Supplementary Information

1. CHEMICALS

The chemicals used in the preparation of hydrogels are listed below:

Acrylamide (AAm)
N,N' - Methylenebis(acrylamide) (MBA)
Ammonium Persulfate (APS)
N,N,N',N' - teramethylethane - 1,2 - dimine (TEMED)
N,N - Dimethylacrylamide (DMA)
N-Isopropylacrylamide (NIPAAm)
All chemicals used in this paper are purchasd from Sigma-Aldrich Co.

2. HYDROGEL PREPARATION

All hydrogels were prepared from aqueous stock solutions of the following chemicals: N,N'-methylene(bis)acrylamide (MBA), N,N-Dimethylacrylamide (DMA), ammonium persulfate (APS), and tetramethylethylenediamine (TEMED) at concentrations of 0.108 g/10.0 mL, 2.60 mL/10.0 mL, 0.0800 g/10.0 mL and 0.250 mL/10.0 mL, respectively. The base acrylamide (AAm) monomer was used in its pure powder form. By mixing different amounts of these chemicals, polymers were spontaneously synthesized. During this process, APS served as an initiator, TEMED as an accelerator, and MBA as a crosslinker.

In all hydrogels, we started with 0.500 g of AAm monomer, 1.00 mL of TEMED solution and 1.00 mL of APS solution. Amounts of MBA solution waws varied to achieve the target crosslinker ratio (MBA/AAm, mol/mol) ranging from 0.500 % to 7.00 %.

For hydrogels with DMA, 10.0 % (DMA/AAm mol/mol) was added. Then, the solution was vortex mixed for approximately one minute and subsequently rested at room temperature (24.0 °C) for 24 hours. For samples that were hydrolyzed, we immersed the samples in 1.00 mol L^{-1} sodium hydroxide for 30 minutes before. For PNI-co-AAm hydrogels, 10.0 % mol (of AAm) of NIPAAm was added before adding crosslinker, accelerator and initiator. The ratio of Total Water/AAm was fixed to be 1000 (mL/mol).

Finally, the samples were rinsed everyday and immersed in DI water for one week to remove unreacted chemicals and equilibrate them to the wet state.
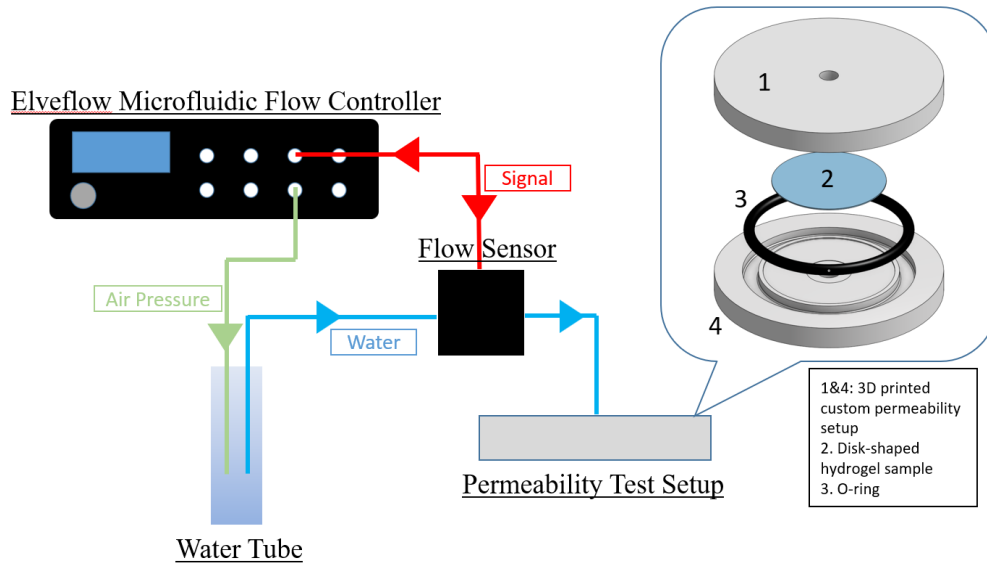
Fig. S1. A scheme of the custom-built permeability tester. The custom permeability setup was designed and printed in our lab. With the setup, the gel thickness, L, and cross-sectional area, A, (Eq. 4) can be fixed at certain values. To ensure the sealing of the setup, an o-ring was applied between top and bottom parts. After the sample was inserted in the setup, two clamps were used to keep the setup being tight through the testing. ΔP (Eq. 4) was controlled by the Elveflow Microfluidic Flow Controller and provided pressure in the water tube. Water in the water tube was pushed out and ran through the flow sensor, where instant reading of the volumetric flow rate, Q (Eq. 4) could be sent back to the controller for an accurate adjustment. Water flow with targeting pressure flew through the sample in the permeability setup and a real-time Q was read by the sensor.
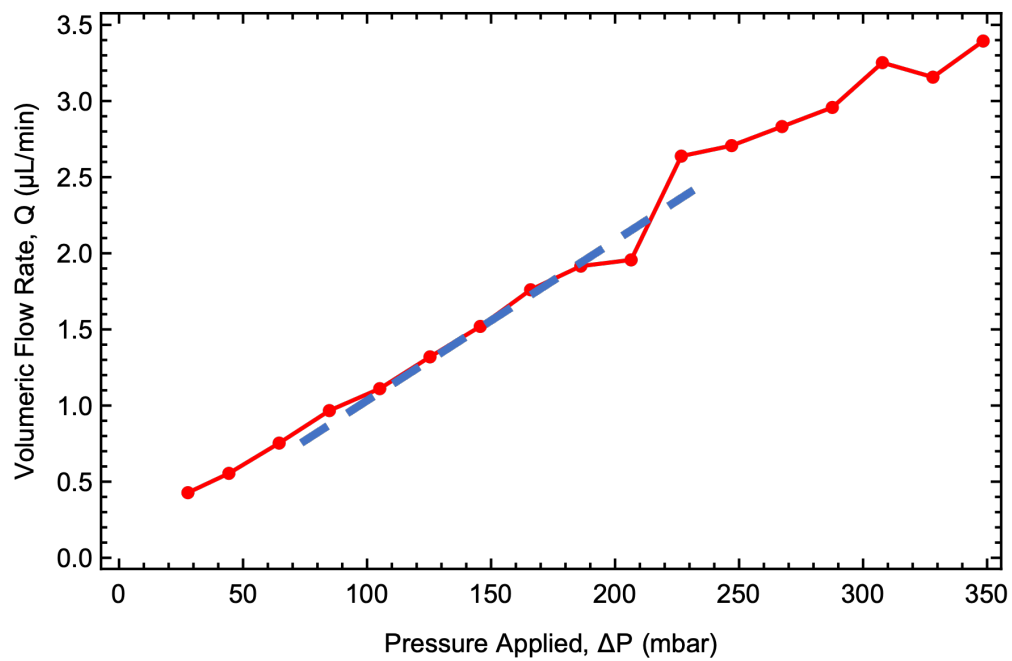
Fig. S2. K of the tested sample was 18.4 kPa (184 mbar). We varied pressure and found that the slope of $Q/\Delta P$ was nearly linear when $\Delta P/K$ was in the range of 0.5–1 (lower pressures introduce measurement uncertainties, within an interval of $\pm 2.00\%$); therefore, we set ΔP such that $\Delta P/K = 0.7$ for all our permeability tests of all samples.

1. Load cell
2. Stainless steel ball bearing
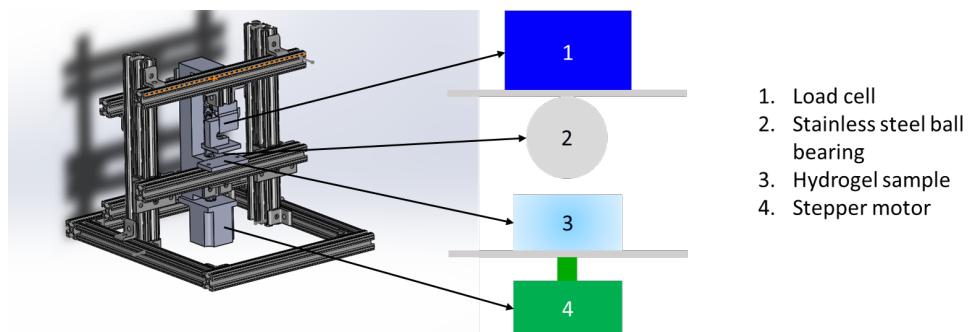3. Hydrogel sample
4. Stepper motor

Fig. S3. We used a custom-built indentation tester to perform these measurements. Samples were prepared in a cylindrical shape and oriented such that a flat surface was indented. All tests are completed within 15 minutes to ensure minimal weight loss from de-swelling to the ambient environment. To confirm this minimal weight loss, we ensured that the weights of the samples before and after indentation tests were less than 1.00 %. Displacement speeds ranged from $5.00\,\mathrm{mm\,min^{-1}}$ to $10.0\,\mathrm{mm\,min^{-1}}$; slower or faster speeds did not affect the force-displacement curves, indicating the sample behaved quasi-statically and quasi-elastically, away from dynamic drainage and viscoelastic effects.
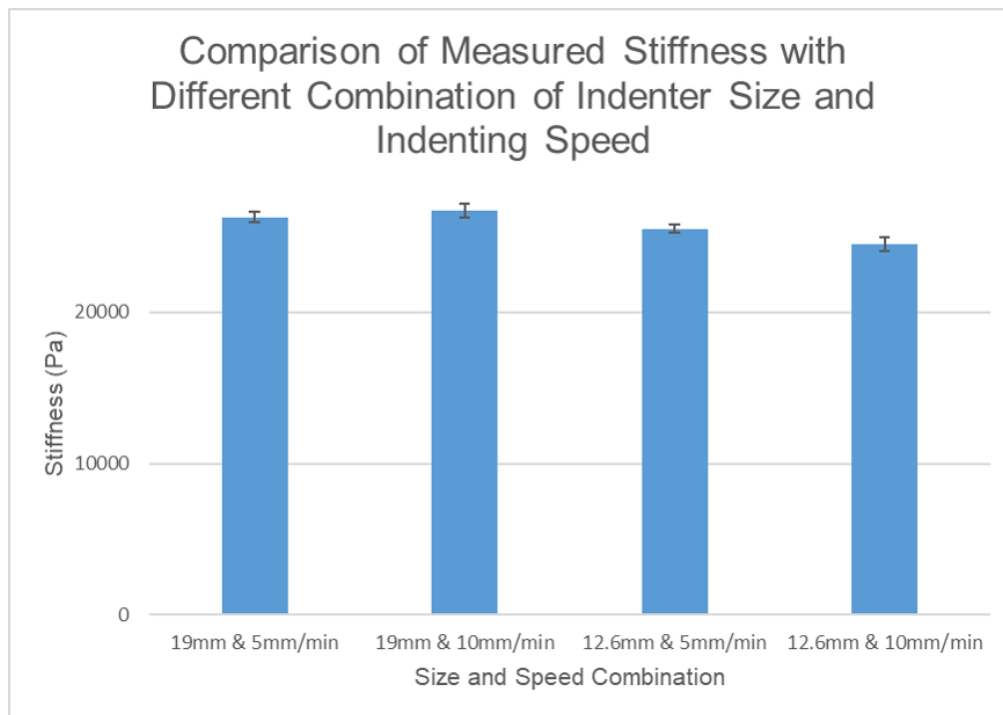
Fig. S4. Pure PAAm with 3.00 % crosslinker ratio sample is shown here as an example. Ball bearing size is set to 19 mm or 12.6 mm (0.75 in or 0.5 in), compression speed is set to 5 mm/min or 10 mm/min. The invariance in stiffness results with indenter size and speed show that our testing procedure is reliable and can be assumed quasistatic.
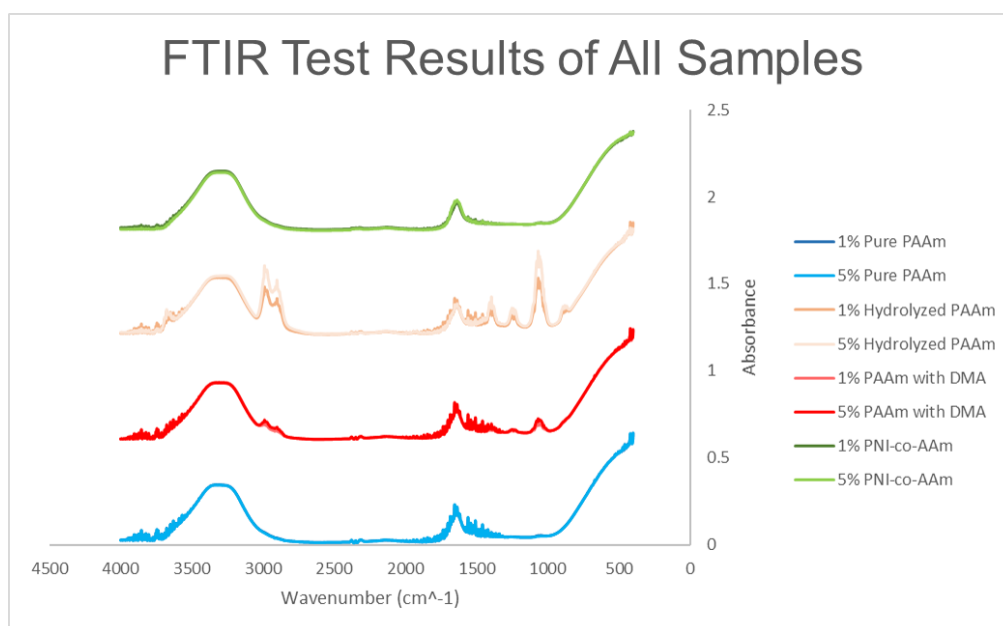
Fig. S5. FTIR test results for samples from four hydrogel families. The results indicate that samples from the same hydrogel family only differ in polymer volume fraction. Plots have been shifted vertically by family for clarity.
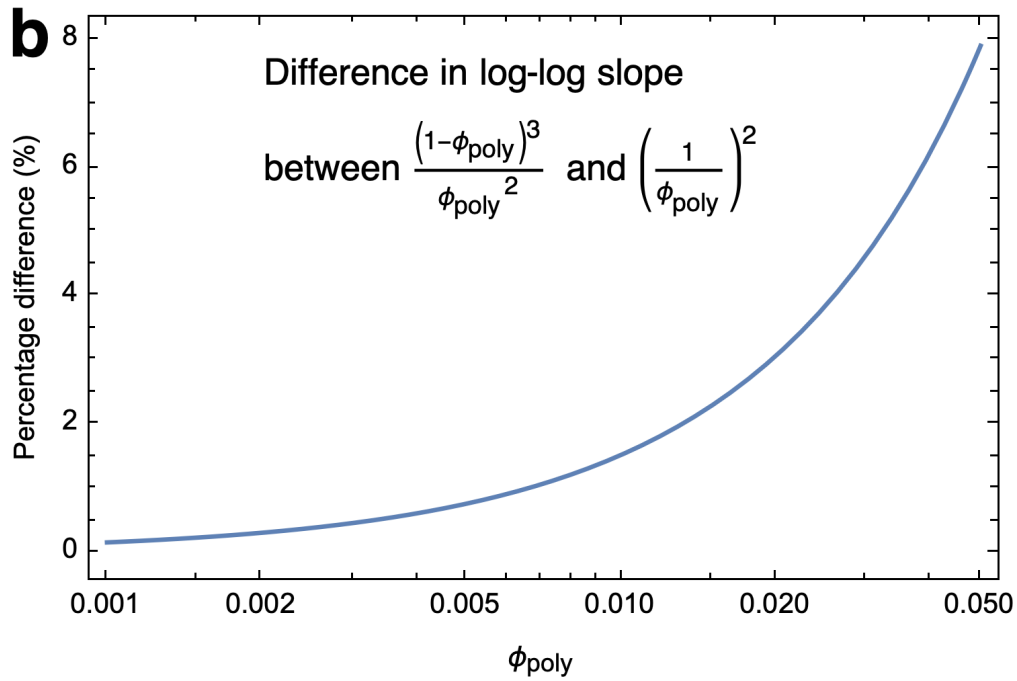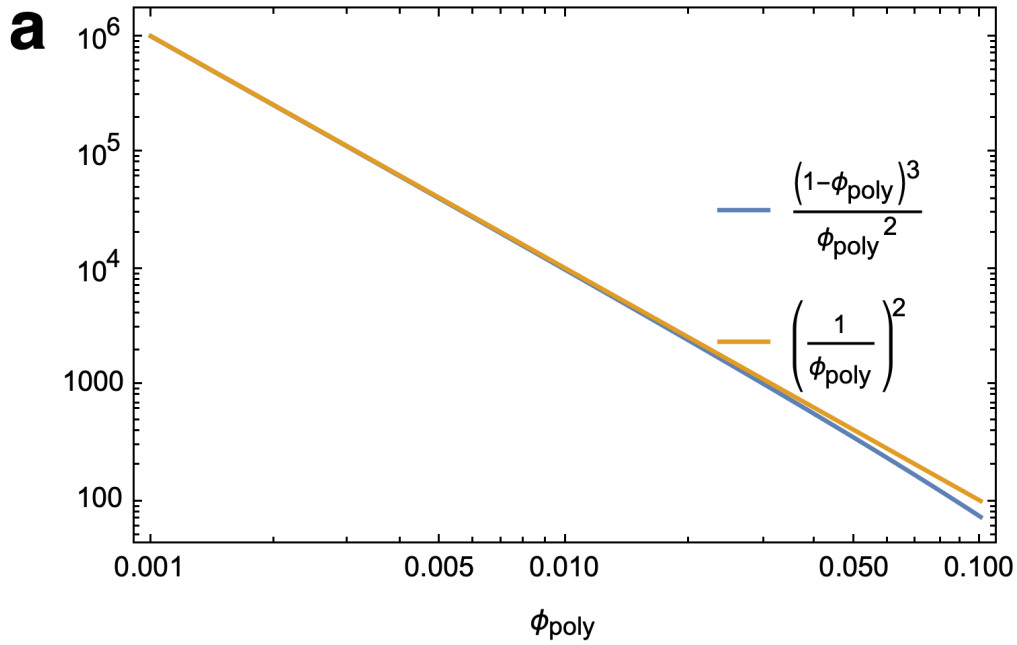
Fig. S6. (a) For small ϕ_{poly} ($\phi_{\text{poly}} < 0.05$) the permeability can be approximated as $\kappa \propto \phi_{\text{poly}}^{-2}$ with minimal error. (b) The calculation shows an error within 8.00 % when ϕ_{poly} is equal or lower than 0.0500
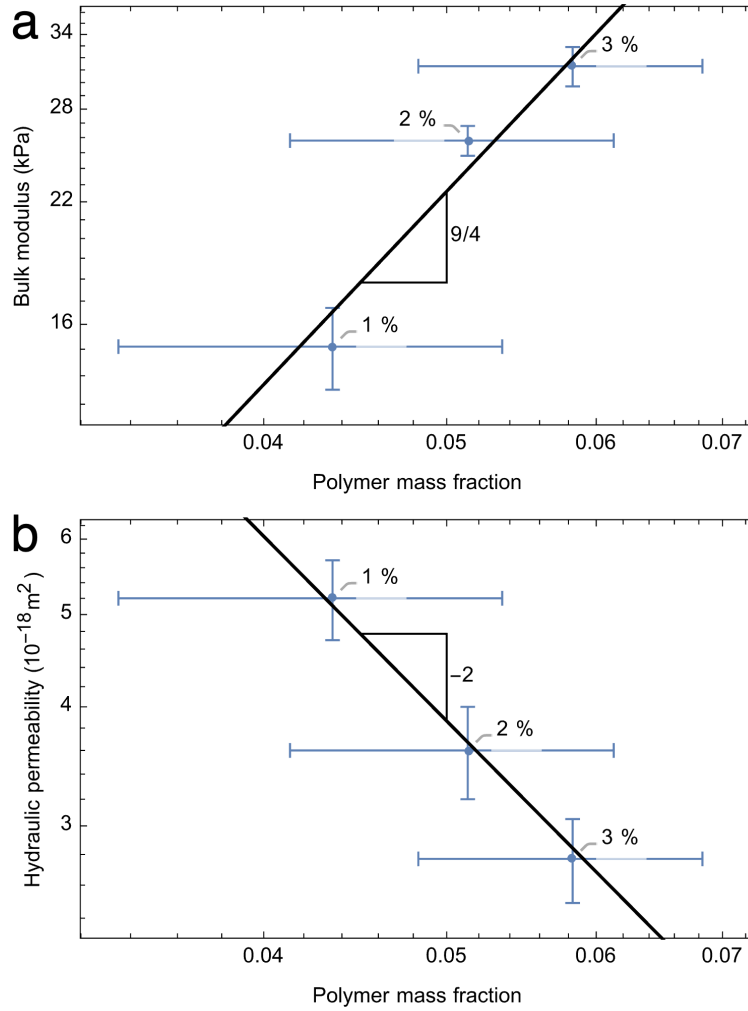
Fig. S7. The stiffness and permeability of pure PAAm hydrogel samples with crosslinker ratios of 1, 2, and 3 % are plotted against the polymer mass fractions. The polymer mass fraction is proportional to the polymer volume fraction; thus, the stiffness should scale with the mass fraction by a 9/4 power according to Eq. 2. (a) A log-log plot of bulk modulus versus polymer mass fraction confirms this 9/4 scaling. Likewise, the permeability should scale with the mass fraction by a −2 power according to Eq. 7. (b) A log-log plot of hydraulic permeability versus polymer mass fraction confirms this −2 scaling. The polymer mass fractions were obtained by measuring the weight of samples dried for over one week under dry air flow and dividing by their weight after equilibrating in pure water for over one week.

Sample	Permeability, κ (10^{-18}m^2)	Modulus, K (kPa)
Pure 0.5%	9.7 ± 1.0	7.0 ± 0.6
Pure 1%	5.2 ± 0.5	15.1 ± 1.6
Pure 2%	3.6 ± 0.4	25.8 ± 1.0
Pure 3%	2.8 ± 0.3	31.3 ± 1.6
Pure 5%	2.9 ± 0.3	31.0 ± 0.9
Hydro 1%	23.1 ± 2.4	7.6 ± 0.7
Hydro 2%	13.9 ± 1.4	14.4 ± 1.7
Hydro 3%	9.6 ± 1.0	18.4 ± 0.7
Hydro 5%	9.5 ± 1.0	20.9 ± 1.2
Hydro 6%	6.1 ± 0.6	35.6 ± 0.9
Hydro 7%	3.1 ± 0.3	85.7 ± 2.4
DMA 0.5%	12.6 ± 1.3	9.3 ± 0.4
DMA 1%	8.3 ± 0.8	14.6 ± 0.3
DMA 2%	5.6 ± 0.6	25.5 ± 0.5
DMA 3%	5.0 ± 0.5	31.8 ± 0.4
DMA 5%	5.4 ± 0.6	27.2 ± 0.5
NIP 0.5%	11.9 ± 1.2	8.0 ± 0.7
NIP 1%	6.0 ± 0.6	16.7 ± 1.0
NIP 2%	3.4 ± 0.4	32.9 ± 0.6
NIP 3%	2.9 ± 0.3	42.6 ± 0.7
NIP 5%	2.8 ± 0.3	43.3 ± 0.5

Table S1. Stiffness, K, and permeability, κ, of all samples from four hydrogel families with varying crosslinker ratio.

Hydrogel Family	Prefactor ($10^{-17}\text{m}^2*\text{kPa}^{8/9}$)
Pure	5.9 ± 0.3
DMA	9.8 ± 0.5
Hydro	14.5 ± 0.7
NIP	7.8 ± 0.4

Table S2. Prefactors depend on the monomer size, solvent interaction and sphericity, thus, different hydrogel families have different values of prefactors.