Supplementary Information

Asymmetric Ion Transport through "Janus" MoSSe Sub-Nanometer Pores

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Methods:

Bootstrap Statistics Calculation:

In all bootstrap analyses, the original time-varying current sample is resampled 10,000 times with a sample size of 3,000. For each resampled dataset, the mean of the current is calculated, and a histogram distribution is obtained. The mean and standard deviation of the time-averaged current are then determined from this distribution to perform the noise analysis. The large number of resamples ensures that the final mean value closely approximates the original mean.

Internal Electric Field Calculation:

To calculate the internal electric field, we have assumed each layer of Mo, S and Se as a separate infinite sheet with a circular pore (red) of radius 'a' in the center, as shown in Figure M1. The infinite sheet approximation is valid as the size of the sheet is much larger than the pore radius. A point on a ring (cyan) r distance away from the center with a sheet charge density, σ,

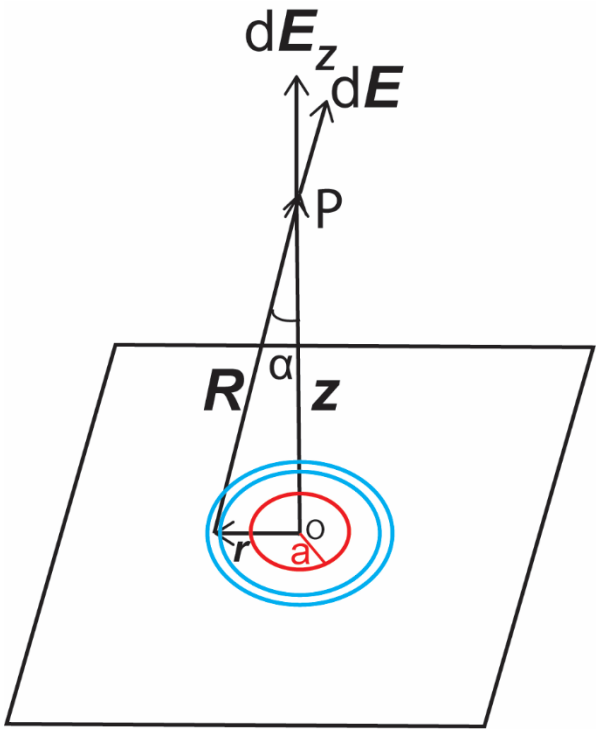

Figure M1: Internal electric field calculation with infinite sheet charge approximation.

creates an electric field d**E** at a point P. Since the ring is circularly symmetrical, the radial components cancel each other out, leaving the electric field dE_z in the z-direction.

$$d\boldsymbol{E} = \frac{1}{4\pi\epsilon}\frac{\sigma\, r\, dr\, d\phi}{|\boldsymbol{z}-\boldsymbol{r}|^2}$$

$$dE_z = \frac{1}{4\pi\epsilon}\frac{\sigma\, r\, dr\, d\phi}{z^2+r^2}\cos(\alpha) = \frac{1}{4\pi\epsilon}\frac{\sigma\, r\, dr\, d\phi}{z^2+r^2}\frac{\mathrm{z}}{\sqrt{(\mathrm{z}^2+r^2)}}$$

$$E_z = \frac{\sigma z}{4\pi\epsilon}\int_{r=a}^{r=\infty}\int_{\phi=0}^{\phi=2\pi}\frac{r\ d\phi dr}{(z^2+r^2)^{\frac{3}{2}}} = \frac{\sigma z}{4\pi\epsilon}\times 2\pi\int_{r=a}^{r=\infty}\frac{r\, dr}{(z^2+r^2)^{\frac{3}{2}}} = \frac{\sigma}{2\epsilon}\frac{z}{\sqrt{(\mathrm{z}^2+a^2)}}$$

We calculated the electric field for each layer of Mo, S and Se to obtain the resultant electric field. σ is calculated by dividing the total charge of each layer by its area. With a layer thickness of 3.31 Å, we obtain an electric field of 1.57×10^5 V/cm at the center of the structure.

Supplementary Figures:

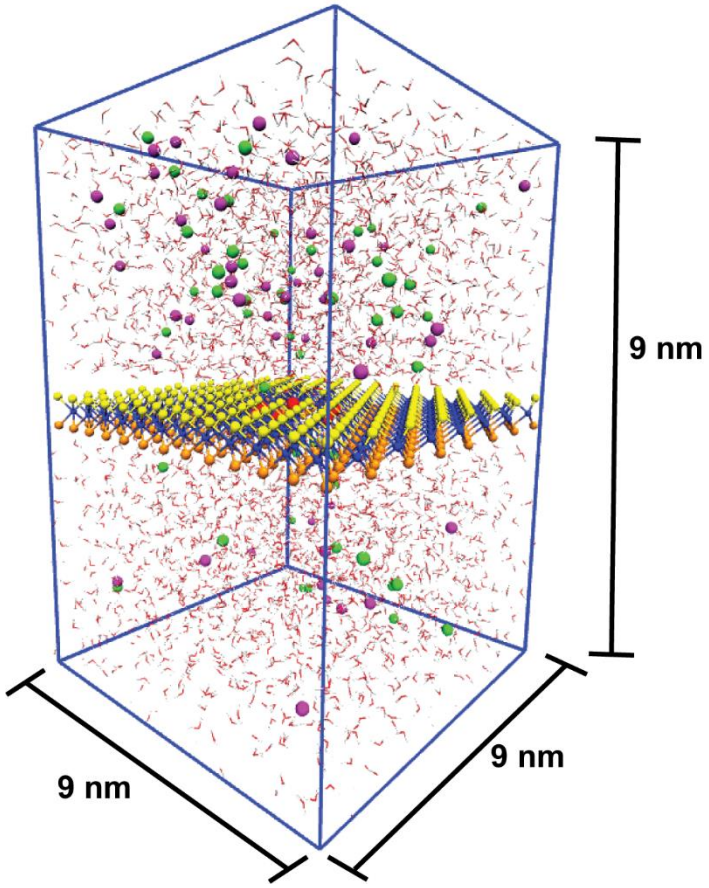

Figure S1: Three-dimensional illustration of the solvated system setup used in all molecular dynamics simulations, along with cell dimensions. Mo, S, and Se atoms are represented by blue, orange, and yellow spheres, respectively, while green and purple spheres represent K^+ and Cl^- ions, respectively. V-shaped red and white lines represent water molecules. The number of water molecules and ions has been reduced in the representation for better visualization.

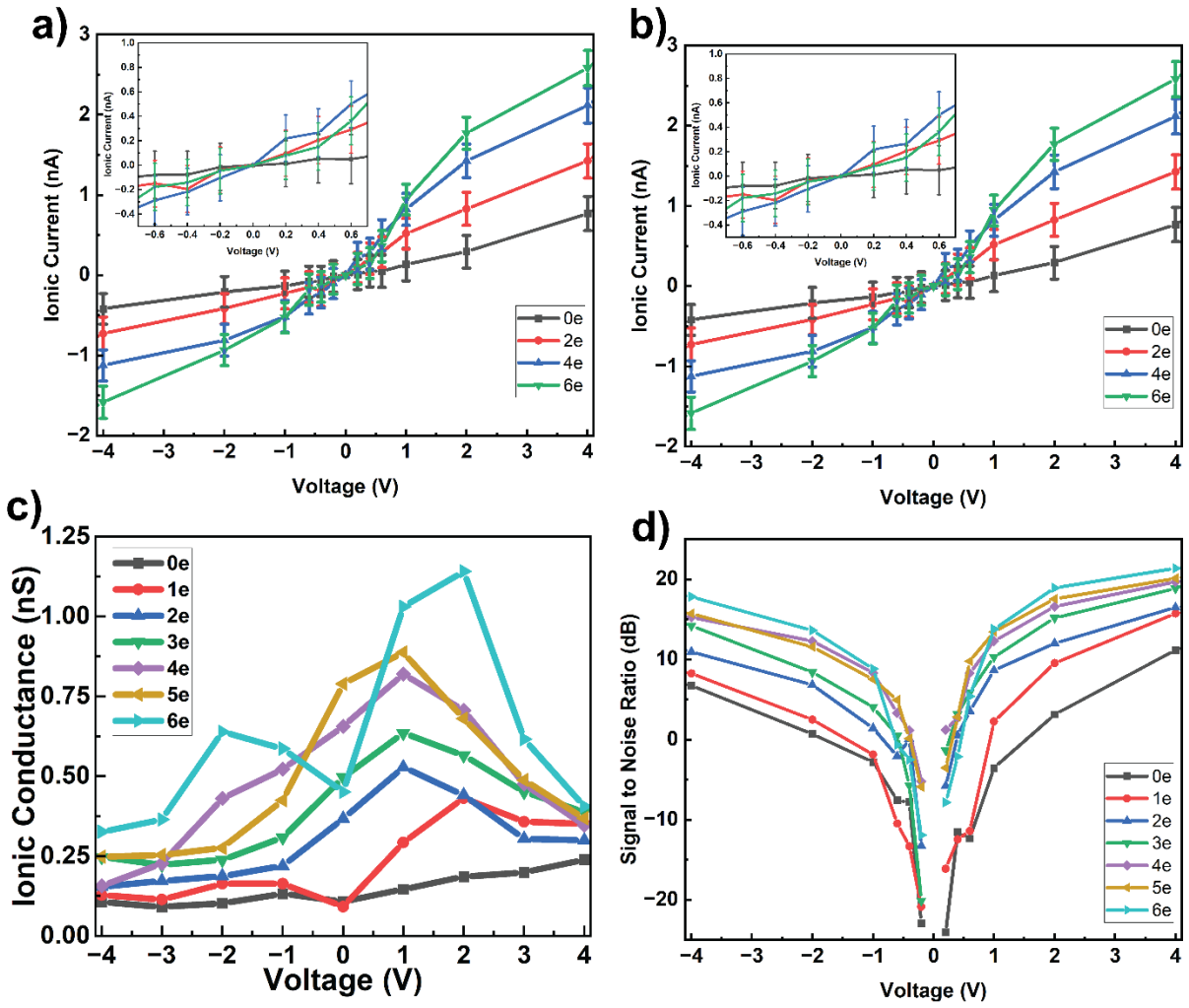

Figure S2: a) Ionic I-V characteristics of the MoSSe sub-nm pore membrane with standard error a) even charges and b) odd charges. The inset provides a close-up view of the I-V curves under low applied biases (< 1 V). c) Ionic conductance as a function of applied biases with different charge configurations. d) Signal-to-noise ratio for different applied biases with all charge configurations.

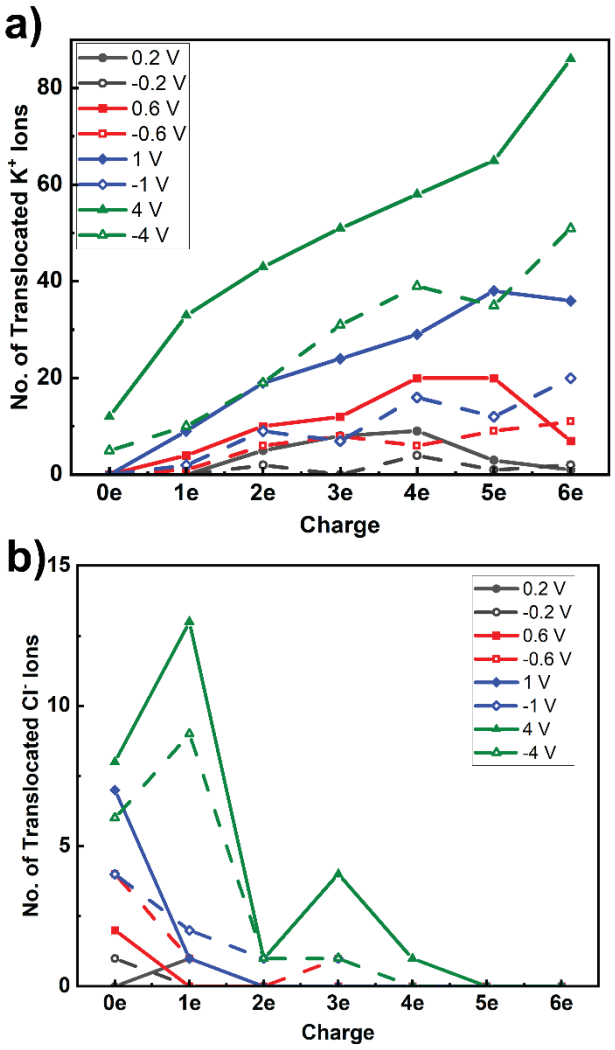

Figure S3: Number of translocated ions as a function of charges on the pore edge under different applied biases for a) K^+ ions and b) Cl^- ions. Solid and dashed lines represent forward and reverse applied biases, respectively.

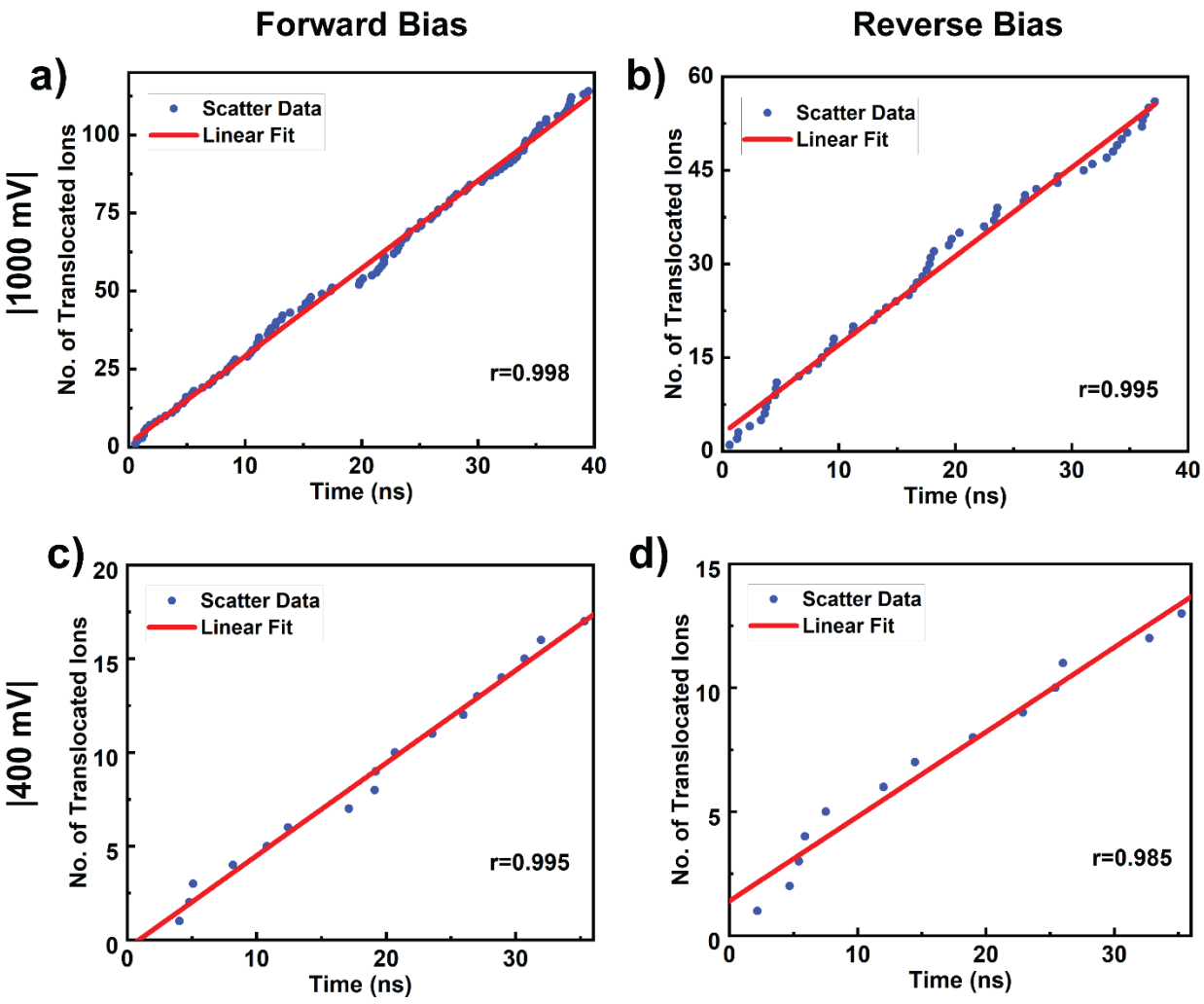

Figure S4: Number of translocated ions vs time for 6e charge configurations under applied biases of a) forward 1V, b) reverse -1 V, c) forward 400 mV, and d) reverse -400 mV. The linear fit lines (red color) show a clear linear correlation between the number of translocated ions and time, supported by Pearson's correlation coefficient (r) displayed in bottom right corner for all the cases. One notices that the points of the scatter data do not exactly start at the time origin, which results from the fact that the translocation of the first K^+ ion does not occur at t=0.

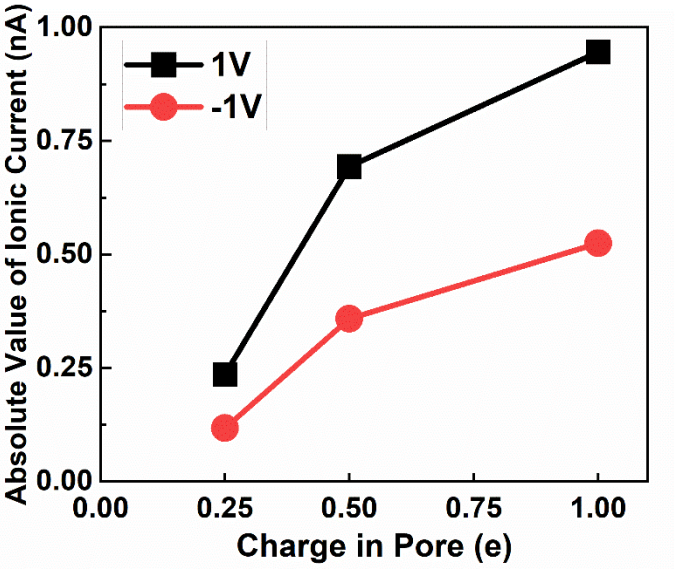

Figure S5: Absolute value of average ionic current as a function of charge deposited on each Mo atoms for 6-charge configuration under applied biases of a) forward 1V and b) reverse -1 V (negative current). The current under forward bias remains higher than the current under reverse bias across all cases.

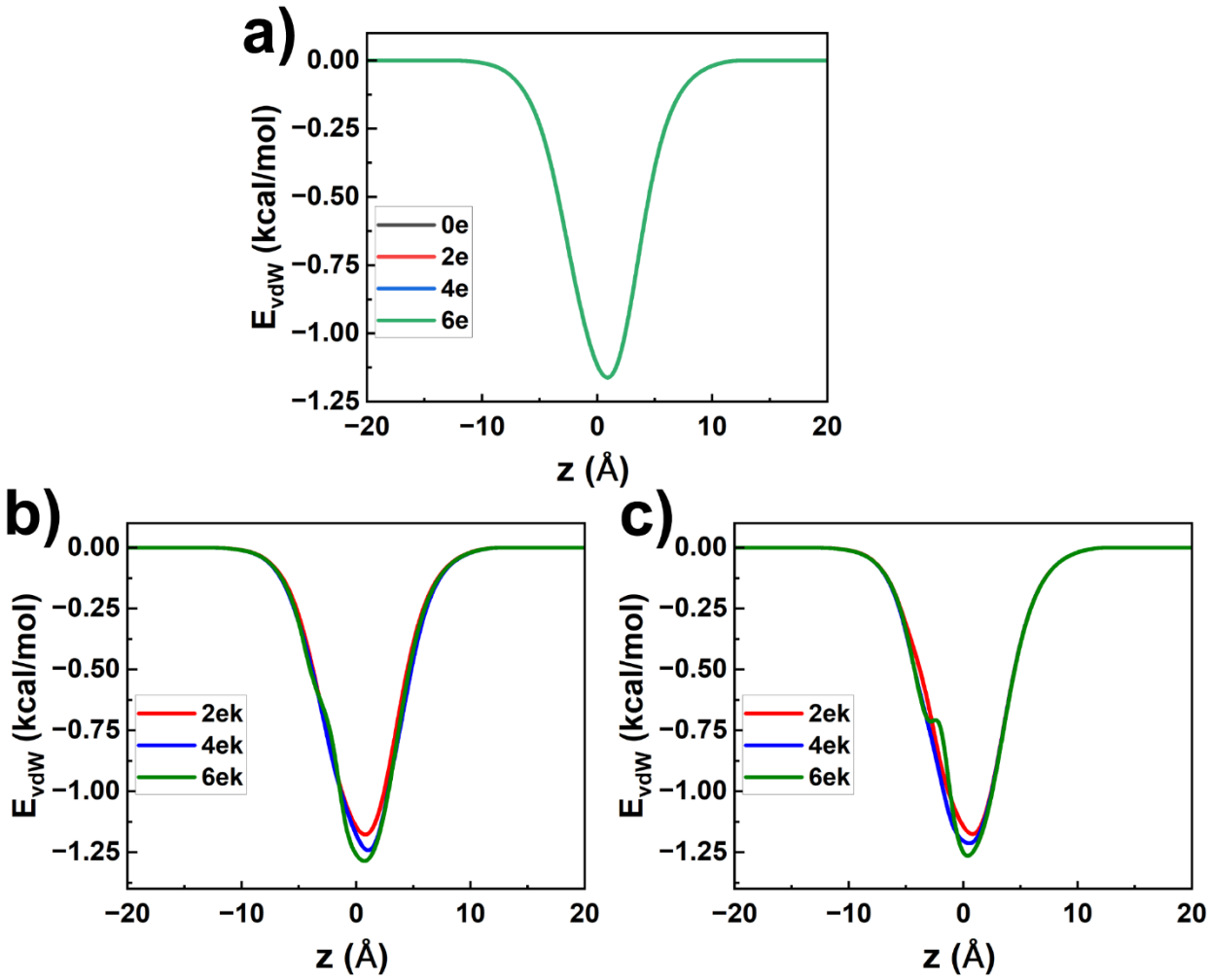

Figure S6: Van der Waals energy profiles for a K^+ ion translocating through a MoSSe sub-nm pore membrane: a) in the absence of adsorbed K^+ ions, in the presence of adsorbed K^+ ions under b) forward bias, and c) reverse bias.

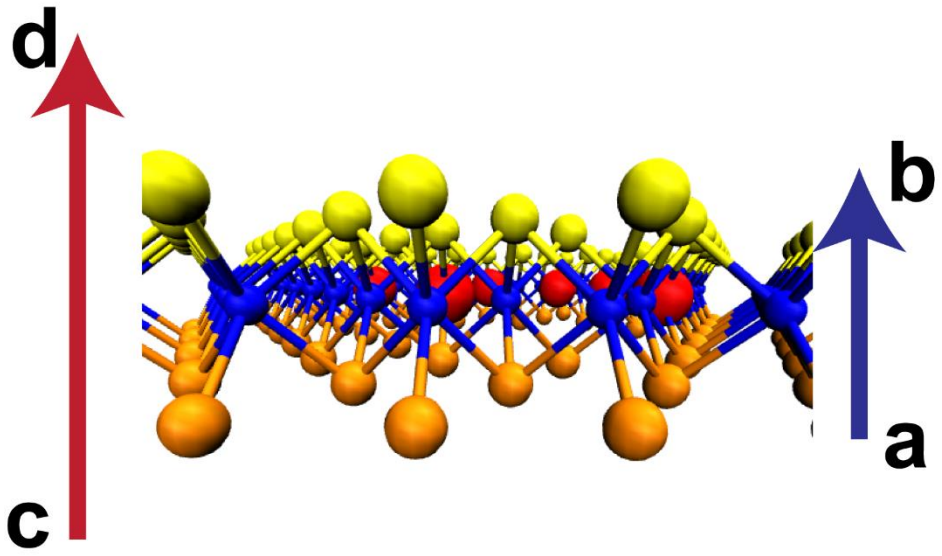

Figure S7: Definition of dwell time and total translocation time. Dwell time refers to the duration taken by a K^+ ion to traverse the MoSSe membrane thickness through the pore (from a to b). Total translocation time encompasses the adsorption time of a K^+ ion onto the membrane and the dwell time (to go from c to d).

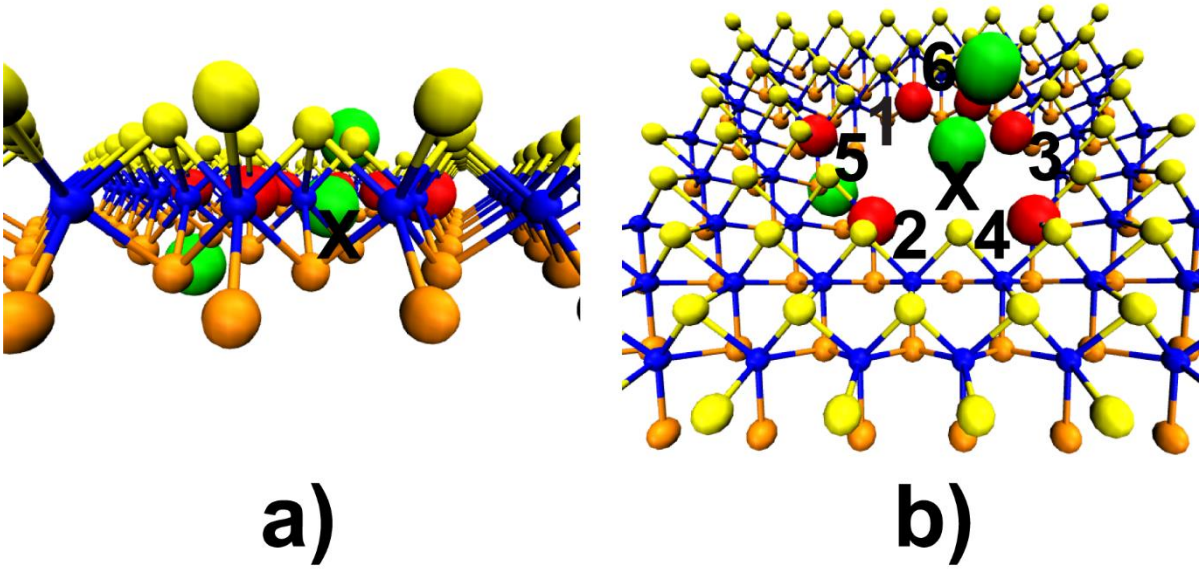

Figure S8: a) Side view and b) isometric view of the MoSSe sub-nm pore membrane, showing the K^+ ion 'X' (in green) being trapped within an arc formed by negative charges (in red) 1, 3, and 6 of the pore edge.

Supplementary Tables:

	1V			-1V		
	Median (ps)	**Mean (ps)**	**Standard Deviation (ps)**	**Median (ps)**	**Mean (ps)**	**Standard Deviation (ps)**
0e	-	-	-	-	-	-
1e	10	10.56	6.82	5	5	0
2e	10	9.47	4.68	5	10.71	8.38
3e	10	11	6.92	10	9.17	3.76
4e	10	10.9	7.08	10	11	6.6
5e	10	12.21	8.18	10	13.18	9.82
6e	17.5	21.36	12.93	17.5	22.22	16.07
Overall	10	12.7	9	10	14.57	11.89

Table S1: Median, mean, and standard deviation of dwell times for various charge configurations under 1V forward and -1V reverse applied biases.