Supplemental Methods and Figures

Analysis of hydraulic permeability in the wet/wet configuration

From a simple mass balance analysis the governing equation for the water volume transfer in the wet/wet format can be derived as

$$\epsilon = -\frac{ln\frac{V_{cup}(t)-V_{cup,EQ}}{V_{cup,o}-V_{cup,EQ}}}{(t)\rho g[1+\frac{A_{cup}}{A_a}]}$$

where $\boldsymbol{\epsilon}$ is the hydraulic permeability of the membrane, $\boldsymbol{\rho}$ is the density of water, $\boldsymbol{g}$ is the centrifugal acceleration calculated for the membrane position in the centrifuge, $\boldsymbol{V_{cup,o}}$ is the initial volume of water in the interior 'cup' obtained prior to centrifugation, $\boldsymbol{V_{cup}(t)}$ is the volume of water in the interior cup at a later time $\boldsymbol{t}$, and $\boldsymbol{V_{cup,EQ}}$ is the equilibrium volume of water in the cup obtained from extended spins or from control runs using intentionally broken membranes. The cross sectional area of the cup, $\boldsymbol{A_{cup}}$, and the annular cross section between the cup and the outer tube area $\boldsymbol{A_a}$ were calculated based on the dimensions of the two devices.

For cases such as pnc-Si where the active membrane material does not span the entire cross section interior cup, the formula above must be multiplied by the additional factor of $\boldsymbol{A_{cup}/A_m}$, where $\boldsymbol{A_m}$ is the active membrane area. The active membrane area of chips used was estimated by light microscopy and a typical value was 0.0032 cm^2 . The cup area and annulus areas were 0.394 cm^2 and 0.925 cm^2 respectively. Note that the use of active area in hydraulic permeability calculations rather than total chip area is appropriate to determine the intrinsic behavior of the nanomaterial and avoid device-dependent values. This was also the approach for example, taken by *Holt et al. Science,* 2006 (*1*) to calculate the hydraulic permeability of CNT membranes created as small active areas on otherwise impermeable silicon chips. Expanding the amount of active area on pnc-Si chips is an engineering challenge that is being met with steady improvements in etching, patterning, and deposition processes. Figure S3d shows the results of some of these improvements have led to new chip designs that have 10 times more active area than the device shown in Figure 1c.

The Dagan equation (*2*) for flow through short pores used to predict flow through pnc-Si is given by:

$$Q_{liquid} = \frac{r^3 \Delta P}{\mu[3 + \frac{8}{\pi}(\frac{l}{r})]}$$

where $\boldsymbol{r}$ is the average pore radius, $\boldsymbol{\Delta P}$ is the pressure drop across the membrane, $\boldsymbol{l}$ is the membrane thickness and $\boldsymbol{\mu}$ is the solution viscosity. This equation becomes the familiar Hagen-Poiseuille equation in the limit that $\boldsymbol{l/r}$ becomes large.

Estimate of uncertainty in predicted hydraulic permeability

Predictions of hydraulic permeability through pnc-Si membranes derived some uncertainty from the use of pore distributions from TEM images of actual membranes in calculations. There were two main sources of error in these calculations. First, the identification of pores in the pore image processor program varied slightly between users. Second, images only capture a small section of the membrane and so the histograms for individual fields on the same membrane varied to some degree. Calling these uncertainties σ_{user} and $\sigma_{membrane}$ respectively, and assuming they are independent contributors to the overall uncertainty, σ_{all}, we can write $(\sigma_{all})^2 = (\sigma_{user})^2 + (\sigma_{membrane})^2$. We developed systematic tests to estimate each of these uncertainties and determine σ_{all} for Figure 2d. σ_{user} was found by having four different qualified users operate the pore processing program and evaluate the pore characteristics of the five types of membranes presented in Figure 2d. $\sigma_{membrane}$ was found by having one user evaluate a set of TEM images from different fields of view for each of the five membrane types.

Pore Image Processing and UV/Ozone Treatments

See legends of supplemental figures.

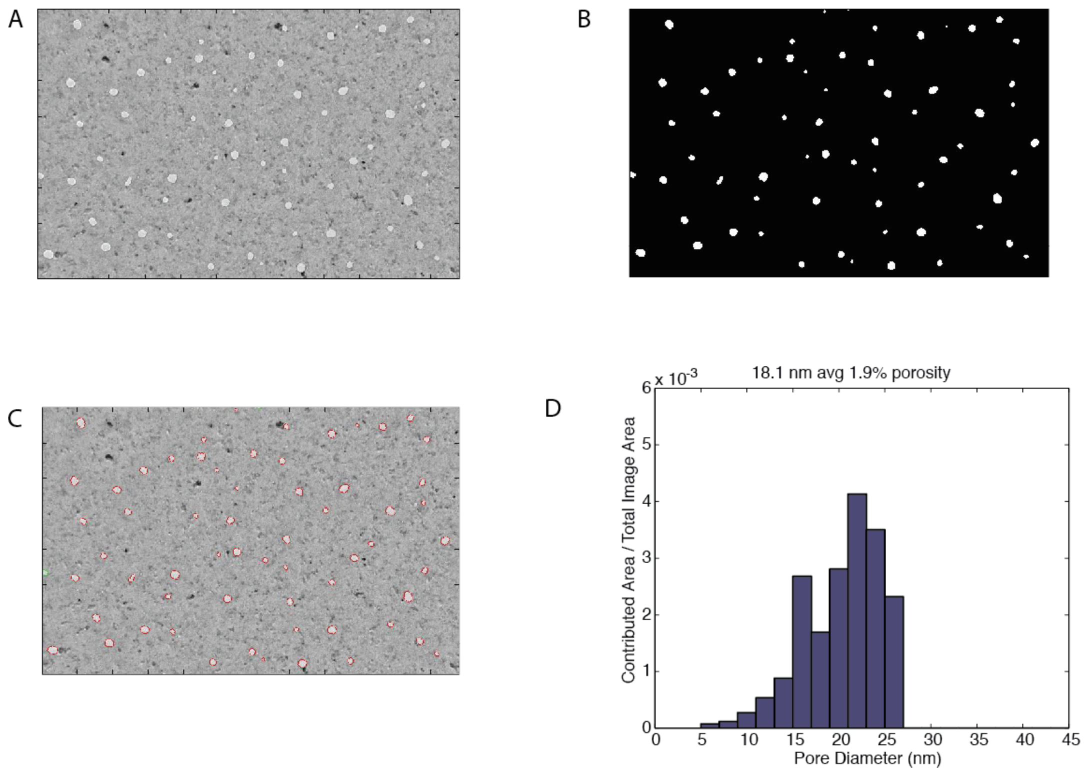

Figure S1: Pore processing of TEM images. We use a custom image processing program written in MATLAB to analyze pore characteristics of pnc-Si membrane TEM images. (A) We first perform background correction by subtracting the average value from a local region of defined size from each pixel. We use a local region that is bigger than the size of the pores, but smaller compared to any light variations in the background. (B) The image is transformed into a black and white image using a threshold value. The threshold value is chosen so that most pore pixels have an intensity value higher than the threshold and background pixels mostly fall below. Values higher than the threshold are white in the new binary image, and those below the threshold are black. Combinations of binary operations are then used to remove artifacts and better define pores in the image before finding the final statistics. An erode operation is used to shrink white areas within neighboring black areas, and a dilate operation is used to grow white areas. Erosions followed by dilations can remove noise and small artifacts from the image while retaining pores. Dilations followed by erosions can be used to fill in pores with inner regions below the threshold. The selection of these operations will vary depending on the image, and leads to user-to-user variability which we have quantified. (C) Pore edges are identified and pore statistics, including diameters, pore areas, and porosity, are found. (D) Pore distributions are typically found by plotting the contributed area for each pore diameter. Average diameter and porosity are indicated above the plot. The pore image processing routines are freely available at http://nanomembranes.org/resources/software/

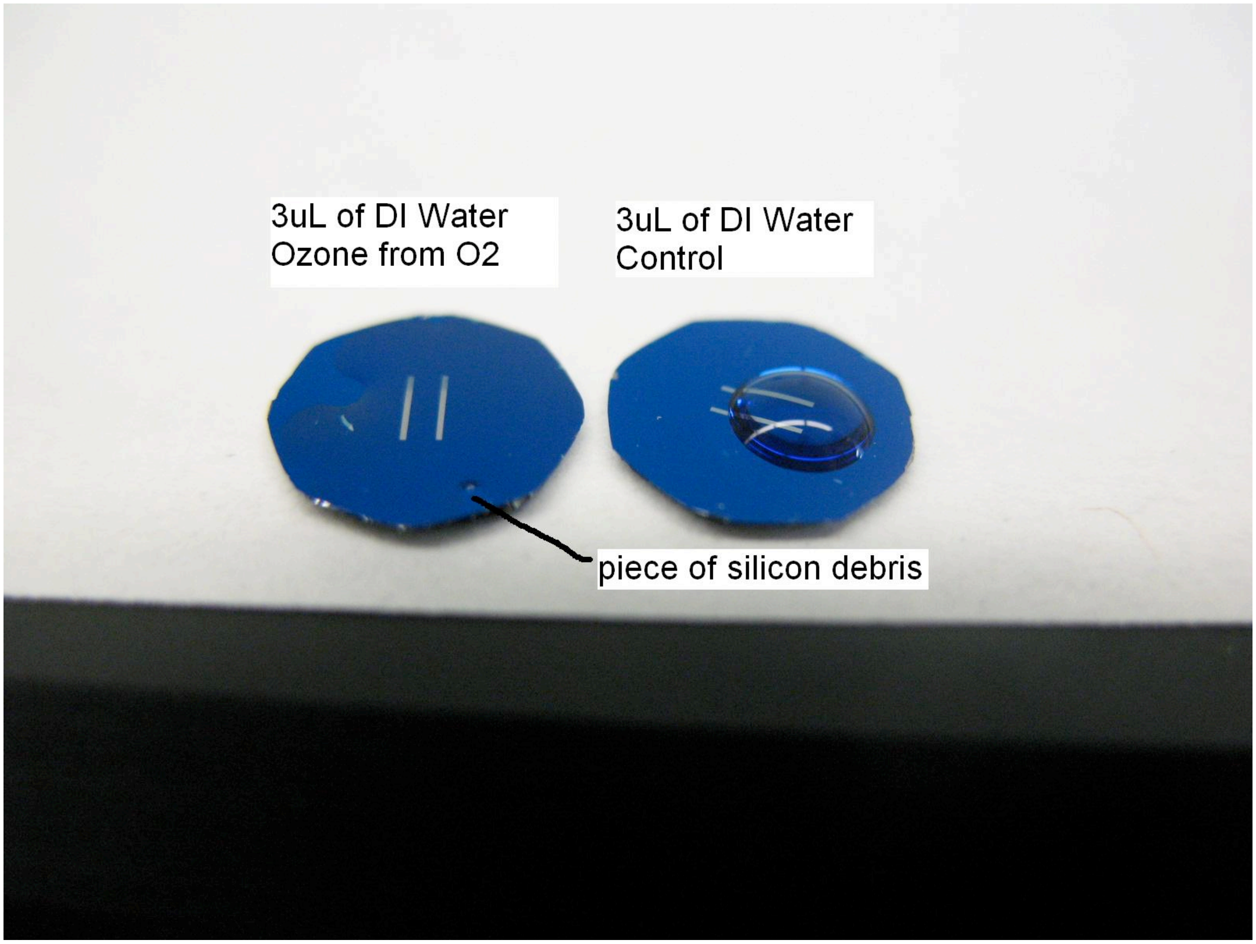

Figure S2: UV Ozone Treatment increases membrane hydrophilicity. In the effort to develop wet/dry hydraulic permeability we treated pnc-Si membranes with Novascan PSD-UVT UV/ozone system. Membranes were exposed to three minutes of oxygen at 50°C followed by three minutes of UV treatment at 50°C. The membranes were undisturbed in the chamber for 15 more minutes to allow the ozone react with the samples. Both figures show 3 uL droplets of DI water. The fluid is so flat on the treated sample that it is hard to discern, except for the disturbance of the water layer caused by a small piece of debris.

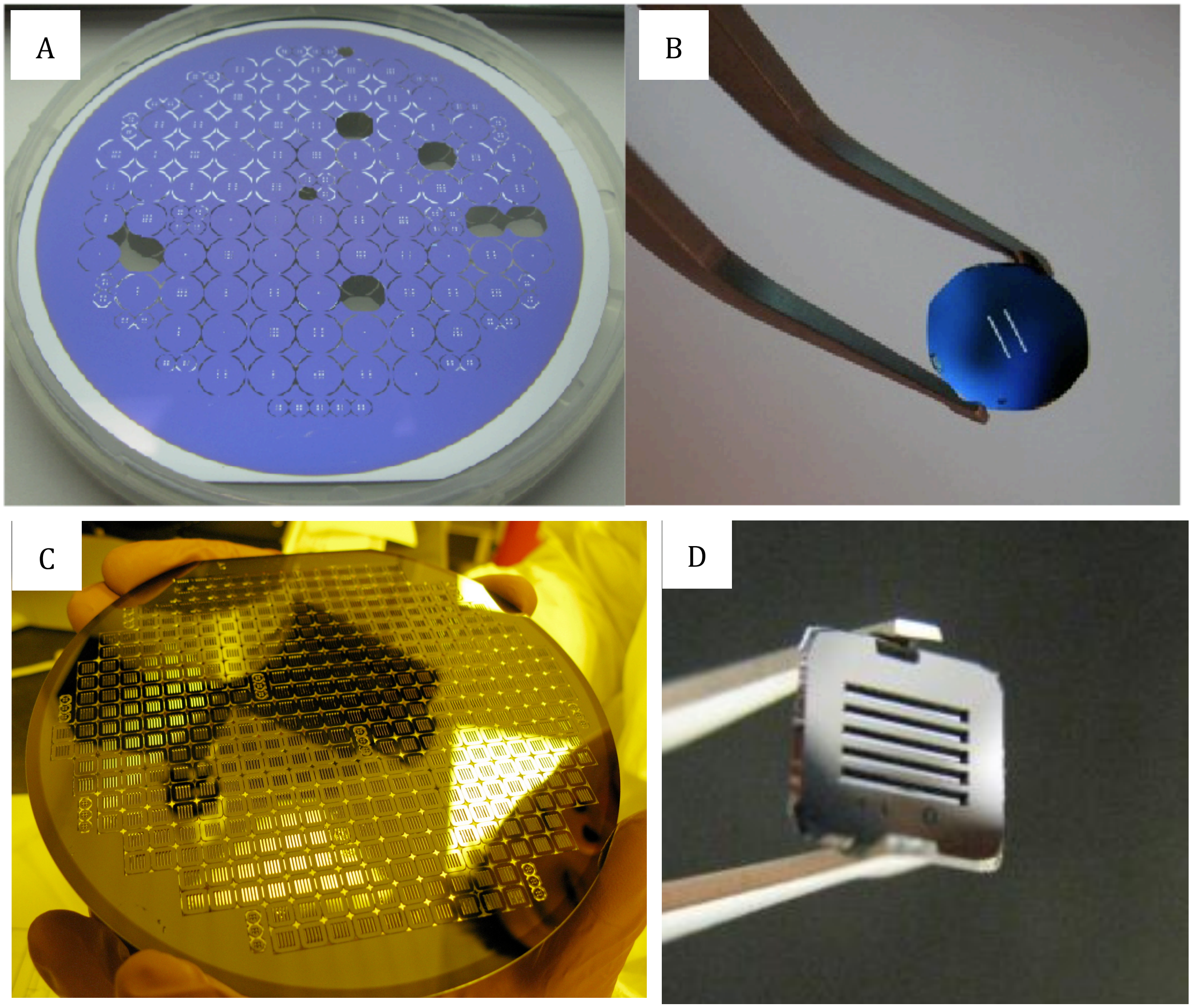

Figure S3: Pnc-Si membrane fabrication is highly scalable. A) Many pnc-Si membrane chips can be made per individual wafer, and this particular 4” wafer, left, has 84 chips formatted for centrifuge tubes and 46 smaller TEM grid chips. B) Pnc-Si chips are detached from the wafer by applying light pressure to the edges of the chips. C) Pnc-Si production has recently been scaled from 4” wafers to 6” and from round to square formats allowing more than 400 membrane devices with high active area to be produced with only marginally more cost compared to 4” wafers. D) Improved chip design now allows 10% of total chip area to be covered with active membrane, approximately 10x more membrane area than the chip shown in C).

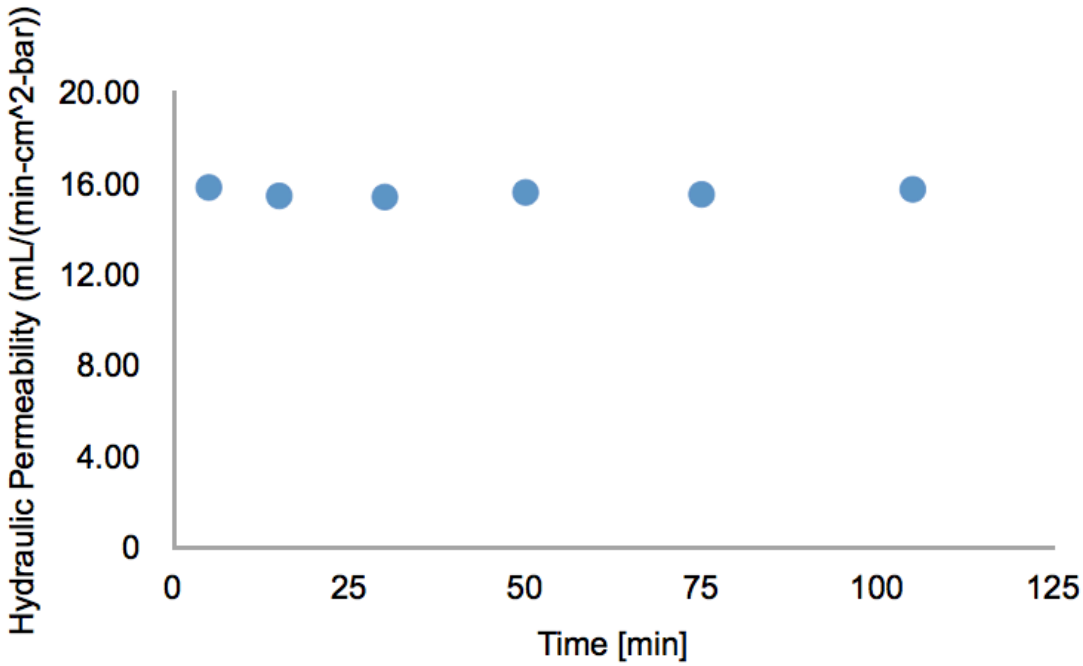

Figure S2: Hydraulic permeability of pnc-Si is time-independent. Distilled and deionized water was forced through a pnc-Si membrane in the pressure cell for over two hours and passed volumes were collected at the times shown to calculate an 'instantaneous' hydraulic permeability.

Table S1. Stock nanoparticle sizes as determined by DLS and TEM.

Manufacturer's Size (nm)	TEM Size (nm)	DLS Size (nm)
9.2 +- 0.9	7.8 +- 0.9 (STD)	9.7 +- 2.4 (STD)
15.2 +- 1.5	12.7 +- 0.9 (STD)	13.3 +- 3.4 (STD)

Citations

1. J. K. Holt *et al.*, *Science* **312**, 1034 (May 19, 2006).
2. Z. Dagan, S. Weinbaum, R. Pfeffer, *J. Fluid Mech* **115**, 505 (1982).