

## Supporting Information

### **Nanofluidic Ion Regulation Membranes Based on Two-dimensional Vacancies Resided CdPS<sub>3</sub> Membrane**

Meng Zhang,<sup>‡a</sup> Chenhui Huang,<sup>‡a</sup> Zhaofeng Zhai,<sup>b</sup> Xiaomin Kang,<sup>a</sup> Jiang Ju<sup>\*c</sup> and Xitang Qian<sup>\*d</sup>

<sup>a</sup> College of Mechanical Engineering, University of South China, Hengyang 421001, P. R. China

<sup>b</sup> Institute of Metal Research, Chinese Academy of Sciences, Shenyang, 110012, P. R. China

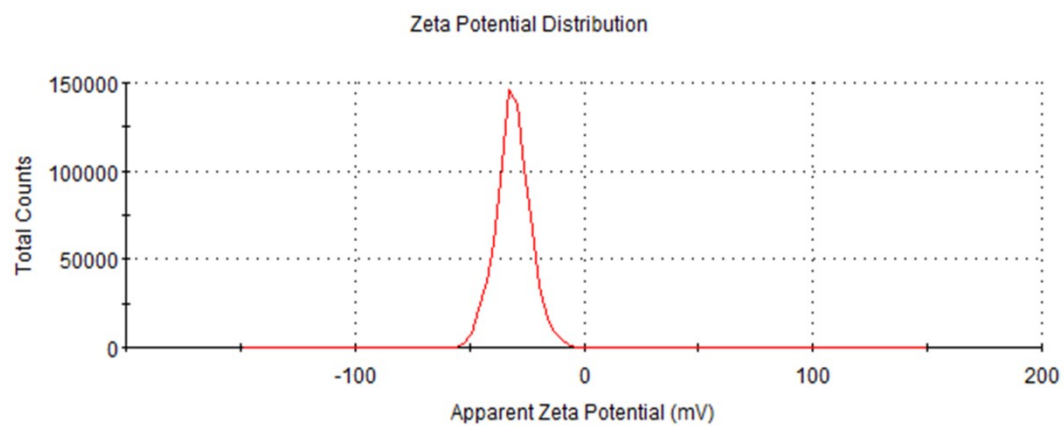
<sup>c</sup> Center for Advanced Nuclear Safety and Sustainable Development, City University of Hong Kong, Hong Kong 999077, P. R. China

<sup>d</sup> Department of Chemical and Biological Engineering, The Hong Kong University of Science and Technology, Kowloon, Hong Kong, P. R. China

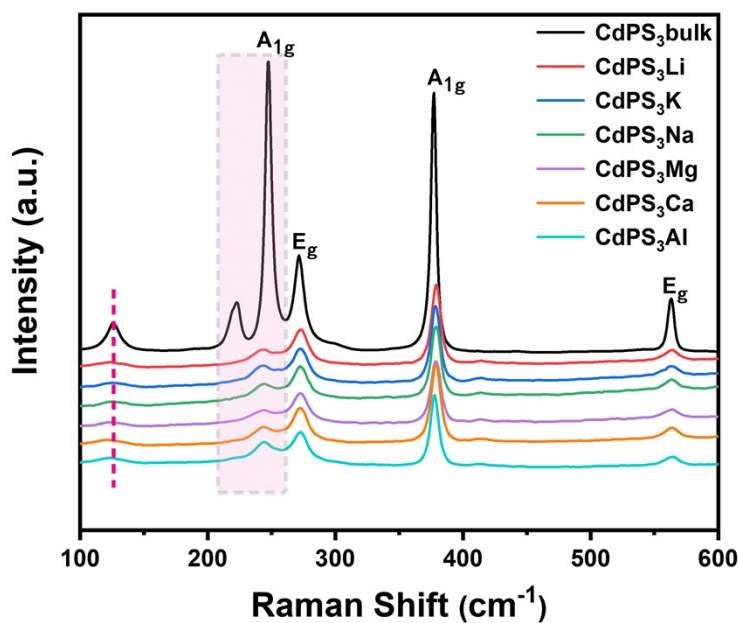
<sup>‡</sup> These authors contribute equally to this work.

\* Corresponding author emails: Xitang Qian [xtqian@ust.hk](mailto:xtqian@ust.hk) and Jiang Ju [Jiang.Ju@cityu.edu.hk](mailto:Jiang.Ju@cityu.edu.hk)

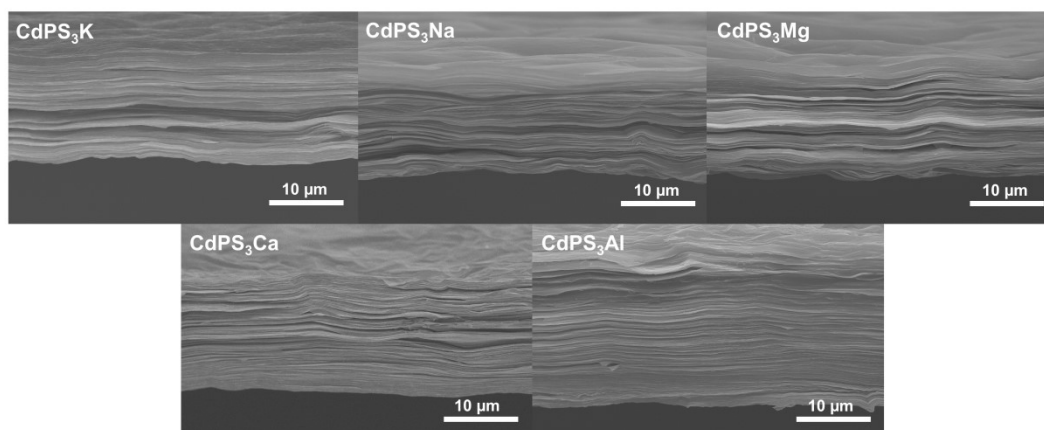
## Figures



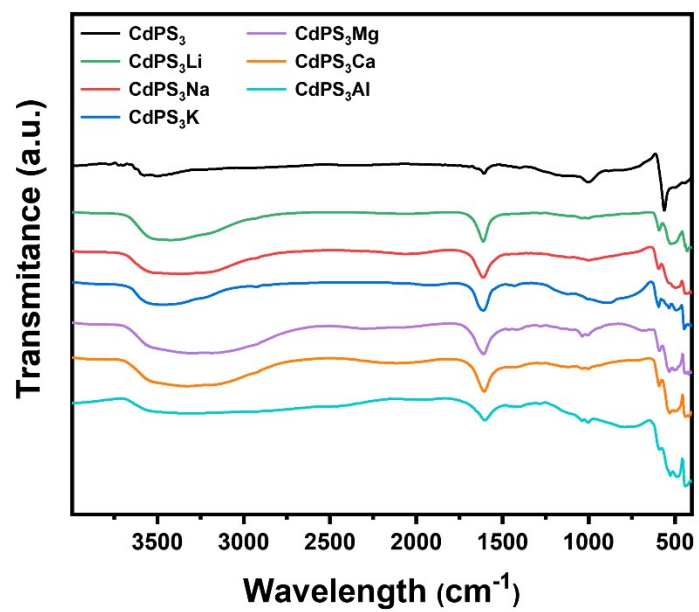
**Fig. S1.** The zeta potential diagram of CdPS<sub>3</sub>Li nanosheet dispersion.



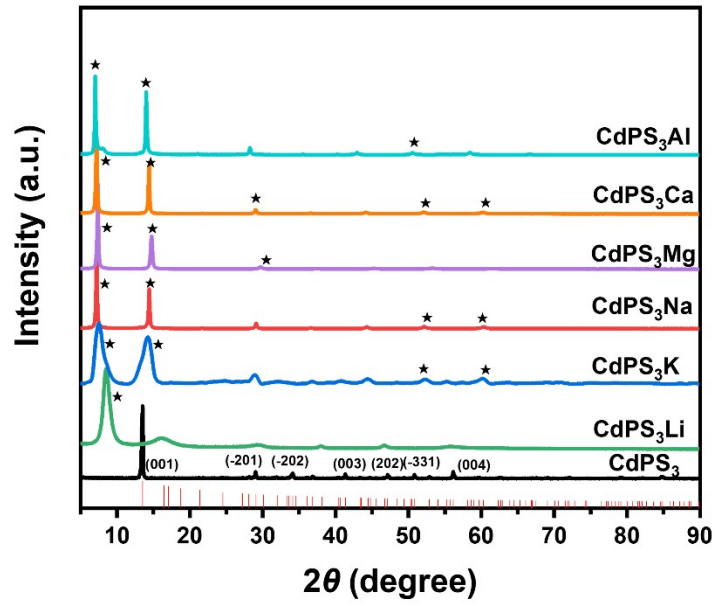
**Fig. S2.** Raman spectra of bulk CdPS<sub>3</sub> and CdPS<sub>3</sub>X. The out-of-plane vibrations of the P<sub>2</sub>S<sub>6</sub> solid exhibit the opposite motion characteristic of the S<sub>3</sub>P-PS<sub>3</sub> unit, as discerned from the strongly polarized A<sub>1g</sub> mode that appears at 378 cm<sup>-1</sup>. The Raman spectral peak at 247 cm<sup>-1</sup> also confirms the significant mode caused by the symmetric stretching vibration of the P-S bond. However, those peaks appearing at 271 and 562 cm<sup>-1</sup> represent in-plane vibrations in response to the S<sub>3</sub>P-PS<sub>3</sub> unit, exemplified by the E<sub>g</sub> mode<sup>1</sup>.



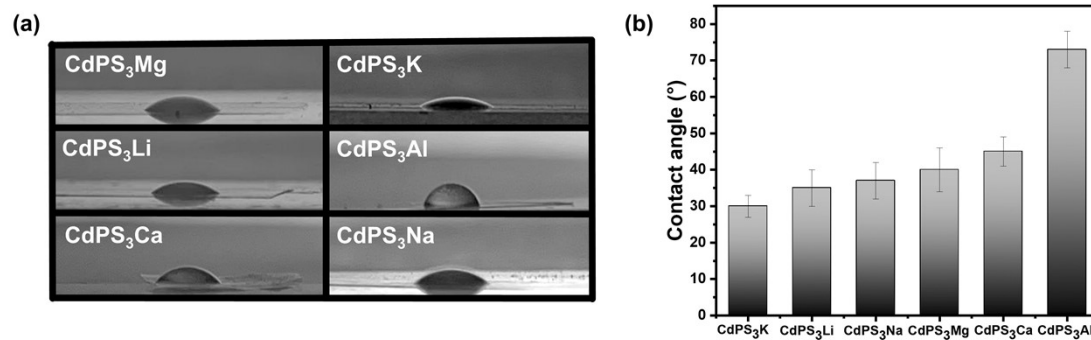
**Fig. S3.** Structure characterizations of CdPS<sub>3</sub>X membranes, showing well-ordered lamellar structures.



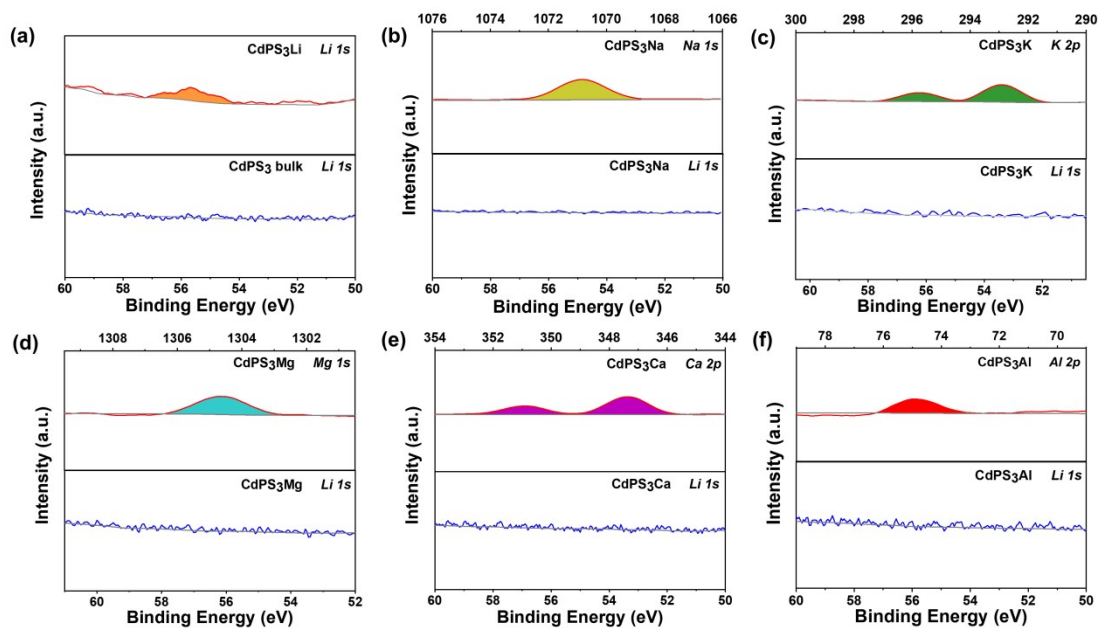
**Fig. S4.** FTIR spectra of bulk CdPS<sub>3</sub> and CdPS<sub>3</sub>X.



**Fig. S5.** XRD patterns of bulk CdPS<sub>3</sub> and CdPS<sub>3</sub>X. The theoretical X-ray diffraction spectrum of the bulk CdPS<sub>3</sub> is shown as a vertical line in the figure, the star represents the multilevel diffraction peak.

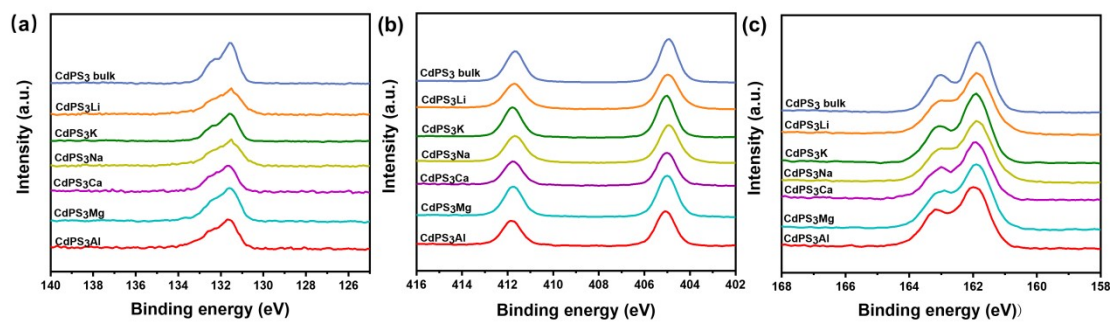


**Fig. S6.** (a) Contact angles of CdPS<sub>3</sub>X membranes towards water. (b) Wettability of CdPS<sub>3</sub>X membranes toward water.

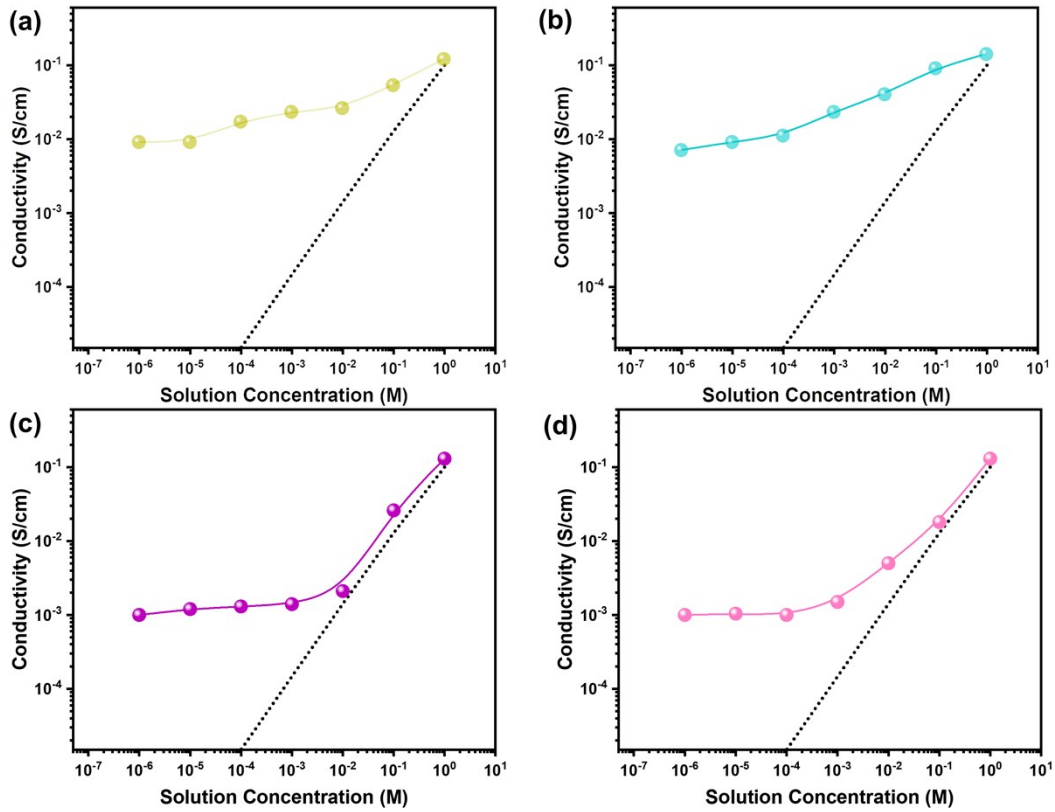


**Fig. S7.** XPS spectra of CdPS<sub>3</sub>X membranes. a-f, (a) Li *1s* orbitals in bulk CdPS<sub>3</sub> and CdPS<sub>3</sub>Li film. (b) Na *1s* orbitals and Li *1s* orbitals in the CdPS<sub>3</sub>Na film. (c) K *2p* orbitals and Li *1s* orbitals in the CdPS<sub>3</sub>K film. (d) Mg *1s* orbitals and Li *1s* orbitals in the CdPS<sub>3</sub>Mg film. (e) Ca *2p* orbitals and Li *1s* orbitals in the CdPS<sub>3</sub>Ca film. (f) Al *2p* orbitals and Li *1s* orbitals in the CdPS<sub>3</sub>Al film.

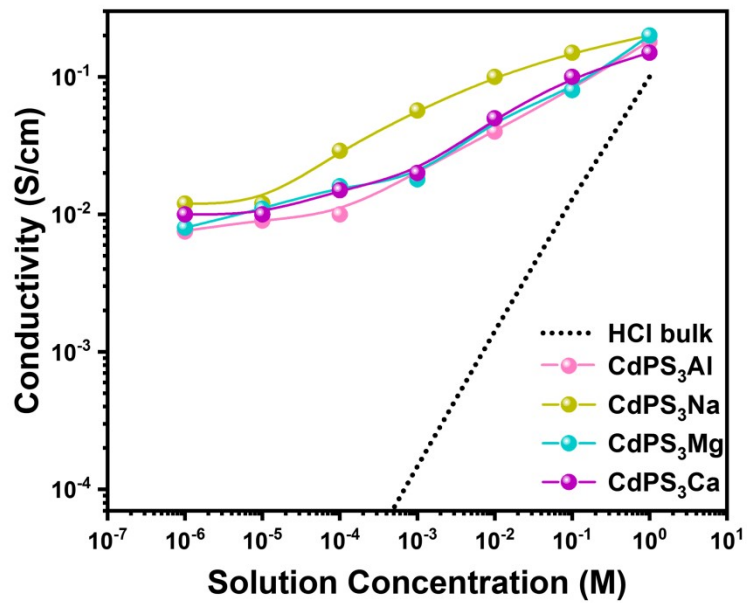




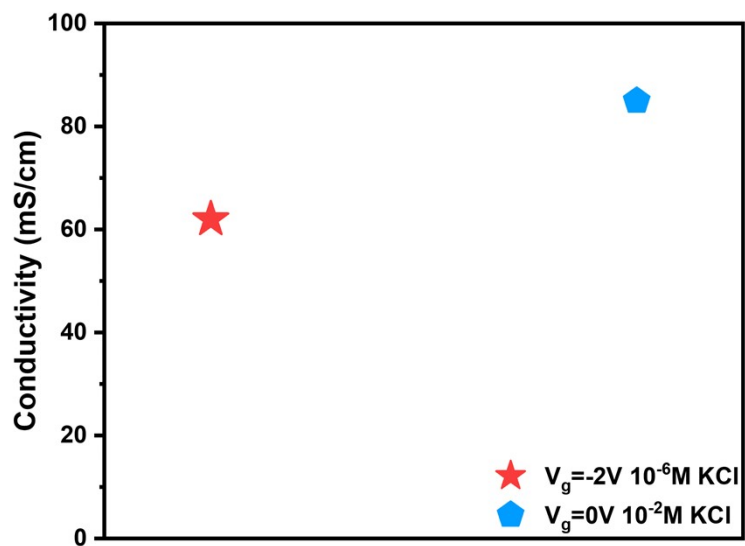
**Fig. S8.** XPS spectra of vc-CdPS<sub>3</sub> membranes. **a-c**, P 2*p* (a), Cd 3*d* (b), and S 2*p* (c) XPS spectra of CdPS<sub>3</sub>X membranes, which show that the valence states of P, Cd and S (+4, +2 and -2, respectively) remain the same in all the membranes.



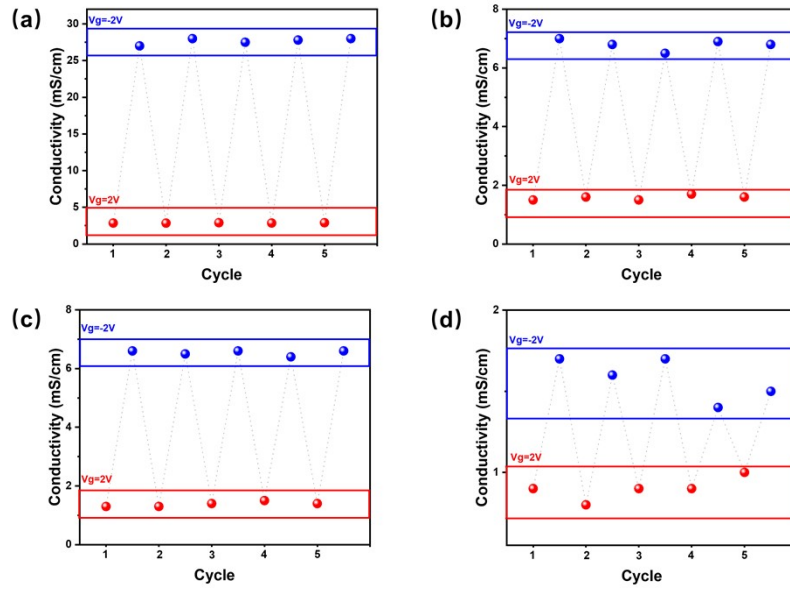
**Fig. S9.** (a) Ionic conductivity of CdPS<sub>3</sub>Na membrane in NaCl solution. (b) Ionic conductivity of CdPS<sub>3</sub>Mg membrane under MgCl<sub>2</sub> solution. (c) Ionic conductivity of CdPS<sub>3</sub>Ca membrane under calcium chloride solution. (d) Ionic conductivity of CdPS<sub>3</sub>Al membrane under aluminum chloride solution. The gray dashed lines represent the bulk conductivity of the corresponding salt solutions.



**Fig. S10.** Ionic conductivity in CdPS<sub>3</sub>X membranes at various HCl concentrations. The gray dashed lines represent the bulk conductivity of HCl solutions.



**Fig. S11.** Comparison of conductivity of CdPS<sub>3</sub>K membrane at different V<sub>g</sub>.



**Fig. S12.** (a) Plot of ions passing through CdPS<sub>3</sub>Na nanochannels as a function of time in 10<sup>-6</sup> M NaCl at different  $V_g$ . (b) Plot of ions passing through CdPS<sub>3</sub>Mg nanochannels as a function of time in 10<sup>-6</sup> M MgCl<sub>2</sub> at different  $V_g$ . (c) Plot of ions passing through CdPS<sub>3</sub>Ca nanochannels as a function of time in 10<sup>-6</sup> M CaCl<sub>2</sub> at different  $V_g$ . (d) Plot of ions passing through CdPS<sub>3</sub>Al nanochannels as a function of time in 10<sup>-6</sup> M AlCl<sub>3</sub> at different  $V_g$ .

## Tables

| <b>Material</b>                | <b>Electrolyte</b> | <b>Conductivity<br/>(mS cm<sup>-1</sup>)</b> | <b>Ref.</b> |
|--------------------------------|--------------------|--|-------------|
| BN                             | KCl solution       | 0.086  | 2           |
| Ti <sub>3</sub> C <sub>2</sub> | KCl solution       | 0.039  | 3           |
| GO                             | KCl solution       | 1  | 4           |
| Cellulose                      | KCl solution       | 2  | 5           |
| MXene                          | KCl solution       | 0.16   | 6           |
| Elastic Wood                   | KCl solution       | 0.5  | 7           |
| Montmorillonite                | KCl solution       | 0.8  | 8           |
| SLMO                           | KCl solution       | 0.6  | 9           |
| CdPS <sub>3</sub> K            | KCl solution       | 10   | This work   |

**Table. S1.** Comparison of ionic conductivity of different materials under 10<sup>-6</sup>M KCl solution (Platform conductivity).

| <b>Material</b>     | <b>Electrolyte</b> | <b>Conductivity<br/>(mS cm<sup>-1</sup>)</b> | <b>Ref.</b> |
|---------------------|--------------------|--|-------------|
| BN                  | KOH solution       | 0.037  | 2           |
| MoS <sub>2</sub>    | KOH solution       | 0.3  | 10          |
| Graphite-NFC        | KOH solution       | 1  | 11          |
| MXene               | KOH solution       | 1.2  | 6           |
| CdPS <sub>3</sub> K | KOH solution       | 20   | This work   |

**Table. S2.** Comparison of ionic conductivity of different materials under 10<sup>-6</sup>M KOH solution (Platform conductivity).

| <b>Material</b>      | <b>Electrolyte</b> | <b>Conductivity<br/>(mS cm<sup>-1</sup>)</b> | <b>Ref.</b> |
|----------------------|--------------------|--|-------------|
| BN                   | HCl solution       | 0.6  | 2           |
| MXene                | HCl solution       | 1  | 6           |
| Montmorillonite      | HCl solution       | 1.4  | 8           |
| GO                   | HCl solution       | 2.5  | 4           |
| Graphite-NFC         | HCl solution       | 3  | 11          |
| LGM                  | HCl solution       | 4.5  | 12          |
| Vermiculite          | HCl solution       | 5.6  | 13          |
| HA-GO                | HCl solution       | 7  | 14          |
| SLMO                 | HCl solution       | 10   | 9           |
| CdPS <sub>3</sub> Al | HCl solution       | 7.5  | This work   |
| CdPS <sub>3</sub> Ca | HCl solution       | 8  | This work   |
| CdPS <sub>3</sub> Mg | HCl solution       | 10   | This work   |
| CdPS <sub>3</sub> Na | HCl solution       | 12   | This work   |
| CdPS <sub>3</sub> K  | HCl solution       | 23   | This work   |

**Table. S3.** Comparison of ionic conductivity of different materials under 10<sup>-6</sup>M HCl solution (Platform conductivity).



| <b>Material</b>                | <b>Electrolyte</b> | <b>Conductivity<br/>(mS cm<sup>-1</sup>)</b> | <b>Ref.</b> |
|--------------------------------|--------------------|--|-------------|
| BN                             | NaCl solution      | 0.07   | 2           |
| Ti <sub>3</sub> C <sub>2</sub> | NaCl solution      | 0.032  | 3           |
| Graphite-NFC                   | NaCl solution      | 1  | 11          |
| BN-NFC                         | NaCl solution      | 0.18   | 15          |
| SLMO                           | NaCl solution      | 0.2  | 9           |
| CdPS <sub>3</sub> Na           | NaCl solution      | 9  | This work   |

**Table. S4.** Comparison of ionic conductivity of different materials under 10<sup>-6</sup> M NaCl solution (Platform conductivity).

| Material                       | Electrolyte                | Conductivity<br>(mS cm <sup>-1</sup> ) | Ref.      |
|--------------------------------|----------------------------|--|-----------|
| BN                             | CaCl <sub>2</sub> solution | 0.035                                  | 2         |
| GO                             | CaCl <sub>2</sub> solution | 0.6                                    | 4         |
| CdPS <sub>3</sub> Ca           | CaCl <sub>2</sub> solution | 1                                      | This work |
| Ti <sub>3</sub> C <sub>2</sub> | AlCl <sub>3</sub> solution | 0.067                                  | 3         |
| CdPS <sub>3</sub> Al           | AlCl <sub>3</sub> solution | 1                                      | This work |
| CdPS <sub>3</sub> Mg           | MgCl <sub>2</sub> solution | 6                                      | This work |

**Table. S5.** Comparison of ionic conductivity of different materials in 10<sup>-6</sup> M CaCl<sub>2</sub>, MgCl<sub>2</sub>, and AlCl<sub>3</sub> solutions (Platform conductivity).

## References

1. M. Naguib, M. Kurtoglu, V. Presser, J. Lu, J. Niu, M. Heon, L. Hultman, Y. Gogotsi and M. W. Barsoum, *Advanced Materials*, 2011, 23, 4248-4253.
2. S. Qin, D. Liu, G. Wang, D. Portehault, C. J. Garvey, Y. Gogotsi, W. Lei and Y. Chen, *Journal of the American Chemical Society*, 2017, 139, 6314-6320.
3. J. Lao, R. Lv, J. Gao, A. Wang, J. Wu and J. Luo, *Acs Nano*, 2018, 12, 12464-12471.
4. K. Raidongia and J. Huang, *Journal of the American Chemical Society*, 2012, 134, 16528-16531.
5. T. Li, S. X. Li, W. Kong, C. Chen, E. Hitz, C. Jia, J. Dai, X. Zhang, R. Briber, Z. Siwy, M. Reed and L. Hu, *Science Advances*, 2019, 5.

6. Y. Wang, H. Zhang, Y. Kang, Y. Zhu, G. P. Simon and H. Wang, *Acs Nano*, 2019, 13, 11793-11799.
7. C. Chen, J. Song, J. Cheng, Z. Pang, W. Gan, G. Chen, Y. Kuang, H. Huang, U. Ray, T. Li and L. Hu, *Acs Nano*, 2020, 14, 16723-16734.
8. M.-L. Liu, M. Huang, L.-Y. Tian, L. H. Zhao, B. Ding, D. B. Kong, Q.-H. Yang and J.-J. Shao, *Acs Applied Materials & Interfaces*, 2019, 11, 6665-6665.
9. H. Jin, J. Li, Z. Xu, Z. Hu, K. Liu, K. Liu, J. Duan, B. Hu, L. Huang and J. Zhou, *Science China-Materials*, 2022, 65, 2578-2584.
10. J. Park, S. Bhoyate, Y.-H. Kim, Y.-M. Kim, Y. H. Lee, P. Conlin, K. Cho and W. Choi, *Acs Nano*, 2021, 15, 12267-12275.
11. Y. Zhou, C. Chen, X. Zhang, D. Liu, L. Xu, J. Dai, S.-C. Liou, Y. Wang, C. Li, H. Xie, Q. Wu, B. Foster, T. Li, R. M. Briber and L. Hu, *Journal of the American Chemical Society*, 2019, 141, 17830-17837.
12. K. Luo, T. Huang, Q. Li, J. Lao, J. Gao and Y. Tang, *Rsc Advances*, 2022, 12, 29640-29646.
13. Y. Wu, T. Zhou, Y. Wang, Y. Qian, W. Chen, C. Zhu, B. Niu, X.-Y. Kong, Y. Zhao, X. Lin, L. Jiang and L. Wen, *Nano Energy*, 2022, 92.
14. T. J. Konch, R. K. Gogoi, A. Gogoi, K. Saha, J. Deka, K. A. Reddy and K. Raidongia, *Materials Chemistry Frontiers*, 2018, 2, 1647-1654.
15. L. Yu, T. Gao, R. Mi, J. Huang, W. Kong, D. Liu, Z. Liang, D. Ye and C. Chen, *Nano Research*, 2023, 16, 7609-7617.