Figure S2: A single layer of layered GO membrane. Green color is for hydrogen atoms, black color is for oxygen atoms, cyan color is for carbon atoms.

Simulation setup for forward osmosis (FO)

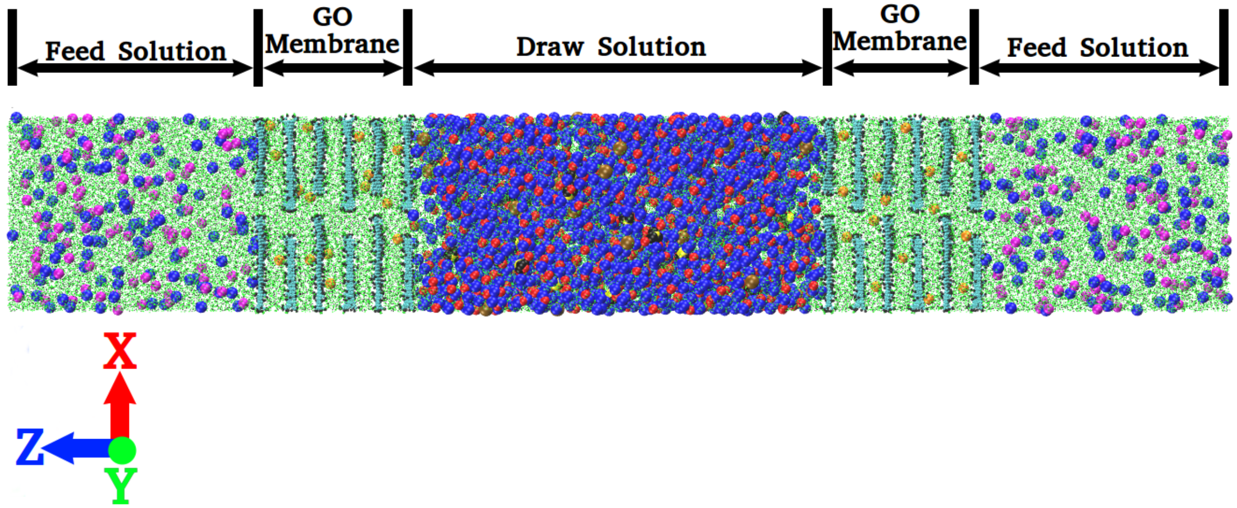


Figure S3: Simulation setup forward osmosis (FO). Green color is for hydrogen atoms, black color is for oxygen atoms, cyan color is for carbon atoms, blue color is for Cl^- ions, magenta color is for Na^+ ions, orange color is for K^+ ions.

Continuous H bond autocorrelation function

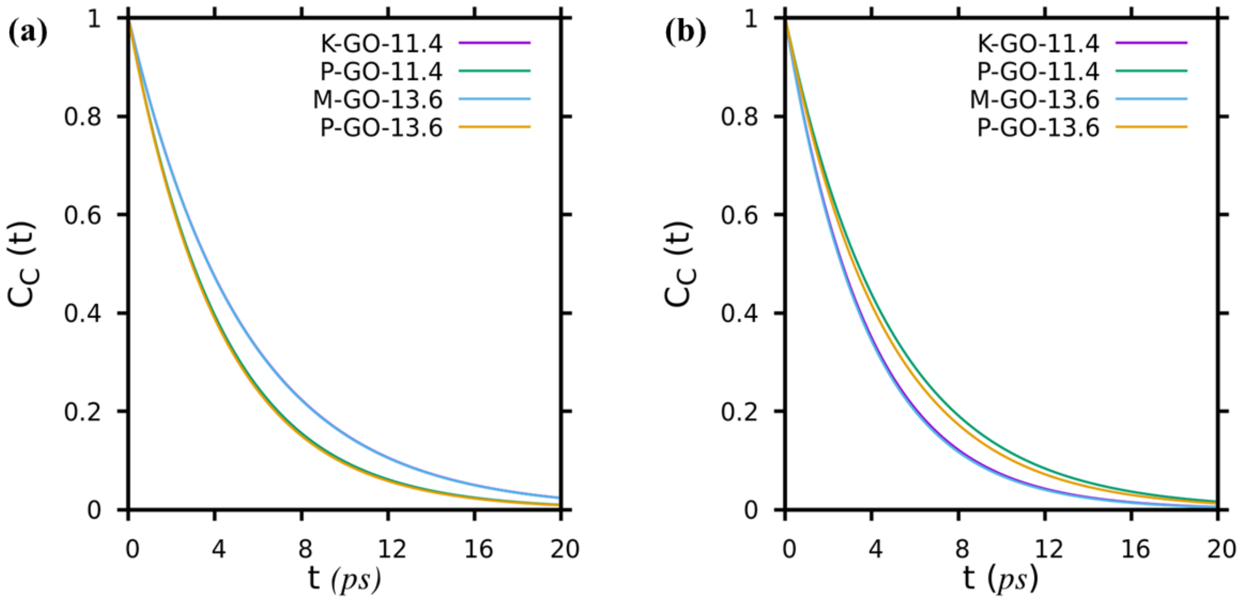


Figure S4: Continuous H bond autocorrelation function between (a) water molecules (b) water and GO.

Na^+ and Cl^- ion rejection by M-GO-13.6 and P-GO-13.6 membrane systems

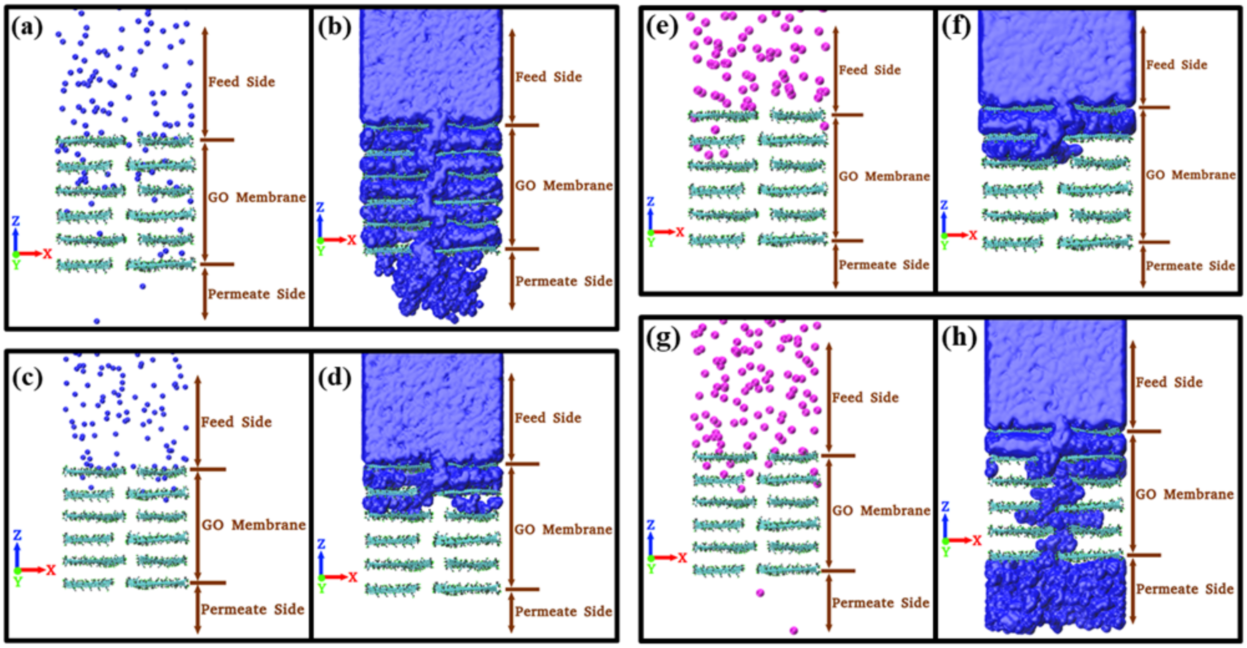


Figure S5: (a) Distribution of Cl^- ions at the end of the simulation for M-GO-13.6 membrane system. (b) Spatial distribution function of Cl^- ions for M-GO-13.6 membrane system. (c) Distribution of Cl^- ions at the end of the simulation for P-GO-13.6 membrane system. (d) Spatial distribution function of Cl^- ions for P-GO-13.6 membrane system. (e) Distribution of Na^+ ions at the end of the simulation for M-GO-13.6 membrane system. (f) Spatial distribution function of Na^+ ions for M-GO-13.6 membrane system. (g) Distribution of Na^+ ions at the end of the simulation for P-GO-13.6 membrane system. (h) Spatial distribution function of Na^+ ions for P-GO-13.6 membrane system.

PMF plots for M-GO-13.6 and P-GO-13.6 membrane systems

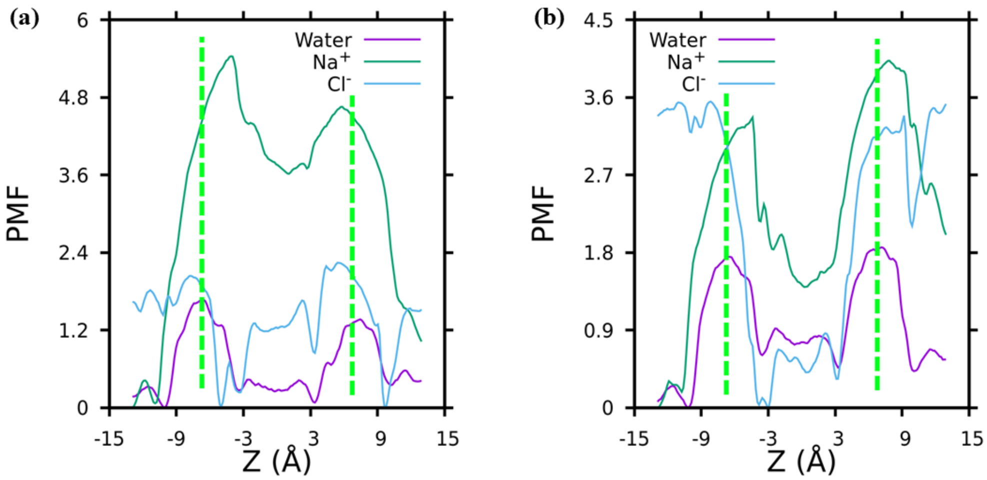


Figure S6: (a) PMF for M-GO-13.6 membrane system. (b) PMF for P-GO-13.6 membrane system. The entry and exit points of the permeating species through the GO laminates are shown with a pair of dotted green lines.

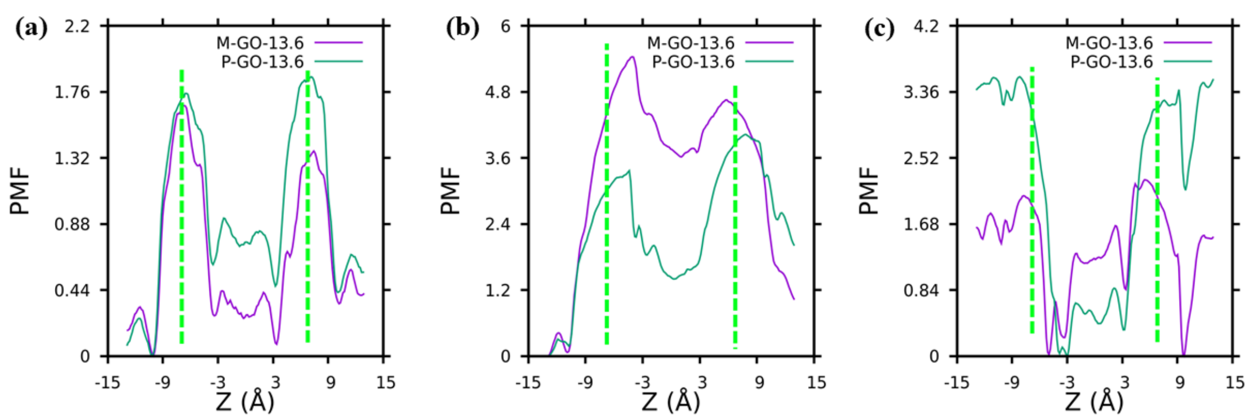


Figure S7: Comparison of PMF between M-GO-13.6 and P-GO-13.6 membrane system for (a) Water, (b) Na^+ ions and (c) Cl^- ions. The entry and exit points of the permeating species through the GO laminates are shown with a pair of dotted green lines.

Radial distribution function ($g(r)$) between the oxygen atoms of water inside the layered GO membranes

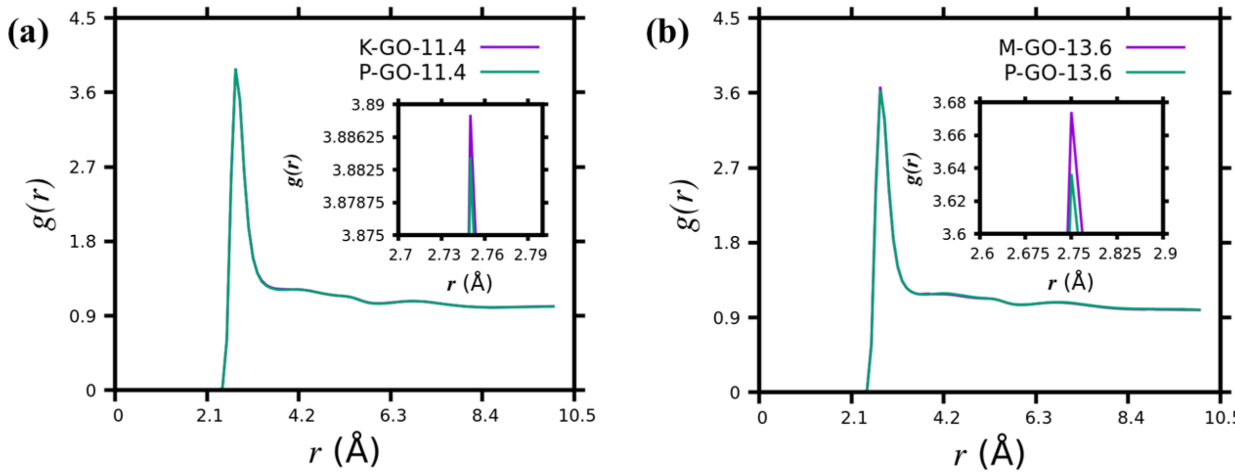
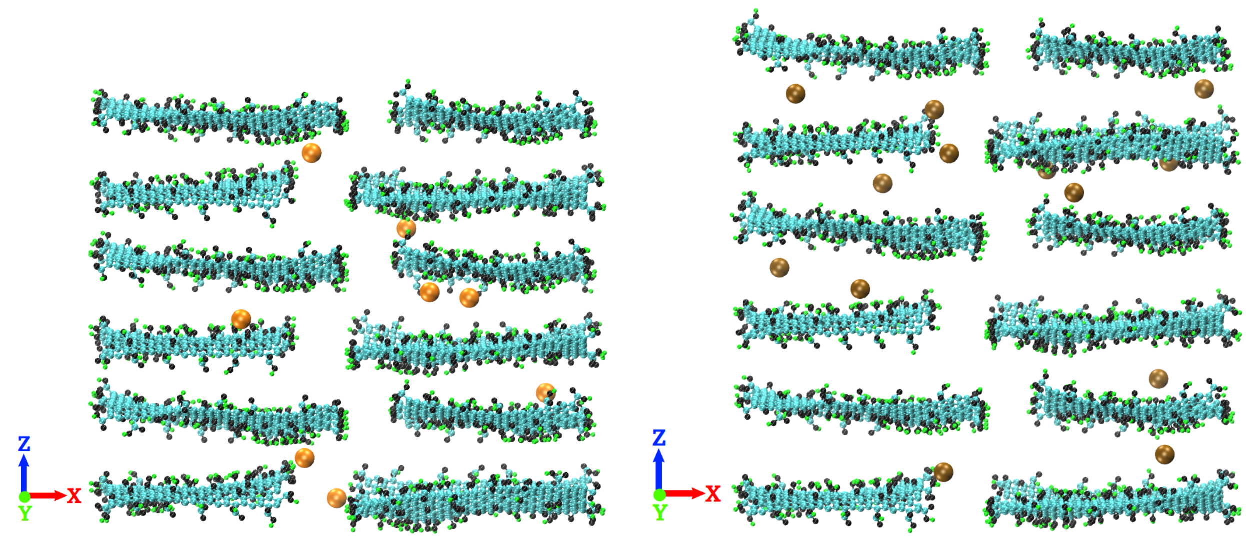


Figure S8: Radial distribution function ($g(r)$) between the oxygen atoms of water inside the layered GO membranes (a) K-GO-11.4 and P-GO-11.4 and (b) M-GO-13.6 and P-GO-13.6.

Figure S8a and S8b shows the radial distribution function between the oxygen atoms of water inside the layered GO membranes. For all the cases shown in Figure S8a and S8b, the first peak of $g(r)$ is observed at 2.75 Å. However, for cation intercalated layered GO membranes (K-GO-11.4 and M-GO-13.6), the intensity of this peak slightly increases as depicted in the insets of Figure S8a and S8b. This indicates that the water molecules inside the layered GO membranes have slightly higher compact arrangement in the presence of the cations.

Dynamics of intercalated cations (K^+ and Mg^{2+})

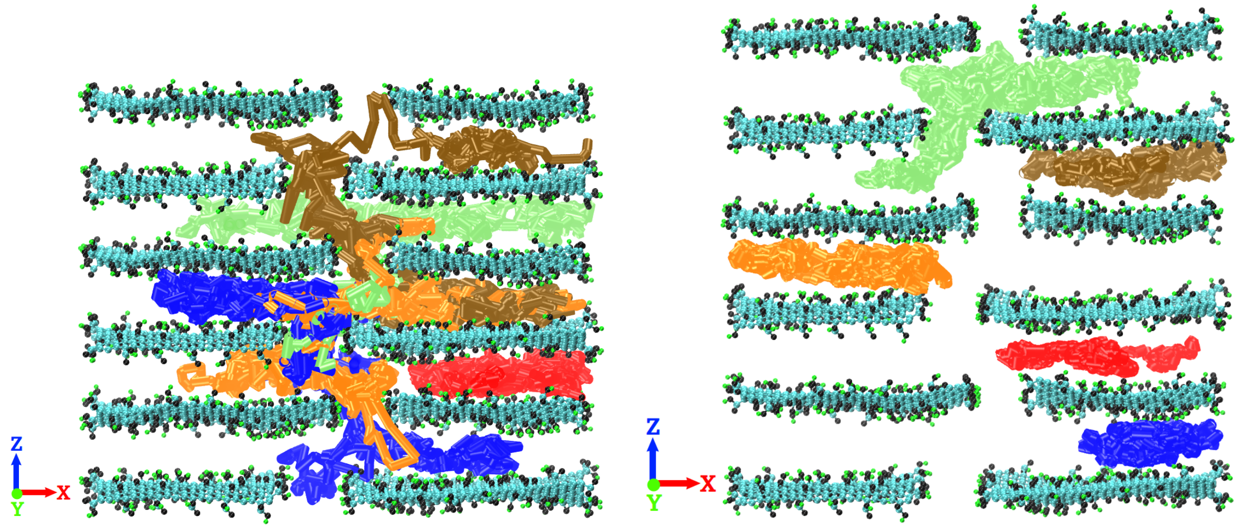


(a) (b)

Figure S9: Distribution of the intercalated cations inside the layered GO membranes at the end of the simulation. (a) K^+ intercalated membrane (K-GO-11.4). (b) Mg^{2+} intercalated membrane (M-GO-13.6). Green color is for hydrogen atoms, black color is for oxygen atoms, cyan color is for carbon atoms, orange color is for K^+ ions and ochre color is for Mg^{2+} ions.

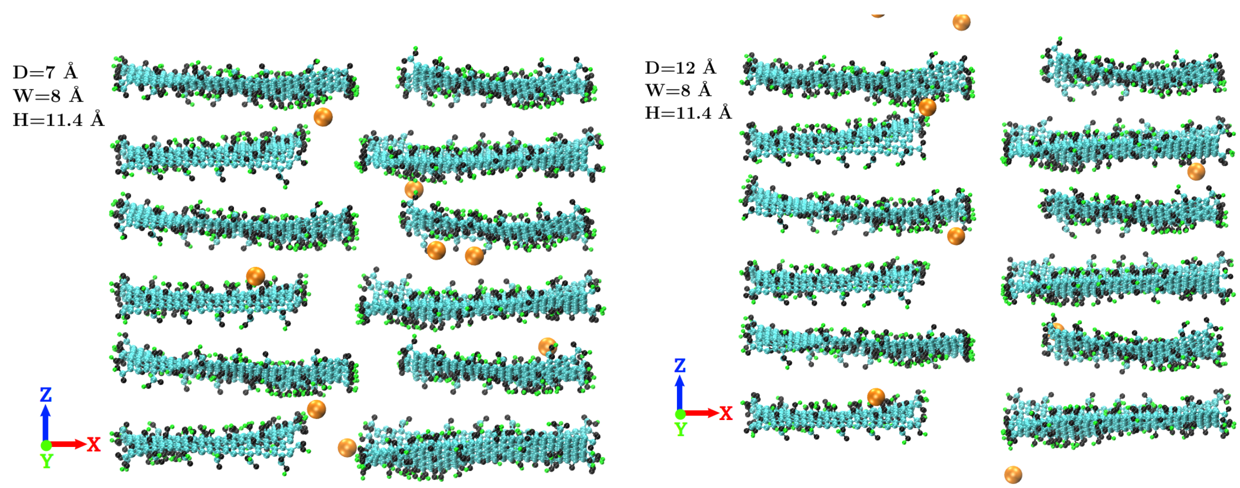
At the end of the simulation 8 K^+ ions remain trapped inside the interlayer gallery of K^+ intercalated layered GO membrane (Figure S9a) and 13 Mg^{2+} ions remain trapped inside the interlayer gallery of Mg^{2+} intercalated layered GO membrane (Figure S9b). Figure S10 shows the representative trajectory of the cations inside the interlayer gallery of layered GO membranes. As can be seen from Figure S10, the K^+ ions are much more mobile (Figure S10a) inside the interlayer gallery of layered GO membranes as compared to Mg^{2+} ions (Figure S10b). Because of their higher mobility inside the interlayer gallery of layered GO membrane, less number of K^+ ions remain trapped inside the interlayer gallery at the end of the simulation as compared to Mg^{2+} ions (8 K^+ ions while 13 Mg^{2+} ions) as shown in Figure S9. On the same note, the amount cations remain resided inside the interlayer gallery also depends upon the geometric parameters of the layered GO membranes.

Figure S11a and S11b show the distribution of intercalated K^+ ions inside the layered



(a) (b)

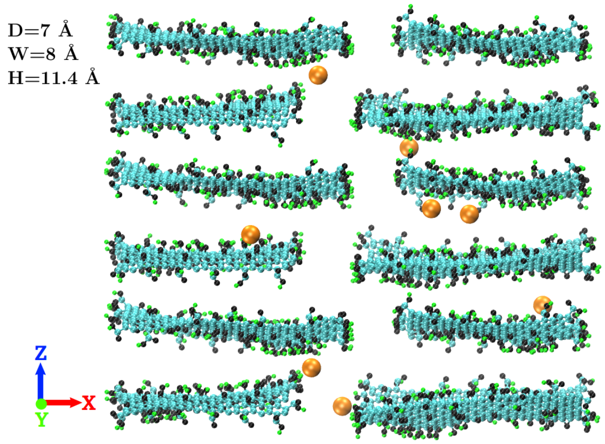
Figure S10: Trajectory of 5 different cations are tracked inside the layered GO membranes. (a) Trajectory of K^+ ions. (b) Trajectory of Mg^{2+} ions. Each color represents the trajectory of a single cation. For the membrane, green color is for hydrogen atoms, black color is for oxygen atoms, cyan color is for carbon atoms.



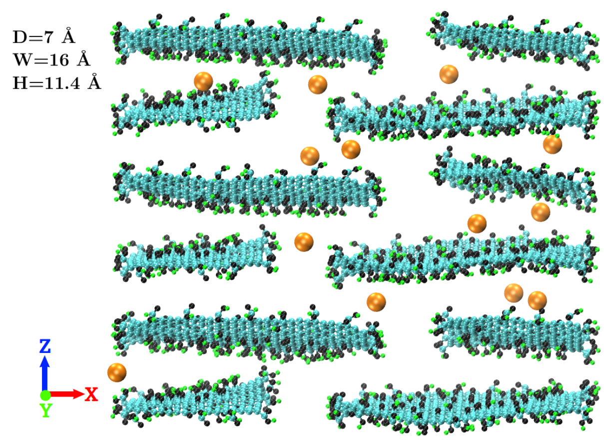
(a) (b)

Figure S11: Distribution of K^+ ions at the end of the simulation inside the layered GO membranes with (a) $\mathbf{D}$ = 7 Å and (b) $\mathbf{D}$ = 12 Å. Green color is for hydrogen atoms, black color is for oxygen atoms, cyan color is for carbon atoms and orange color is for K^+ ions.

GO membranes with $\mathbf{D}$ = 7 Å and $\mathbf{D}$ = 12 Å respectively at the end of the simulation. For layered GO membrane with $\mathbf{D}$ = 7 Å, 8 K^+ ions remain resided inside the interlayer gallery



(a)



(b)

Figure S12: Distribution of K^+ ions at the end of the simulation inside the layered GO membranes with (a) **W** = 8 Å and (b) **W** = 16 Å. Green color is for hydrogen atoms, black color is for oxygen atoms, cyan color is for carbon atoms and orange color is for K^+ ions.

of layered GO membrane (Figure S11a) while for layered GO membrane with **D** = 12 Å, 5 K^+ ions remain resided inside the interlayer gallery of layered GO membrane (Figure S11b). So, with the increase in the parameter **D** (pore width), the number of intercalated cations that were retained inside the membrane reduces.

Similarly, Figure S12a and S12b show the distribution of intercalated K^+ ions inside the layered GO membranes with **W** = 8 Å and **W** = 16 Å respectively at the end of the simulation. For layered GO membrane with **W** = 8 Å, 8 K^+ ions remain trapped inside the interlayer gallery of layered GO membrane as shown in Figure S12a. On the other hand for layered GO membrane with **W** = 16 Å, 13 K^+ remain trapped inside the interlayer gallery of layered GO membrane (Figure S12b). So, with the increase in the parameter **W**, the number of intercalated cations which remain trapped inside the interlayer gallery of layered GO membrane increases. The parameter **W** (pore offset distance) is a measure of the lateral dimension of the GO nanosheets used to construct the layered GO membranes.[1–4] In other words, one of the way to retain a greater number of intercalated cations inside the interlayer gallery of layered GO membrane is to use GO nanosheets of larger lateral dimensions to construct the GO membrane.

Effect of the parameter W and D

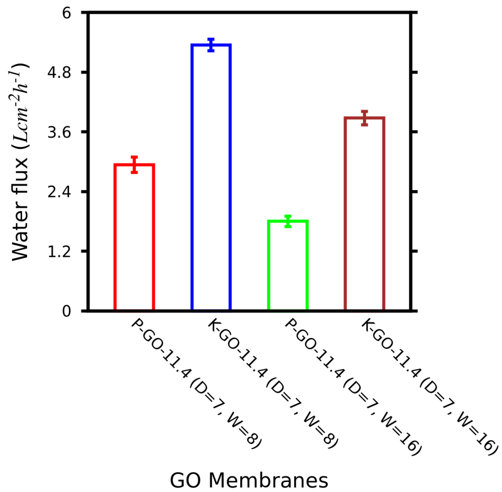
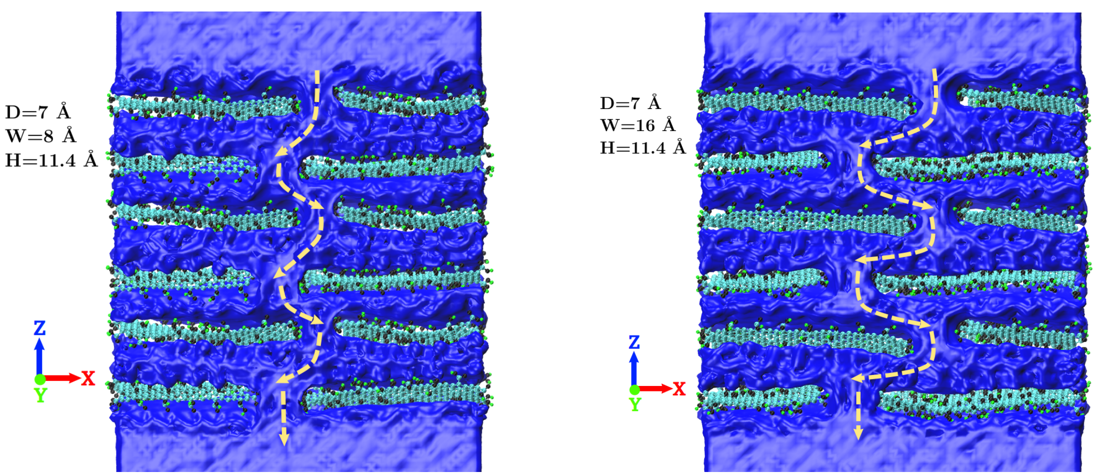


Figure S13: Water flux through layered GO membranes.



(a) (b)

Figure S14: Spatial distribution of water for layered GO membranes for (a) **W** = 8 Å and (b) **W** = 16 Å. The permeation pathways inside the membranes are shown with a dashed yellow line.

In this present study, the value of the geometric parameters **D** (pore width) and **W** (pore offset distance) are considered as 7 Å and 8 Å respectively. As the primary focus of this present study is to investigate the effect of presence of cations inside the interlayer gallery of layered GO membranes on the water and salt dynamics, these geometric parameters (**D**, **W**)

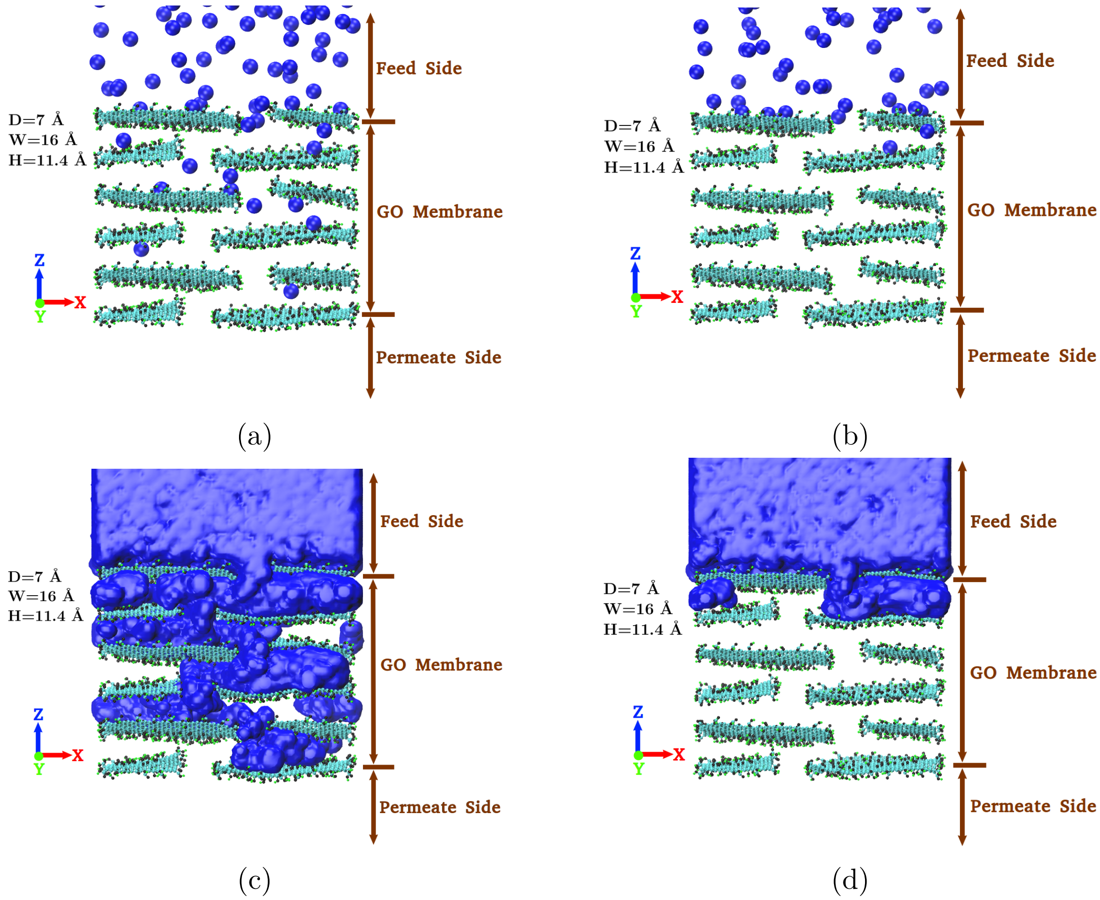


Figure S15: Snapshot of the distribution of Cl^- ions at the end of the simulation for (a) K^+ intercalated membrane (b) pristine membrane. Spatial distribution of Cl^- ions for (c) K^+ intercalated membrane (d) pristine membrane. The parameter **W** = 16 Å.

are kept constant to maintain consistency among all the simulations. However, the role of these parameters are very crucial in determining the performance of a layered GO membrane which is also reported in our previous studies.[1–3] In this section, we briefly reported the effect of the parameters **D** and **W** on the performance of cation intercalated and pristine layered GO membranes.

To investigate the effect of parameter **W** on the performance of cation intercalated and pristine layered GO membranes, we have constructed two configurations of layered GO membranes with **W** = 8 Å and **W** = 16 Å. For both these membrane configurations the value of the parameters **D** and **H** are 7 Å and 11.4 Å respectively. The value of **H** is 11.4 Å, as K^+

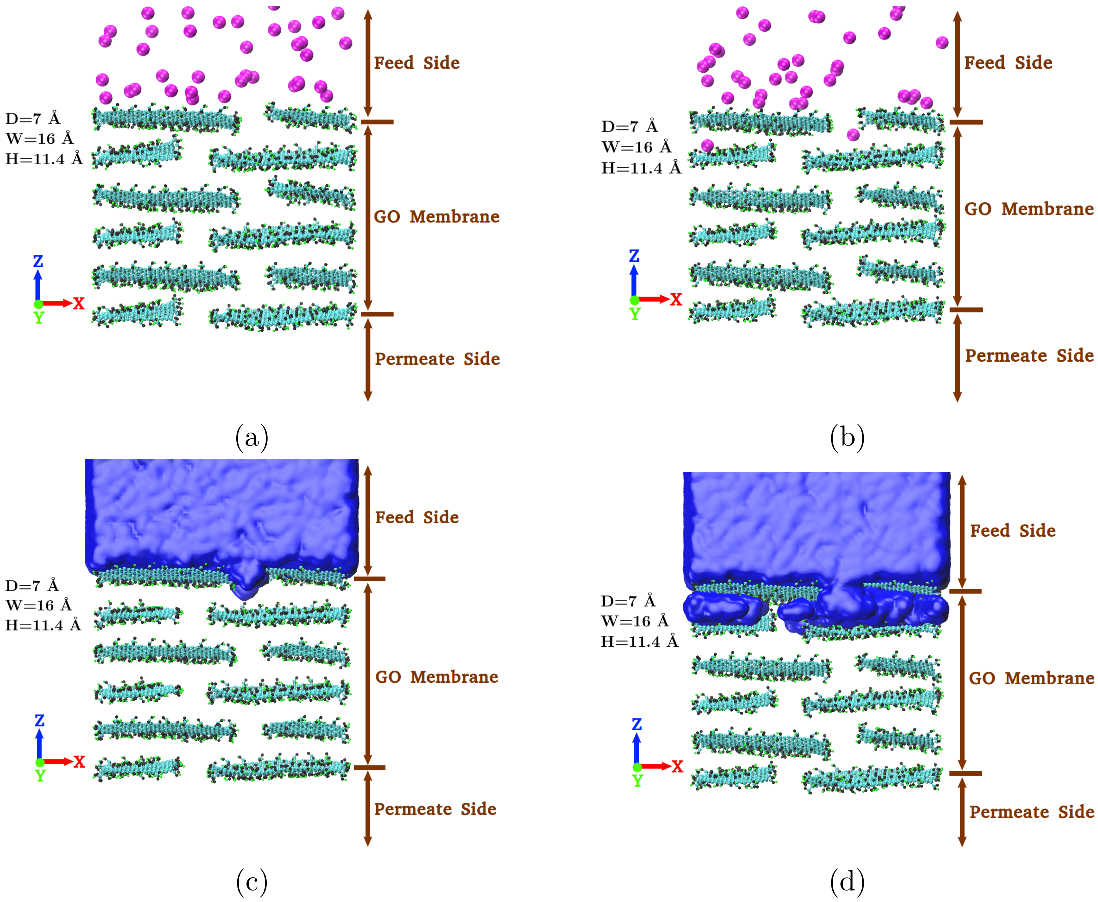


Figure S16: Snapshot of the distribution of Na^+ ions at the end of the simulation for (a) K^+ intercalated membrane (b) pristine membrane. Spatial distribution of Na^+ ions for (c) K^+ intercalated membrane (d) pristine membrane. The parameter $\mathbf{W}$ = 16 Å.

is considered as the intercalating cation for these membranes.

As can be seen from Figure S13, P-GO-11.4 membrane with $\mathbf{W}$ = 8 Å has higher water flux than P-GO-11.4 membrane with $\mathbf{W}$ = 16 Å. Similarly, K-GO-11.4 membrane with $\mathbf{W}$ = 8 Å has higher water flux than K-GO-11.4 membrane with $\mathbf{W}$ = 16 Å. This signified that with the increase in the parameter $\mathbf{W}$, the water permeance through layered GO membrane decreases. With the increase in $\mathbf{W}$, water has to traverse a more “zig-zag” pathways inside the membrane to permeate through, as shown in Figure S14. In other words, water molecules have to cover a larger distance to permeate through the membrane as the parameter $\mathbf{W}$ increases with turn reduces the water permeance through the membrane. This observation is

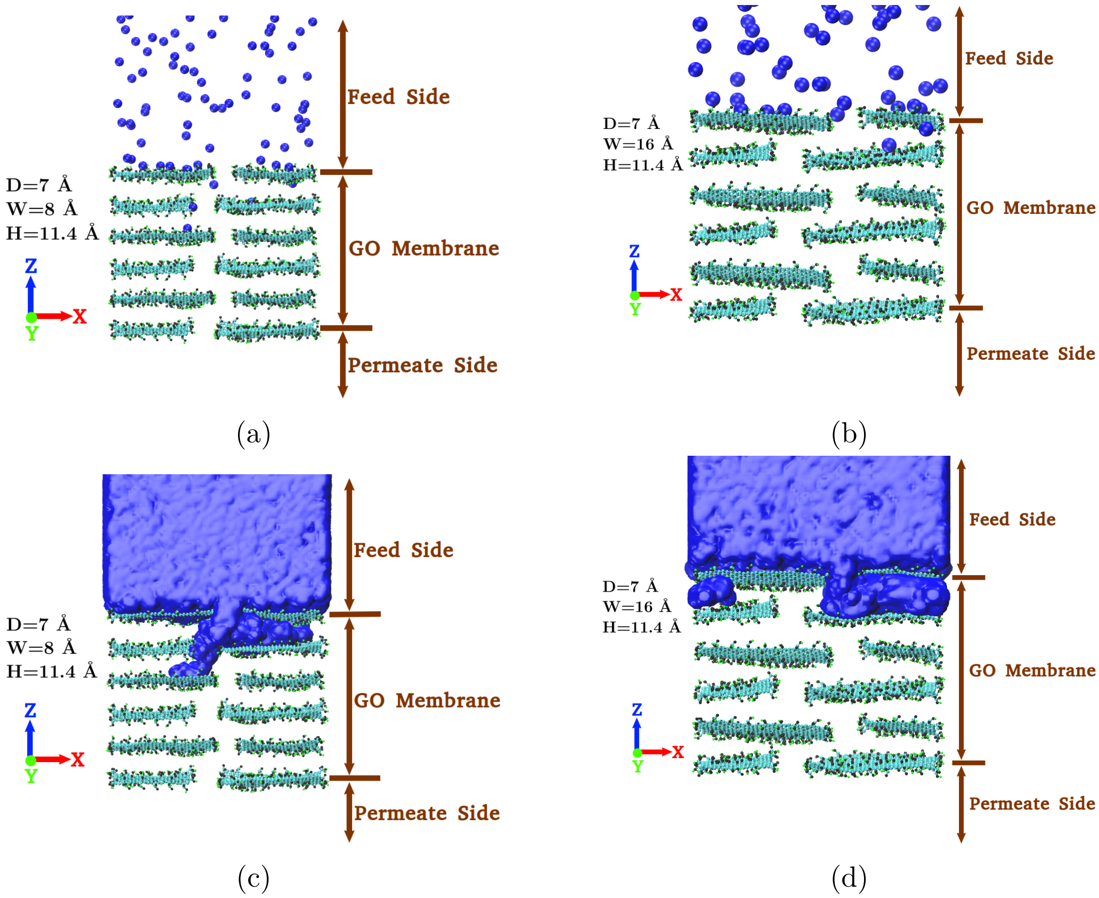


Figure S17: Snapshot of the distribution of Cl^- ions at the end of the simulation for pristine layered GO membrane with (a) $\mathbf{W}$ = 8 Å (b) $\mathbf{W}$ = 16 Å. Spatial distribution of Cl^- ions for pristine layered GO membranes with (c) $\mathbf{W}$ = 8 Å (d) $\mathbf{W}$ = 16 Å.

in accordance with our previous experimental and simulation studies on layered GO membranes.[1–3]

As we mentioned in the manuscript, considering the thickness of the membrane used in the simulation and the timescale of the simulation, the salt rejection ability of the layered GO membranes in this present study is compared to one another based on "*the number of salt ions intercalated inside the membrane from the feed solution and how far these ions have intercalated inside the membrane*".

Figure S15a and S15b show the distribution of Cl^- ions at the end of the simulation for cation intercalated and pristine membrane respectively (with $\mathbf{W}$ = 16 Å). Similarly,

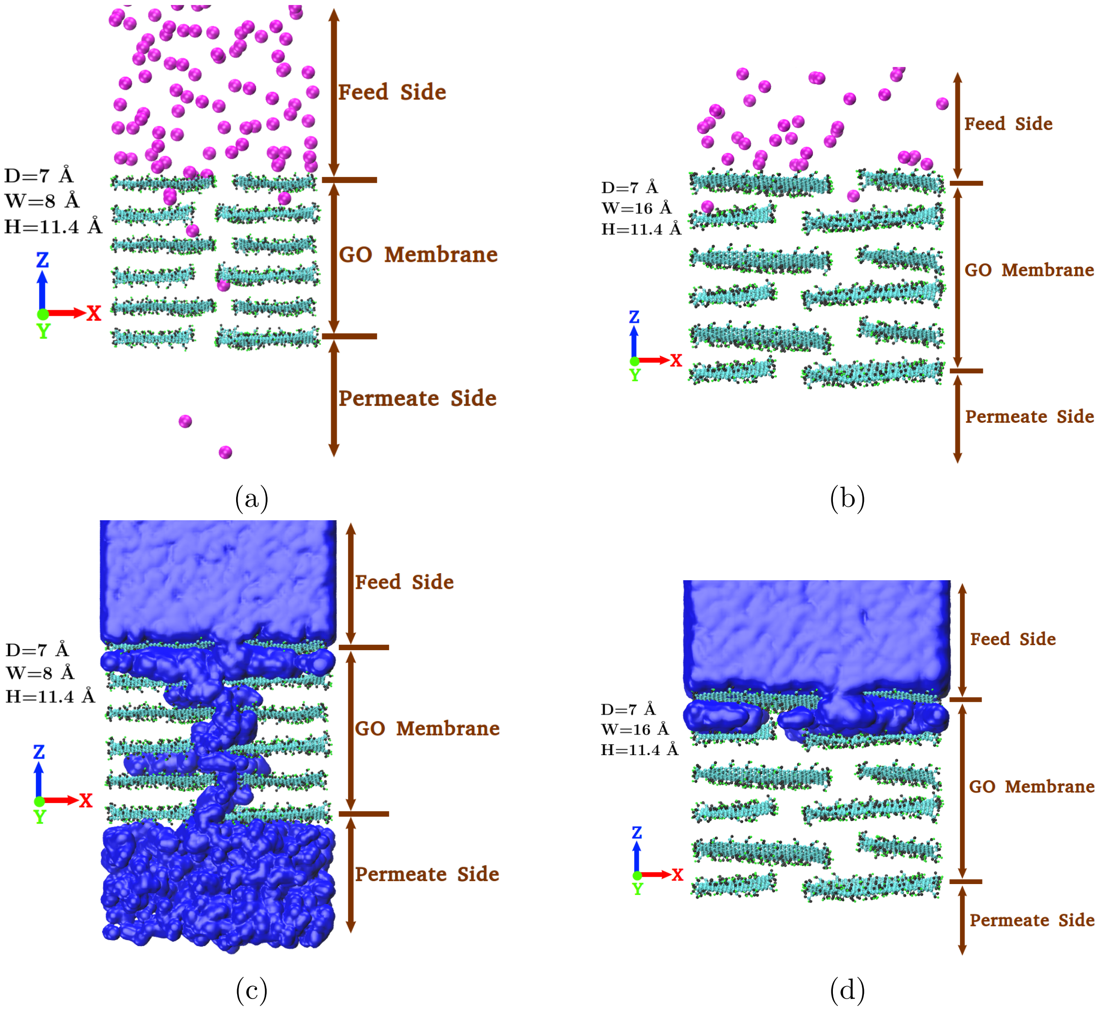


Figure S18: Snapshot of the distribution of Na^+ ions at the end of the simulation for pristine layered GO membrane with (a) $\mathbf{W}$ = 8 Å (b) $\mathbf{W}$ = 16 Å. Spatial distribution of Na^+ ions for pristine layered GO membranes with (c) $\mathbf{W}$ = 8 Å (d) $\mathbf{W}$ = 16 Å.

Figure S15c and S15d show the spatial distribution of Cl^- ions for cation intercalated and pristine membrane respectively (with $\mathbf{W}$ = 16 Å). The distribution of ions at the end of the simulation (its a snapshot at the end of the simulation) gives an idea on the number of ions intercalated inside the membrane. On the other hand, the spatial distribution of the ions gives an idea on how far the ions have intercalated inside the membrane and the path the ions have traversed through the membrane during the entire course of the simulation. Form Figure S15, it is evident that for cation intercalated membrane has lower rejection for Cl^- ions as compared to pristine membrane.

Figure S16a and S16b show the distribution of Na^+ ions at the end of the simulation for K^+ intercalated and pristine layered GO membrane respectively (with $\mathbf{W}$ = 16 Å). Figure S16c and S16d show the spatial distribution of Na^+ ions for K^+ intercalated and pristine layered GO membrane respectively (with $\mathbf{W}$ = 16 Å). As can be seen from Figure S16, K^+ intercalated layered GO membrane has slightly higher rejection towards Na^+ ions.

Figure S17a and S17b show the distribution of Cl^- ions at the end of the simulation for pristine layered GO membranes with $\mathbf{W}$ = 8 Å and $\mathbf{W}$ = 16 Å respectively. Similarly, Figure S17c and S17c show the spatial distribution of Cl^- ions for layered GO membranes with $\mathbf{W}$ = 8 Å and $\mathbf{W}$ = 16 Å respectively. For layered GO membrane with $\mathbf{W}$ = 8 Å, more number of Cl^- ions intercalated inside the membrane which also penetrate a larger distance through the membrane as compared to layered GO membrane with $\mathbf{W}$ = 16 Å.

Figure S18a and S18b show the distribution of Na^+ ions at the end of the simulation for pristine layered GO membranes with $\mathbf{W}$ = 8 Å and $\mathbf{W}$ = 16 Å respectively. Figure S18c and S18d show the spatial distribution of Na^+ ions for pristine layered GO membranes with $\mathbf{W}$ = 8 Å and $\mathbf{W}$ = 16 Å respectively. For layered GO membranes with $\mathbf{W}$ = 8 Å, more number of Na^+ ions intercalated inside the membrane and penetrate through a larger distance through the membrane as compared to layered GO membrane with $\mathbf{W}$ = 16 Å.

So, from Figures S17 and S18 it is evident that pristine layered GO membranes with higher value of $\mathbf{W}$, has higher salt rejection (Na^+ and Cl^+ ions) compared to layered GO membranes with a lower value $\mathbf{W}$. This observation also holds good for cation intercalated layered GO membranes. This increase in the salt rejection of layered GO membranes with the increase in the parameter $\mathbf{W}$, has also been reported in our previous studies on layered GO membranes.[1–3]

We have also investigated the effect of the parameter $\mathbf{D}$ (pore width) on the performance of the pristine and cation intercalated layered GO membranes. For this two layered GO membrane configurations are constructed with $\mathbf{D}$ = 7 Å and $\mathbf{D}$ = 12 Å. For both these configurations the value of the parameters $\mathbf{W}$ and $\mathbf{H}$ are 8 Å and 11.4 Å respectively. The value of the parameter **H** is 11.4 Å as K^+ ion is considered as the intercalating cation.

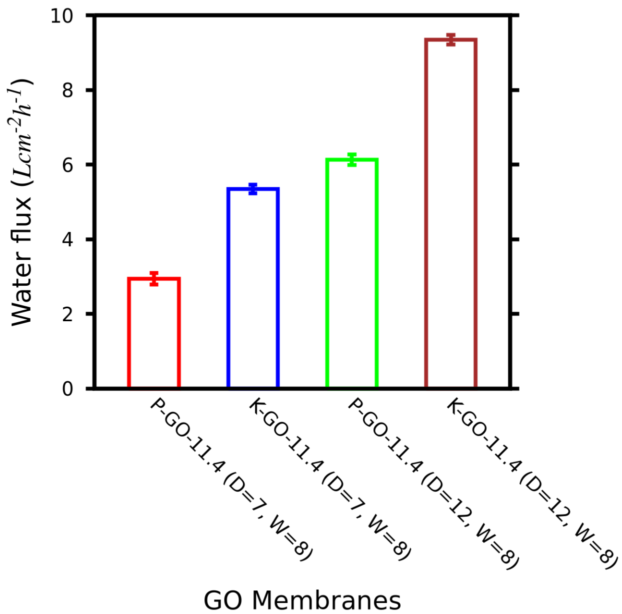


Figure S19: Water flux through layered GO membranes.

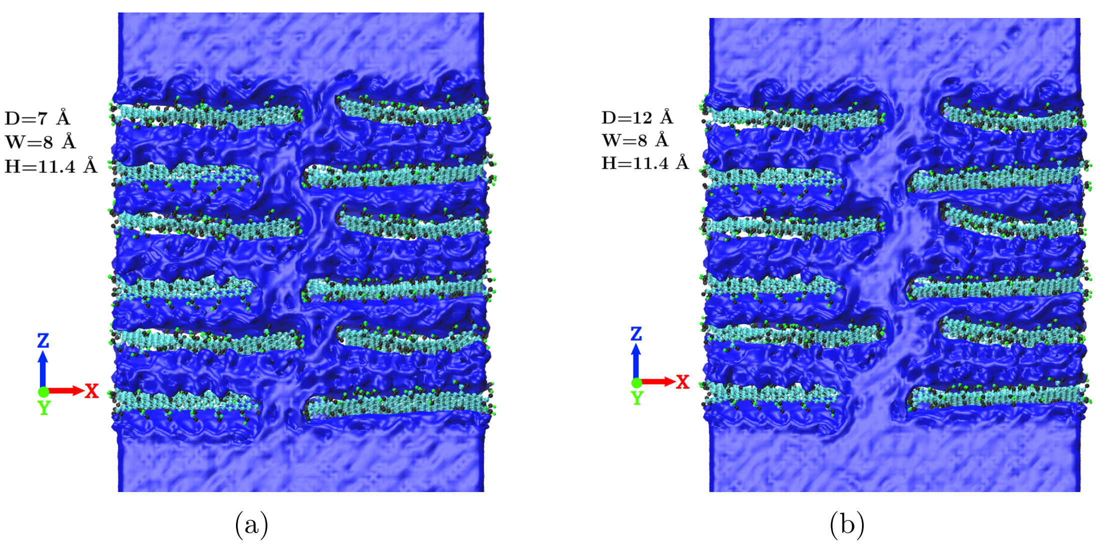
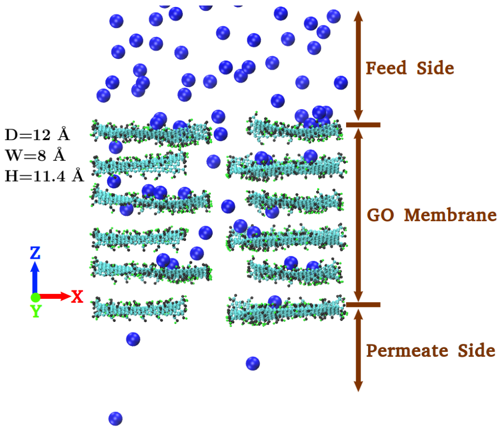
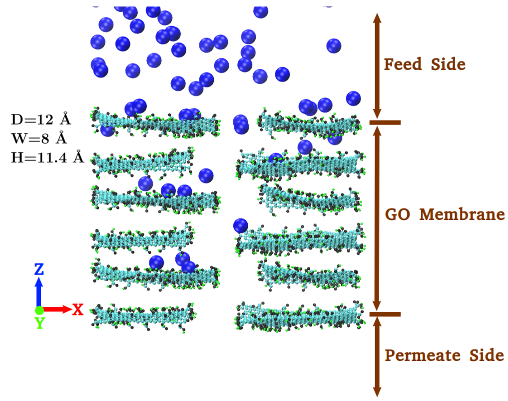


Figure S20: Spatial distribution of water for layered GO membranes for (a) **D** = 7 Å and (b) **D** = 12 Å.

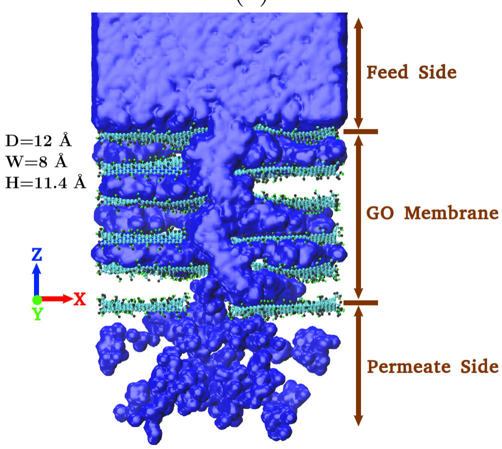
As shown in Figure S19, for pristine layered GO membranes (P-GO-11.4), water flux is higher for membrane with **D** = 12 Å as compared to the membrane with **D** = 7 Å. Similarly,



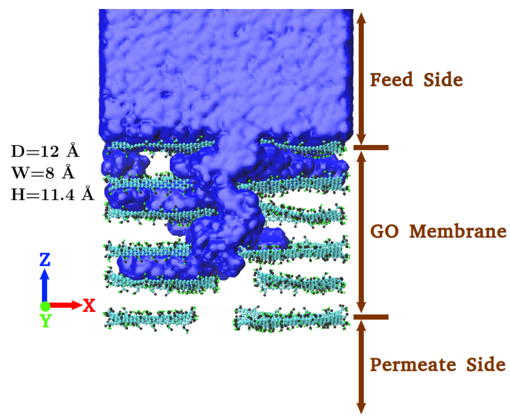
(a)



(b)



(c)



(d)

Figure S21: Snapshot of the distribution of Cl^- ions at the end of the simulation for (a) K^+ intercalated membrane (b) pristine membrane. Spatial distribution of Cl^- ions for (c) K^+ intercalated membrane (d) pristine membrane. The parameter $\mathbf{D}$ = 12 Å.

for K^+ intercalated membrane (K-GO-11.4), membrane with $\mathbf{D}$ = 12 Å has higher water flux as compared to the membrane with $\mathbf{D}$ = 7 Å. So, with the increase in the parameter $\mathbf{D}$, water permeance through the layered GO membrane increases for pristine as well as cation (K^+) intercalated membranes. However, for the same set of geometric parameters ($\mathbf{D}$, $\mathbf{W}$, $\mathbf{H}$), the cation (K^+) intercalated layered GO membranes have higher water flux as compared to pristine layered GO membranes. As shown in Figure S20, the area of the water permeation pathways through the layered GO membrane with $\mathbf{D}$ = 12 Å is higher as compared to layered GO membrane with $\mathbf{D}$ = 7 Å. Because of this increase in the area of

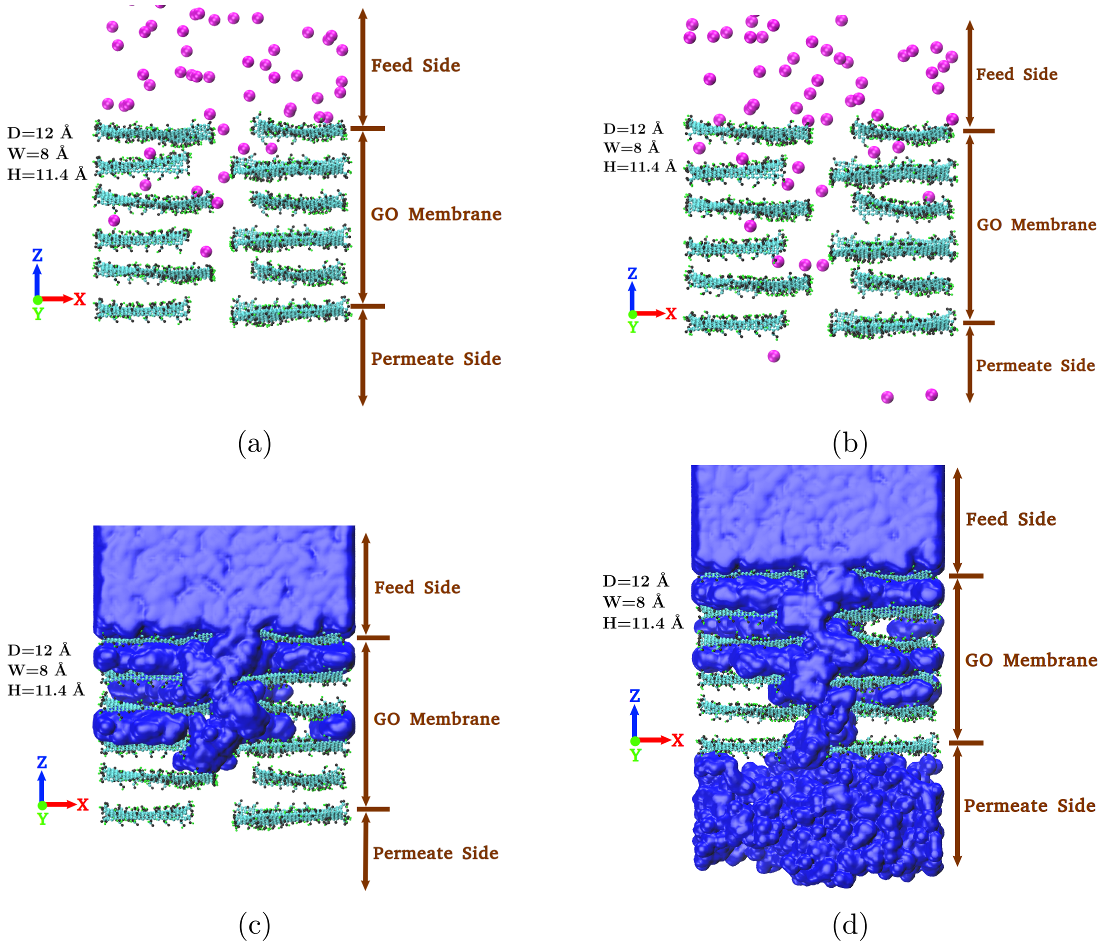


Figure S22: Snapshot of the distribution of Na^+ ions at the end of the simulation for (a) K^+ intercalated membrane (b) pristine membrane. Spatial distribution of Na^+ ions for (c) K^+ intercalated membrane (d) pristine membrane. The parameter $\mathbf{D}$ = 12 Å.

the permeation pathways, water permeance through layered GO membranes increases with the increase in the parameter $\mathbf{D}$.

Figure S21a and S21b show the distribution of Cl^- ions at the end of the simulation for cation intercalated and pristine layered GO membranes respectively (with $\mathbf{D}$ = 12 Å). Similarly, Figure S21c and S21d show the spatial distribution of Cl^- ions for cation intercalated and pristine layered GO membrane respectively. As can be seen from Figure S21, for K^+ intercalated layered GO membranes, more number of Cl^- ions entered inside the membrane and penetrate a larger distance through the membrane as compared to pristine layered GO membrane.

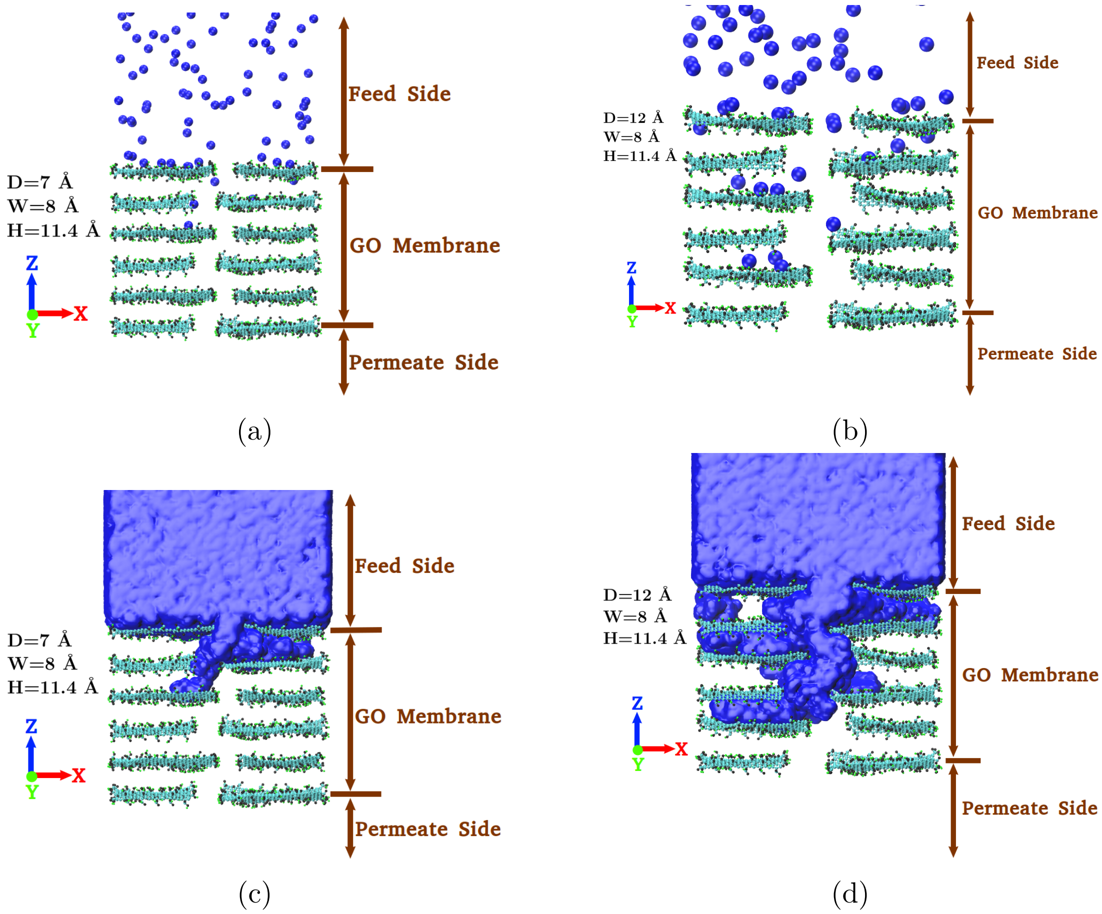


Figure S23: Snapshot of the distribution of Cl^- ions at the end of the simulation for pristine layered GO membranes with (a) $\mathbf{D}$ = 7 Å and (b) $\mathbf{D}$ = 12 Å. Spatial distribution of Cl^- ions for pristine layered GO membrane with (c) $\mathbf{D}$ = 7 Å and (d) $\mathbf{D}$ = 12 Å.

Figure S22a and S22b show the distribution of Na^+ ions at the end of the simulation for cation intercalated and pristine layered GO membrane respectively (the value of the parameter $\mathbf{D}$ is 12 Å). similarly, Figure S22c and S22d show the spatial distribution of Na^+ ions for cation intercalated and pristine layered GO membrane respectively (the value of the parameter $\mathbf{D}$ is 12 Å). For cation (K^+) intercalated layered GO membrane, less number of Na^+ ions intercalated inside the membrane and penetrate a lesser distance through the membrane as compared to pristine layered GO membrane (Figure S22). So, from Figure S21 and S22 it is evident that for layered GO membrane with $\mathbf{D}$ = 12 Å also, the intercalation of K^+ ions leads to decrease in the Cl^- rejection while the rejection Na^+ ions increases.

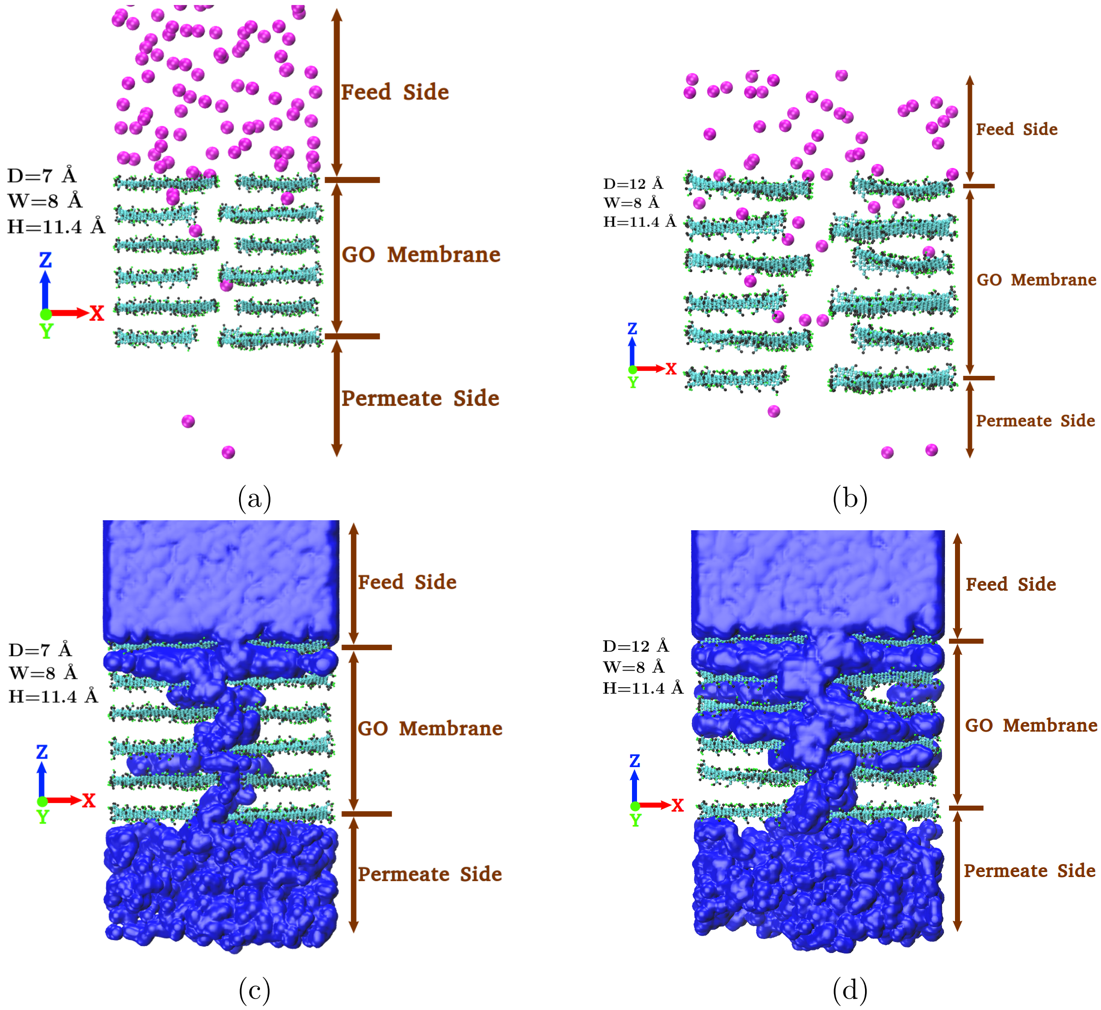


Figure S24: Snapshot of the distribution of Na^+ ions at the end of the simulation for pristine layered GO membranes with (a) $\mathbf{D}$ = 7 Å and (b) $\mathbf{D}$ = 12 Å. Spatial distribution of Na^+ ions for pristine layered GO membrane with (c) $\mathbf{D}$ = 7 Å and (d) $\mathbf{D}$ = 12 Å.

Figure S23a and S23b show the distribution of Cl^- ions at the end of the simulation for pristine layered GO membrane with $\mathbf{D}$ = 7 Å and $\mathbf{D}$ = 12 Å respectively. Similarly, Figure S23c and S23d show the spatial distribution of Cl^- ion for pristine layered GO membranes with $\mathbf{D}$ = 7 Å and $\mathbf{D}$ = 12 Å respectively. For layered GO membrane with $\mathbf{D}$ = 12 Å more number of Cl^- ions intercalated inside the membrane as compared to layered GO membrane with $\mathbf{D}$ = 7 Å (Figure S23a and S23b). These Cl^- ions penetrate through the layered GO membrane to a larger extent for the membrane with $\mathbf{D}$ = 12 Å as compared to the membrane with $\mathbf{D}$ = 7 Å (Figure S23c and S23d).

Figure S24a and S24b show the distribution of Na^+ ions at the end of the simulation for pristine layered GO membrane with **D** = 7 Å and **W** = 12 Å. Figure S24c and S24d show the spatial distribution of Na^+ ions for pristine layered GO membrane with **D** = 7 Å and **D** = 12 Å respectively. For layered GO membrane with **D** = 12 Å more number of Na^+ ions intercalated inside the membrane as compared to layered GO membrane with **D** = 7 Å (Figure S24a and S24b). On the same note, for layered GO membrane with **D** = 12 Å, 3 Na^+ ions have completely permeated through the membrane from the feed side to the permeate side, while for layered GO membrane with **D** = 7 Å, 2 Na^+ ions have completely permeated through the membrane from feed side to the permeate side. So from Figure S23 and S24, it is evident that with the increase in the parameter **D**, salt (Na^+ and Cl^- ions) rejection of pristine layered GO membrane decreases. The same observation holds good for cation (K^+) intercalated layered GO membranes.

So it has been observed that with the increase in the parameter **W**, water permeance through layered GO membrane decreases while the salt rejection increases. On the other hand with the increase in the parameter **D**, water permeance through layered GO membrane increases while the salt rejection decreases.

Distribution of permeation time

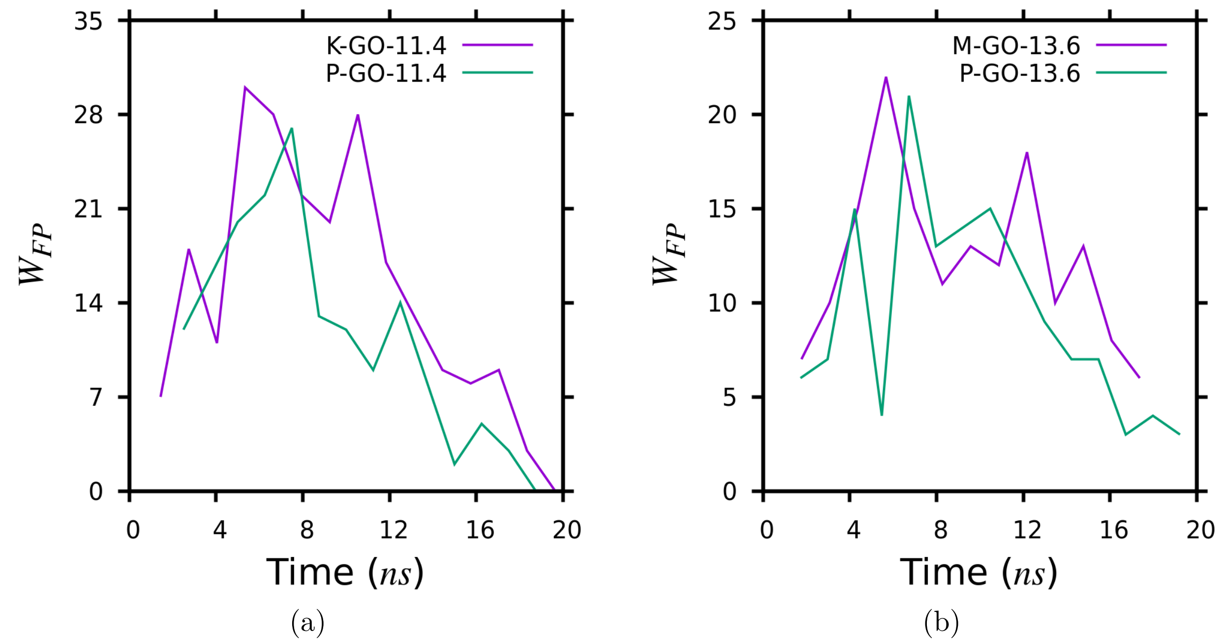
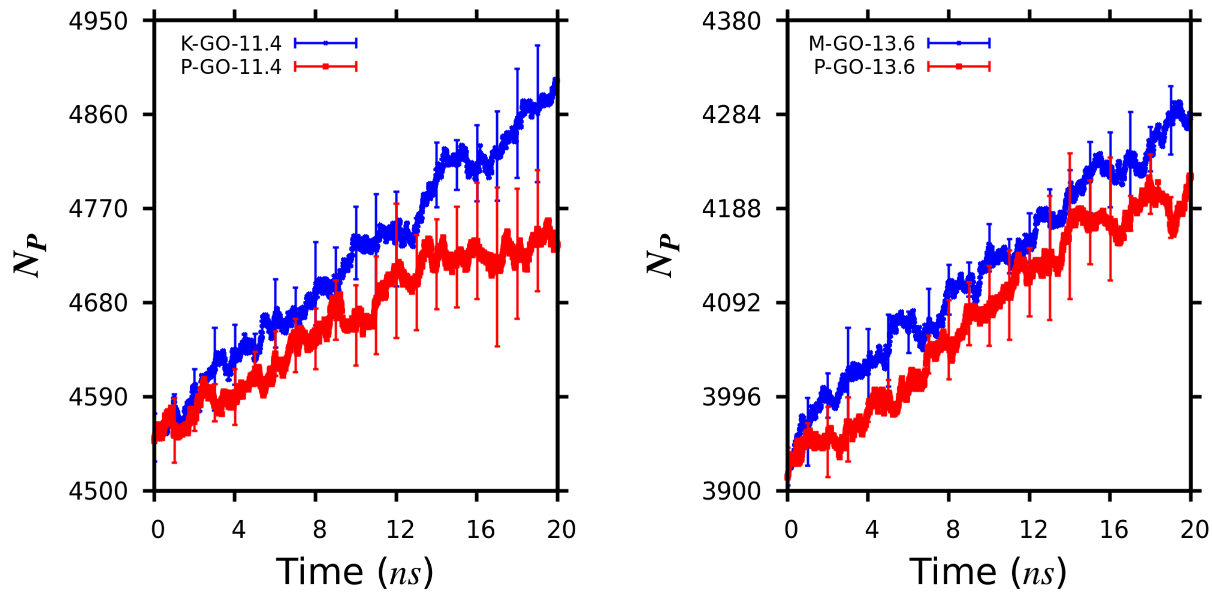


Figure S25: Distribution of permeation time for (a) K-GO-11.4 and P-GO-11.4 membrane system (b) M-GO-13.6 and P-GO-13.6 membrane system.

Figure S25 shows the distribution of permeation time for the layered GO membranes. The permeation time refers to the time taken by a water molecule to permeate through the membrane from the feed side to the permeate side. To calculate the permeation time distribution, we consider the water molecules that have entered inside the membrane from the feed side and completely permeated through the membrane towards the permeate side (W_{FP}). As can be seen from Figure S25a, for K-GO-11.4 membrane system, the majority of water molecules inclined towards lower permeation time as compared to P-GO-11.4 membrane system. Similarly, for M-GO-13.6 membrane system, the majority of water molecules inclined towards lower permeation time as compared to P-GO-13.6 membrane system as show in Figure S25b. This lower permeation time for cation intercalated layered GO membranes (K-GO-11.4/M-GO-13.6) signifies its higher water permeability as compared to corresponding pristine layered GO membranes (P-GO-11.4/P-GO-13.6).

Error analysis of the kinetic permeation curves



(a) (b)

Figure S26: Variation of number of water molecules in the permeate side ($\boldsymbol{N_P}$) with time.

For the error analysis of the kinetic permeation curves (Figure 2 of the manuscript), the standard deviation of the data points are calculated as follows:

$$\sigma = \sqrt{\frac{\sum_{i=1}^{N} (x_i - \mu)^2}{N}} \tag{1}$$

where σ is the standard deviation, N is the number of data points, x_i is the i^{th} data point and μ is the mean value of the data points. The kinetic permeation curves with error bars are shown in Figure S26.

Reliability analysis

To measure the reliability of the kinetic permeation curves (Figure 2 of the manuscript) we calculated the value of Cronbach's alpha (α) for the corresponding data. Cronbach's alpha is a measure of reliability of a set of variables. For a set of k variables Cronbach's alpha compare the sum of the variances of the variables to the variance of the sum as follows:

$$\alpha = \frac{k}{k-1}\left(1 - \frac{\sum_{j=1}^{k} \sigma_j^2}{\sigma_{sum}^2}\right) \tag{2}$$

where σ_j^2 is the variance of the j^{th} variable and σ_{sum}^2 is the variance of the sum of the variables. Theoretically, α should be between 0 and 1. As a general rule, $\alpha >= 0.7$ is considered to represent a reliable set of variables.

Table 1: Reliability analysis of the kinetic permeation curves (Figure 2 of the manuscript)

Membrane abbreviations	Cronbach's alpha (α)
K-GO-11.4	0.944003
P-GO-11.4	0.882823
M-GO-13.6	0.903094
P-GO-13.6	0.969949

The values of Cronbach's alpha (α) for the kinetic permeation curves are tabulated in in Table 1. As can be seen from Table 1, for all the membrane systems (K-GO-11.4, P-GO-11.4, M-GO-13.6 and P-GO-13.6) the value of α for the water permeation kinetic is greater than 0.88 which is greater than 0.7. This signifies the high reliability of the kinetic permeation curves.

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