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Diffusion limitations and translocation barriers in atomically thin biomimetic pores

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Ionic transport in nano- to sub-nano-scale pores is highly dependent on translocation barriers and potential wells. These features in the free-energy landscape are primarily the result of ion dehydration and electrostatic interactions. For pores in atomically thin membranes, such as graphene, other factors come into play. Ion dynamics both inside and outside the geometric volume of the pore can be critical in determining the transport properties of the channel due to several commensurate length scales, such as the effective membrane thickness, radii of the first and the second hydration layers, pore radius, and Debye length. In particular, for biomimetic pores, such as the graphene crown ether we examine, there are regimes where transport is highly sensitive to the pore size due to the interplay of dehydration and interaction with pore charge. Picometer changes in the size, e.g., due to a minute strain, can lead to a large change in conductance. Outside of these regimes, the small pore size itself gives a large resistance even when electrostatic factors and dehydration compensate each other to give a relatively flat - e.g., near barrierless - free energy landscape. The permeability, though, can still be large and ions will translocate rapidly after they arrive within the capture radius of the pore. This, in turn, leads to diffusion and drift effects dominating the conductance. The current thus plateaus and becomes effectively independent of pore free energy characteristics. Measurement of this effect will give an estimate of the magnitude of kinetically-limiting features and experimentally constrain the local electromechanical conditions.

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