Supplementary Material: Ionic phenomena in nanoscale pores through 2D materials

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I. CONDUCTANCE VERSUS PORE RADIUS FROM EXPERIMENT

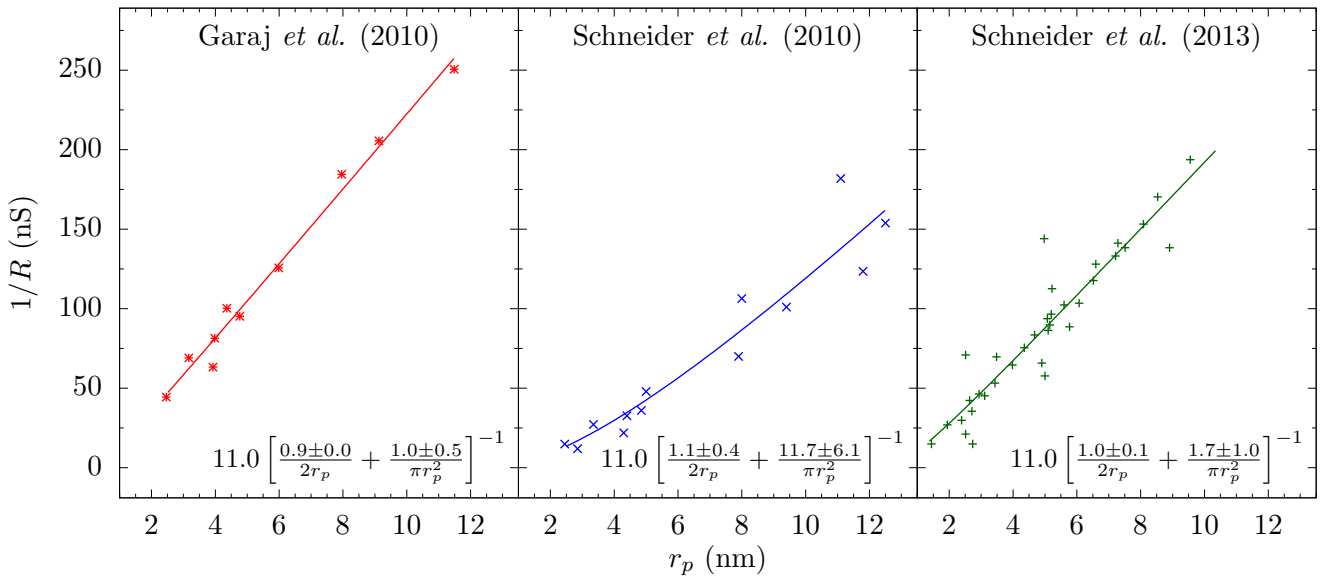

FIG. 1 Conductance fits for experimental data from Garaj *et al.* (2010), Schneider *et al.* (2010), and Schneider *et al.* (2013). Note that the fits here are slightly different than ones given in the respective papers, as indicated in the main text. However, the basic conclusion is the same: Garaj *et al.* (2010) and Schneider *et al.* (2013) obtained a dominant access contribution, whereas Schneider *et al.* (2010) obtained a dominant pore contribution. The pore radii in these experiments are measured using the TEM image of the pores which roughly corresponds to r_p in Fig. 7 of the main text – i.e., the average distance from the pore center to the pore edge excluding the carbon electron cloud (approximately its vdW radius). However, for the size of the pores presented here, the difference between the various definitions of pore radius is not significant (similarly for the exact pore geometry).

II. CONDUCTANCE VERSUS PORE RADIUS FROM ALL-ATOM MOLECULAR DYNAMICS

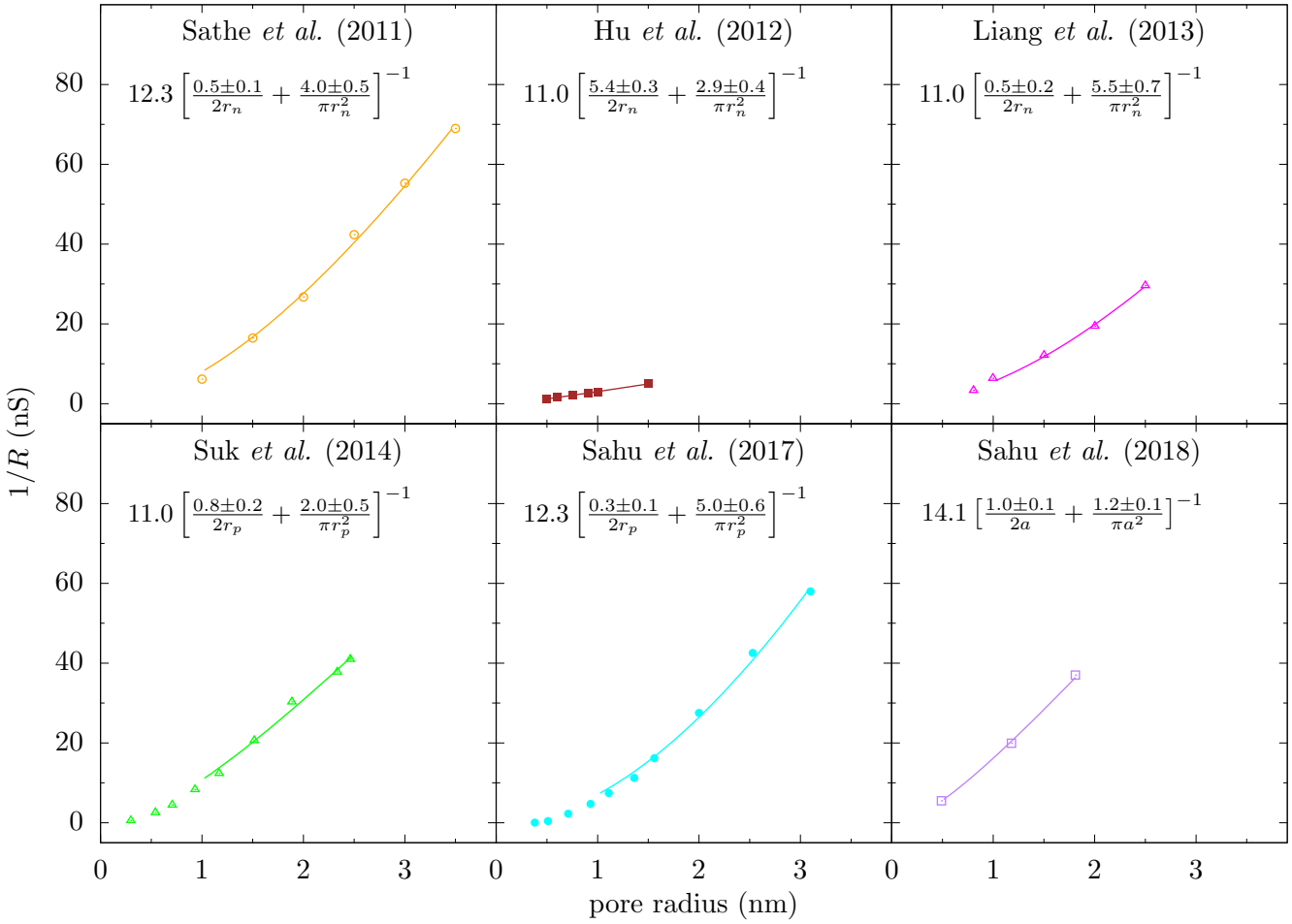

FIG. 2 Conductance fits for all-atom MD data from Sathe *et al.* (2011), Hu *et al.* (2012), Liang *et al.* (2013), Suk and Aluru (2014), Sahu *et al.* (2017), and Sahu and Zwolak (2018). The definition of pore radius varies in these studies. Sathe *et al.* (2011), Hu *et al.* (2012), and Liang *et al.* (2013) take the pore radius as the nominal radius employed in cutting the pore – i.e., all carbon atoms with coordinates that satisfied $r_n^2 \leq x^2 + y^2$ were removed to create the pore and r_n was taken as pore radius (note that r_n is not unique with this definition, a range of r_n give the same physical pore. We use the values as given in th respective papers). Suk and Aluru (2014) take the pore radius as the distance from the pore center to where the water density dropped below 2 % of the bulk water density. Sahu *et al.* (2017) define the pore radius as the average distance from the center of the pore to the inner edge of the pore atoms, i.e., r_p as shown with red dashed line in Fig. 7 of the main text. Both the Suk and Aluru (2014) and Sahu *et al.* (2017) definitions give similar values for the pore radius. However, Sahu and Zwolak (2018) define the pore radius from the current density profile in the pore, as shown by a in Fig. 7 of the main text. Solid lines are shown where data was fit. We only fit the data in which the pore is large enough to ignore the effects of hydration, except in the case of Hu *et al.* (2012), which only has small pores. We note that dehydration increases the pore resistance drastically and thus overshadows the access resistance. For instance, in the case of Sahu *et al.* (2017), only the dehydration regime was of interest. Error bars were not considered in the fits since not all the data were reported with error bars (in particular, the ones in the top panel).

III. PHYSICAL/CHEMICAL PROPERTIES AND APPLICATIONS

	Graphene	hBN	MoS_2
Thickness (vdW)	0.335 nm	0.334 nm	0.65 nm
Young's modulus (Breaking strength)	1 TPa (130 GPa) (Lee *et al.*, 2008)	0.27 TPa (23 GPa) (Bertolazzi *et al.*, 2011)	0.865 TPa (70.5 GPa) Falin *et al.* (2017)
Wettability	Hydrophobic (Schneider *et al.*, 2013)	Mild hydrophilic (Zhou *et al.*, 2013)	Hydrophilic (Liu *et al.*, 2013b)
Surface charge density	-0.6 C/m^2(pH 8) (Rollings *et al.*, 2016) -0.039 C/m^2(pH 5) (Shan *et al.*, 2013)	-0.07C/m^2 to -0.16 C/m^2(pH 7) (Weber *et al.*, 2017)	-0.024 C/m^2 to -0.088 C/m^2 (pH 5) (Feng *et al.*, 2016a)

TABLE I Comparison of the physical/chemical properties 2D materials relevant to topics covered in the main text. We list here the atomic thickness – defined including the van der Waals radii – for a simple comparison. The effective thickness for ion transport can be significantly larger than the thicknesses in this table and it depends on, for example, voltage (see the main text). The large value of Young's modulus and breaking strength demonstrates the extraordinary strength of 2D materials. For comparison, the Young's modulus and breaking strength of steel are 200 GPa and 500 MPa, respectively (Ledbetter *et al.*, 1980). The hydrophobicity/hydrophilicity listed in the table are for the nanopore in the membrane rather than the flat surface of the membrane; thus, although MoS_2 membrane is hydrophobic, the pore is hydrophilic due to Mo-rich edges. The surface charge densities listed here are just a few representative examples as it can vary widely based on the fabrication technique (e.g., exfoliated versus chemical vapor deposition growth of the material), defect density, and other environmental factors (pH , for instance). Additionally, the charge density for porous membrane can be different than that for a pristine membrane. As a point of comparison, the surface charge for silicon nitride is typically reported to be around 20 mC/m^2 to 60 mC/m^2 at pH 7 to pH 8 (Ho *et al.*, 2005; Smeets *et al.*, 2006).

Graphene	Gas separation (Celebi *et al.*, 2014; Koenig *et al.*, 2012), Osmotic power (Gai *et al.*, 2014; Walker *et al.*, 2017), Desalination(Cohen-Tanugi and Grossman, 2012; Rollings *et al.*, 2016; Surwade *et al.*, 2015), Sensing and sequencing (Garaj *et al.*, 2010; Merchant *et al.*, 2010; Schneider *et al.*, 2010) (Heerema and Dekker, 2016; Sathe *et al.*, 2011; Wells *et al.*, 2012)
GO	Desalinaiton (Abraham *et al.*, 2017; Chen *et al.*, 2017; Joshi *et al.*, 2014; Nair *et al.*, 2012), Gas separation (Li *et al.*, 2013), Solvent recovery (Yang *et al.*, 2017), Osmotic power (Ji *et al.*, 2017)
MoS_2	Desalination (Hirunpinyopas *et al.*, 2017), Osmotic power (Feng *et al.*, 2016a) Sensing and sequencing (Feng *et al.*, 2015; Liu *et al.*, 2014)
hBN	Sensing and sequencing (Liu *et al.*, 2013a; Zhou *et al.*, 2013) Osmotic power (Walker *et al.*, 2017)
2D-heterostructures	Desalination (Esfandiar *et al.*, 2017; Gopinadhan *et al.*, 2019)
Biomimetic & fundamental studies	Ion channel mechanisms (Sahu *et al.*, 2019) and biomimetics (He *et al.*, 2013; Sint *et al.*, 2008), Ionic coulomb blockade (Feng *et al.*, 2016b) Dehydration (exp) (Abraham *et al.*, 2017; Esfandiar *et al.*, 2017; Joshi *et al.*, 2014) Dehydration (th) (Sahu *et al.*, 2017; Sahu and Zwolak, 2017)

TABLE II Various applications of 2D material and their derivatives. This list is not exhaustive. It includes the papers we discussed in the main text but these applications are often touched on in many studies and are part of larger and very active research fields. Many fundamental studies have their origin in related work with traditional solid-state pores, see the main text and the references in the citations above.

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