Electronic Supplementary Material (ESI) for Nanoscale Advances.

Supplementary Information

Percolative Proton Transport in Hexagonal Boron Nitride Membranes with Edge-functionalization

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1. Functionalization of h-BN

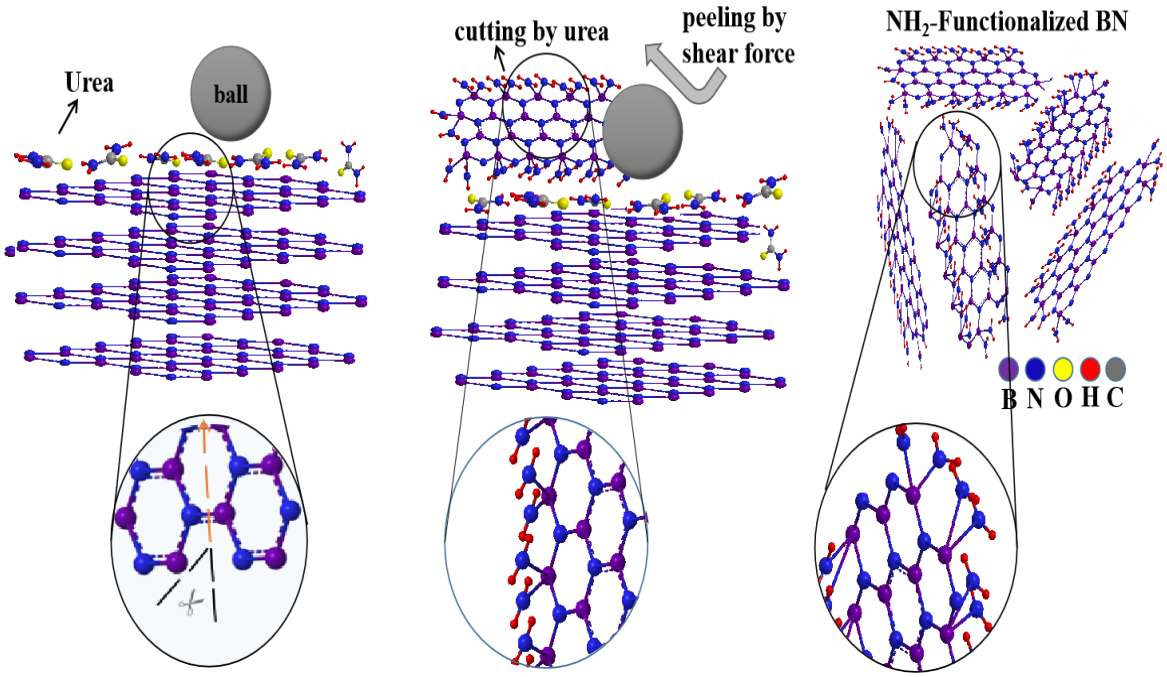

Figure S1: Schematic illustration of solid-state ball milling commercially available h-BN and urea powder.

Ball milling of pristine h-BN with urea particles under inert atmosphere results in size reduction of h-BN particles due to high shear forces and high energy collisions with the stainless steel balls. Edge functionalization of h-BN with amine groups is also realized.

2. Zeta potential of the h-BN nanosheets

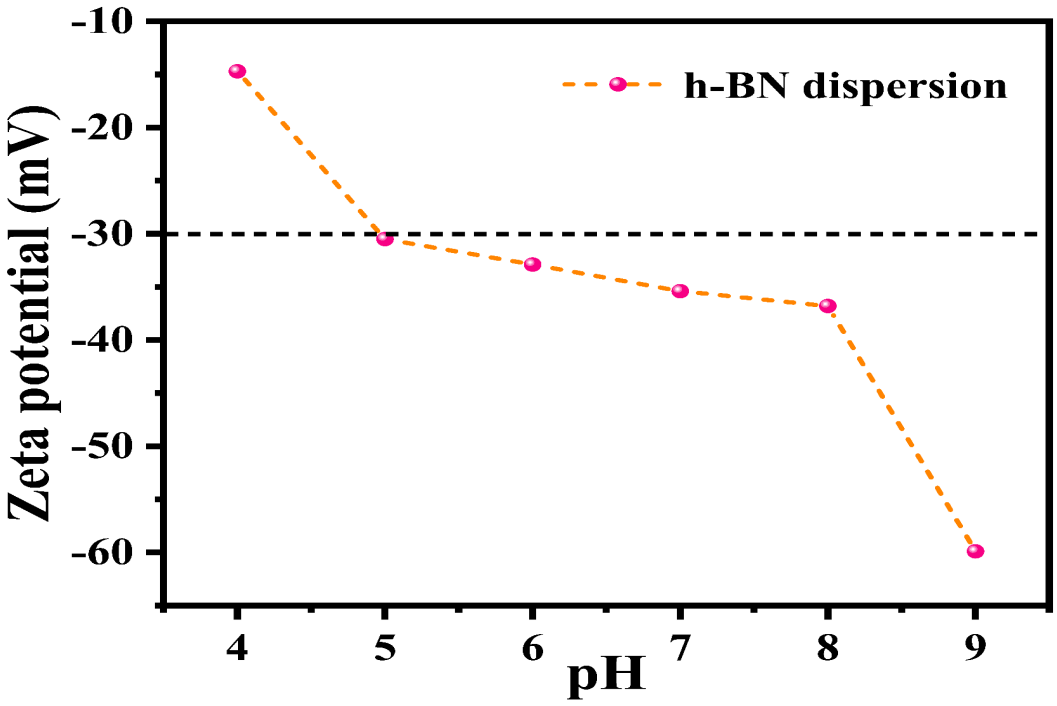

Figure S2: Zeta potential of the h-BN nanosheets in colloidal dispersion in a pH range of 4-9

Surface charges of functionalized h-BN were confirmed by zeta potential measurements. Zeta potentials of dispersion became increasingly negative as pH varied from 4 to 9. The zeta potential variation with pH confirmed surface charges' strong dependence on pH. A stable suspension has a zeta potential value of either +30 mV or higher or -30 mV or lower.

3.SEM images of the functionalized h-BN

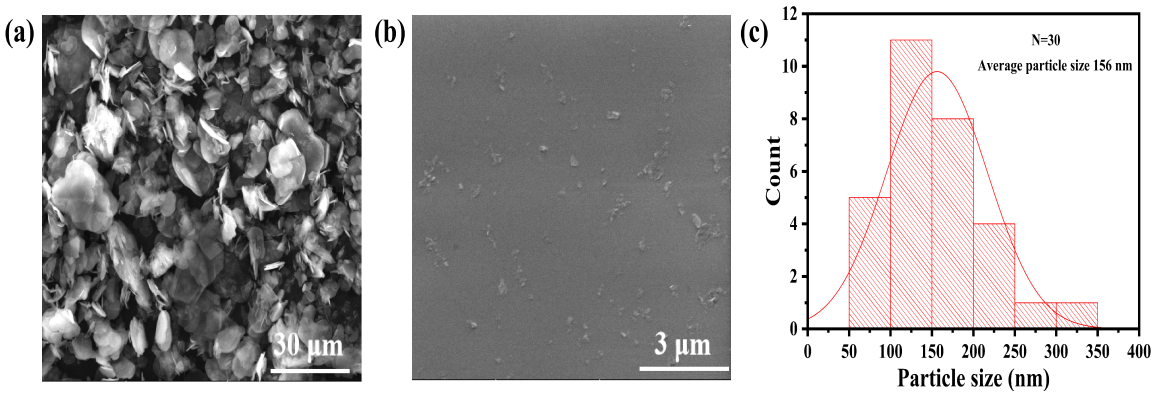

Figure S3: **(a)**,**(b)**, SEM images of pristine h-BN and functionalized h-BN, respectively. **(c)**, the statistical distribution of lateral size for functionalized h-BN sheets.

The lateral size of the flakes was calculated using ImageJ. Overlapping flakes were not considered during the ImageJ analysis. The statistical distribution of the lateral size provides that most of the h-BN flakes have a lateral size of 100-200 nm.

4. XPS spectra for pristine & less functionalized h-BN

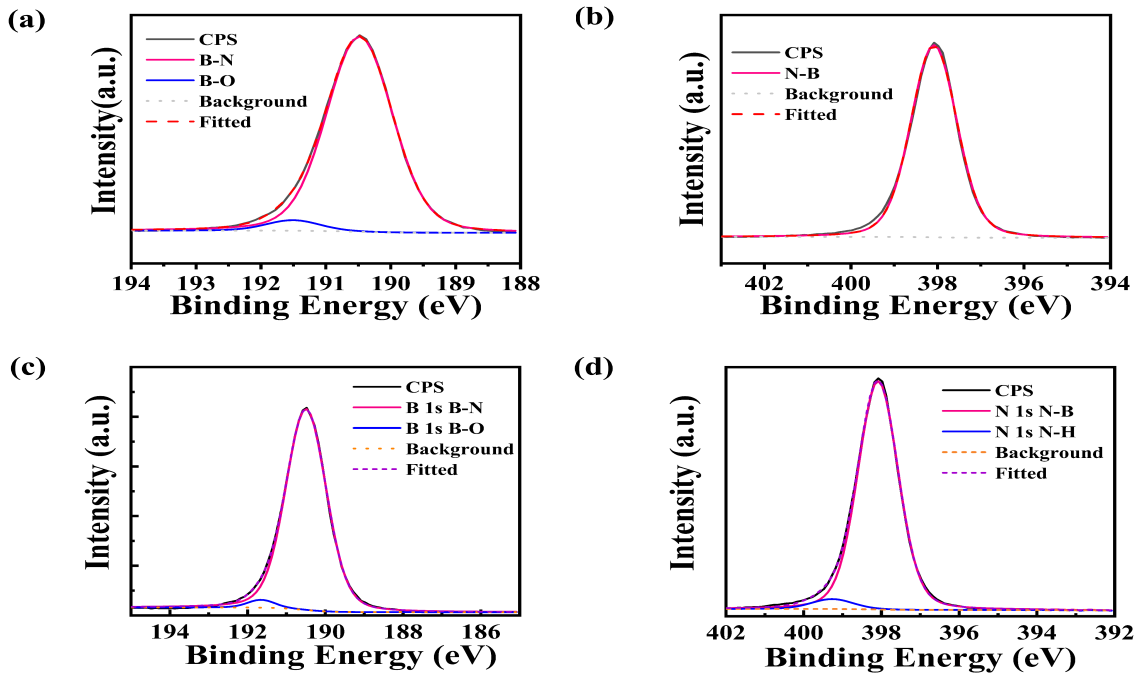

Figure S4: XPS spectra of pristine h-BN **(a)** Core-level B1s, **(b)** Core-level N1s, and less functionalized h-BN **(c)** Core-level B1s, **(d)** Core-level N1s.

5. TGA curve of functionalized h-BN.

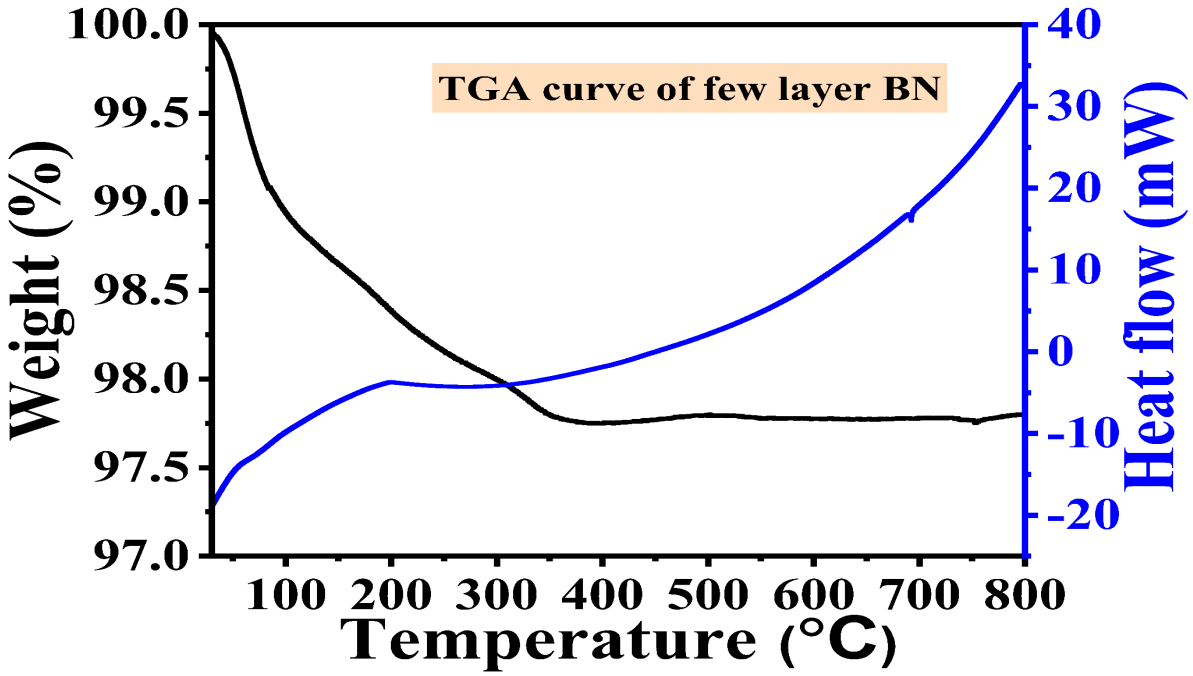

Figure S5: Thermogravimetry plot for functionalized h-BN

6. FTIR spectra

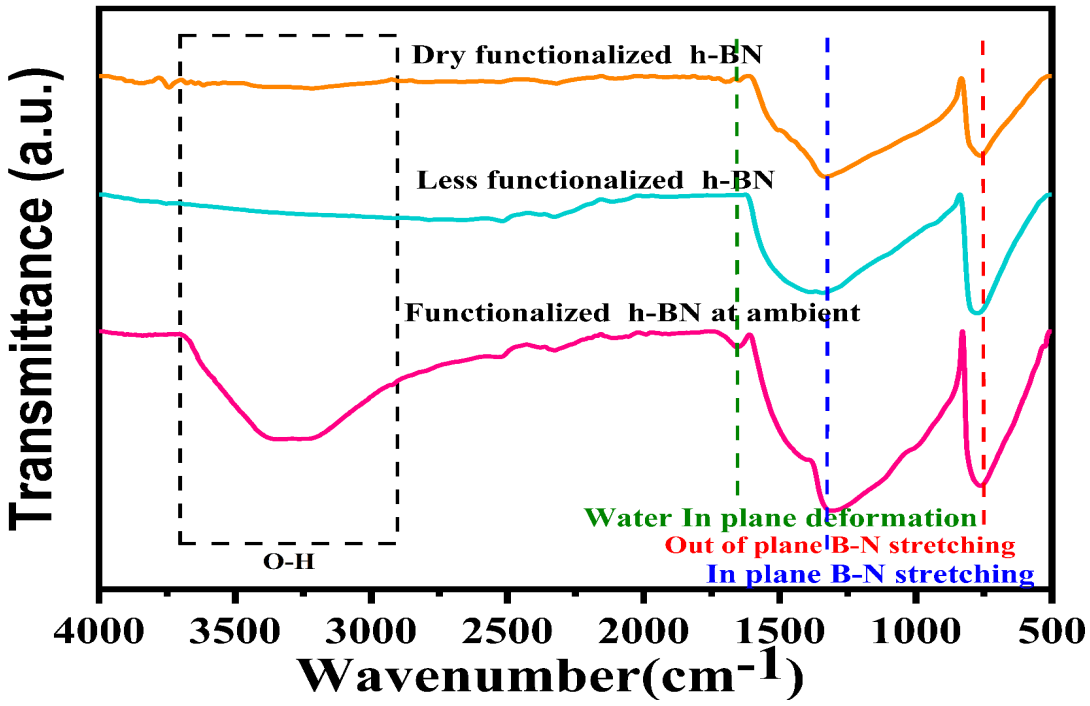

Figure S6: FTIR spectra of functionalized h-BN under dry and ambient RH conditions. Also shown for comparison is the FTIR spectrum of less functionalized h-BN under ambient RH conditions.

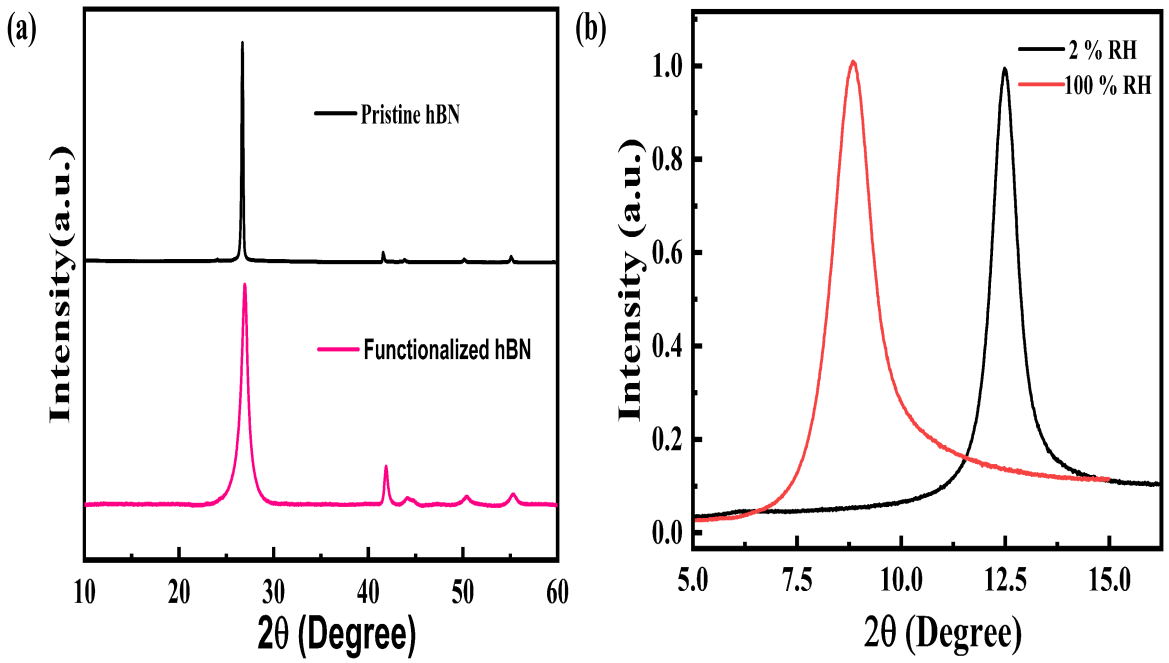

Figure S7: (a)XRD plots at ambient humidity for pristine and functionalized h-BN (b) Graphene oxide (GO): XRD peak shift at 2% and 100% RH.

8. Nyquist plots for temperature-dependent impedance data

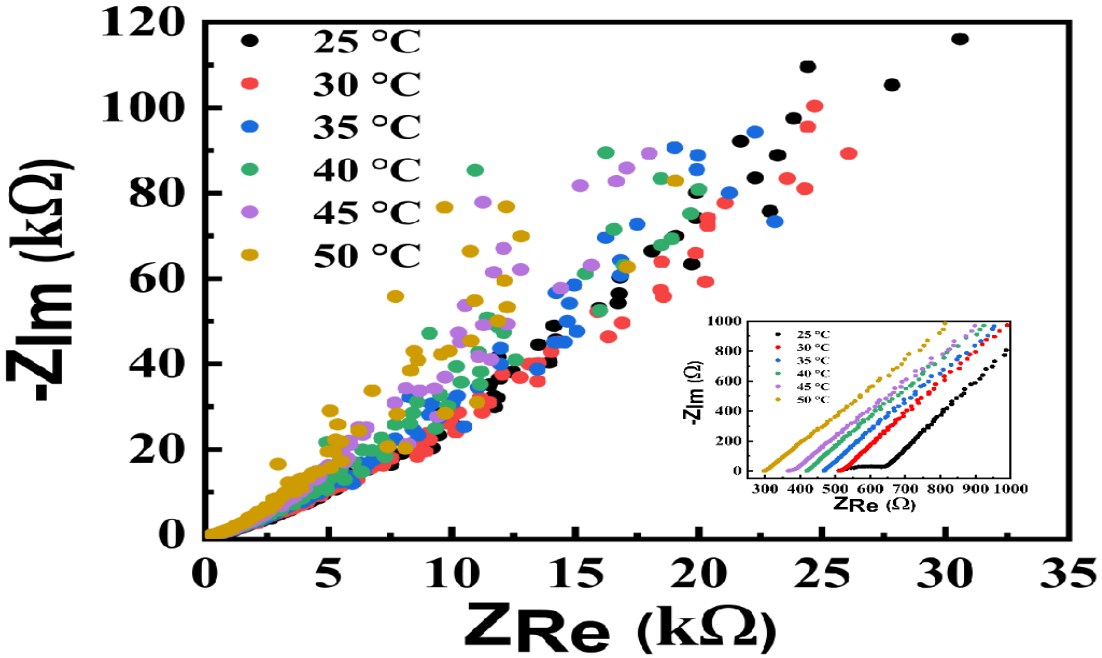

Figure S8: Nyquist plots for h-BN membrane over the temperature interval 25 – 50° C. [Inset: Nyquist plot in the high frequency interval]

9. Percolative proton pathways

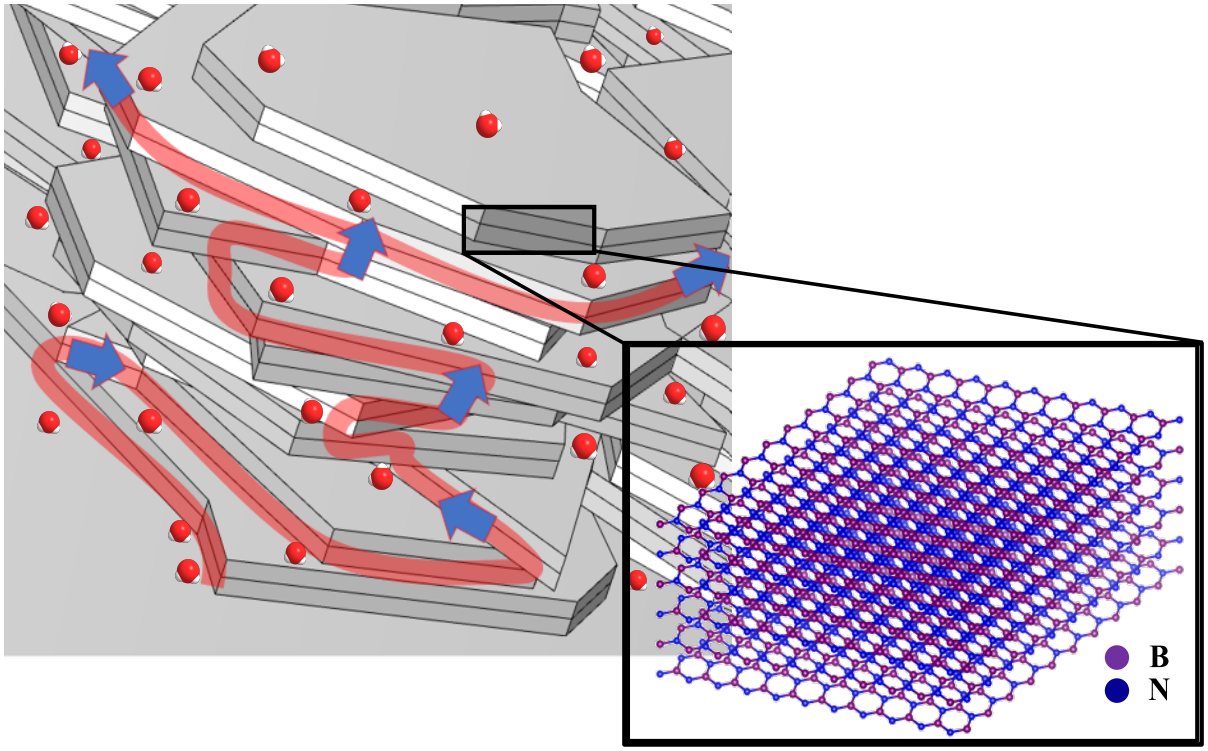

Figure S9: Schematic illustration of the proton percolative pathways along the edges of h-BN crystallites.

10. Roughness profile

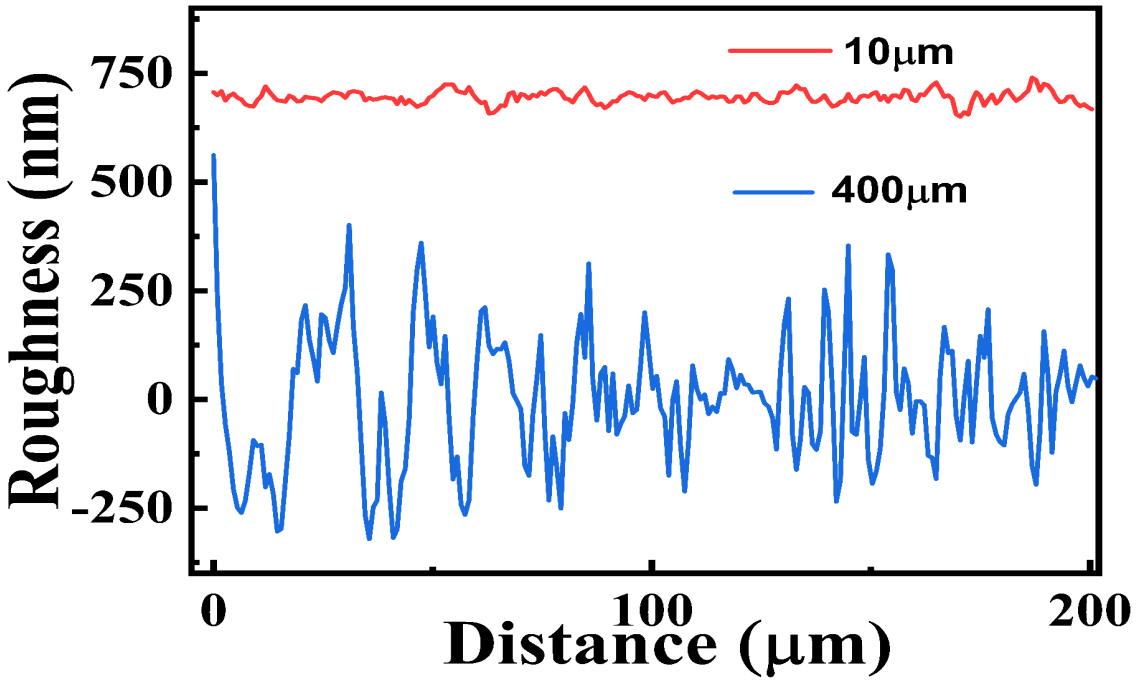

Figure S10: Roughness profile for 10 μm and 400 μm membrane.

The roughness profile of membranes extracted from optical profilometer data after baseline subtraction associated with gradual bending of the membrane. The average roughness of 400 μm membrane is 148.2 nm which is 10 times greater than 10 μm membrane.