## Ultrahigh Inter-Cation Selectivity from Chemically Nanoconfined Fluidic Channels

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Overcoming the structural limitations at the nanoscale presents a significant challenge in the design of membranes for effective and scalable separation processes. In this context, nanostructured graphene-based membranes emerge as a promising solution, offering a versatile platform that can be tailored to meet specific requirements. However, the high yield and processability of graphene oxides (GOs) introduce complexities in controlling their features and properties during synthesis. This study aims to explore the material properties of various GOs to gain a deeper understanding of the key characteristics that influence their performance in ionselective membranes. Notably, we employ a multidisciplinary approach for the first time to reveal how structural and chemical features impact ion transport at the nanoscale, ultimately enhancing the monovalent-selective capacity of GO laminates. Our findings show that in a low defect regime, the prevalent concentration of organosulfate groups-often resulting from conventional synthesis methods-plays a crucial role beyond mere steric effects. These groups significantly contribute to improved ionic selectivity between mono- and multivalent ions. We emphasize that these stronger interlayer interactions fostered by organosulfate groups provide an effective means to control membrane swelling with angstrom precision. As a result, we observe a remarkable four-fold increase in K+/Mg<sup>2+</sup> inter-cation selectivity, achieving 94 % Mg<sup>2+</sup> rejection under osmotic transport conditions. Furthermore, even when interplanar distances are expanded to boost water fluxes, the inter-cation selectivity remains competitive with that of state-of-the-art membranes.

## References

- [1] S. Hong *et al.*, Nano Lett., 17 (2017) 728
- [2] S. Hong et el., ACS Mater. Lett., 5 (2023) 341

## Figures

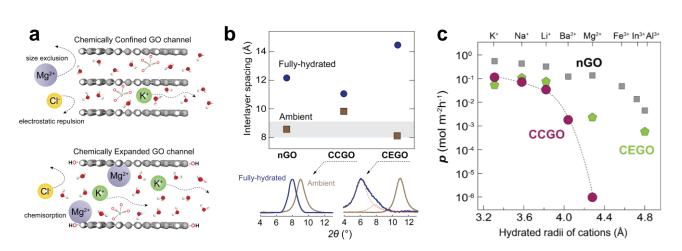


Figure 1: Ion-selective transport through chemically nanoconfined fluidic channels in graphene oxide-based membranes