Electronic Supplementary Information: Poroelastic shape relaxation of hydrogel particles

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Figure S1 | Determination of characteristic indentation depth. Image corresponding to Fig. 2B showing the (a) inscribed circle of radius R_{in} and (b) circumscribed circle of radius R_{out} , from which the characteristic indentation depth $h \equiv R_{out} - R_{in}$ is determined.



Figure S2 | Minimal influence of drying. To assess the influence of drying, we leave a hydrogel bead swollen in the aqueous NH₄SCN solution on a flat glass surface for 10 h. As shown in the images above, the shrinkage of the swollen bead is minimal — the diameter decreases by only $\sim 8\%$. Moreover, considering that the poroelastic time scale over which the shape relaxation dynamics occur is much shorter, over a time scale ranging from ~ 15 min to ~ 1.5 h, we expect that the influence of solvent evaporation will be minimal.



(a)

 $-\left(\Psi-\Psi_i
ight)/(\Psi_f-\Psi_i)$

 $10^{-2} \underbrace{10^{-2}}_{0} \underbrace{10^{-2}}_{10^{-2}} \underbrace{10^{-1}}_{10^{-1}} \underbrace{10^{0}}_{10^{0}}$ Normalized Time t/τ Normalized Time t/τ

Figure S3 | Measurements of normalized circularity. The measurements of circularity $\Psi(t)$ are fit using the stretched exponential function $\Psi(t) = \Psi_f - (\Psi_f - \Psi_i) e^{-(t/\tau)^{\beta}}$, where Ψ_f, Ψ_i, β , and τ are fitting parameters. To further demonstrate the quality of the fitting, the measurements shown in Fig. 3b are re-plotted in two different ways. (a) We plot $1 - (\Psi(t) - \Psi_i) / (\Psi_f - \Psi_i)$ versus t/τ with a logarithmic horizontal axis; in this representation, we observe that the data show a slight downward curve, reflecting that the exponent $\beta > 1$. This representation also shows that the fitting works well up to $t \sim 3\tau$, at which point the noise in the measurement dominates. (b) We plot $-\ln[1 - (\Psi(t) - \Psi_i) / (\Psi_f - \Psi_i)]$ versus t/τ on log-log axes; in this representation, we observe that the exponent $\beta > 1$. This representation also shows that the exponent $\beta > 1$. This representation also shows that the fitting works well up to $t \sim 3\tau$, at which point the noise in the measurement dominates. (b) We plot $-\ln[1 - (\Psi(t) - \Psi_i) / (\Psi_f - \Psi_i)]$ versus t/τ on log-log axes; in this representation, we observe that the data fall on a line whose slope is greater than one, again reflecting that the exponent $\beta > 1$. This representation also shows that the fitting works well except at the shortest times $t < 0.1\tau$ and longest times $t > 3\tau$, which are an order of magnitude smaller and three times larger than the poroelastic time scale of interest τ , respectively.

Determining poroelastic relaxation dynamics by tracking the position of individual points on the hydrogel surface. In a given experiment, we track three different points at the hydrogel surface; we note that an alternative method could be to track the distance between pairs of adjacent points on the hydrogel surface. In particular, for each point *i*, we binarize our images of the hydrogel using a given threshold value and track the position R_i of the point over time *t*. In some cases, slight drying or movement of the hydrogel introduces a linear offset to these measurements, which we subtract from the data; we then fit $R_i(t)$ with an exponential function as shown in Fig. 3a, which yields $\tau_i \pm \delta \tau_i$, where the uncertainty $\delta \tau_i$ reflects the uncertainty in the least-squares fitting. Taking the mean of the three τ_i thus obtained for a given hydrogel yields the relaxation time τ (circles in Fig. 4) while propagating the uncertainties $\delta \tau_i$ yields the overall uncertainty (vertical error bars on the circles in Fig. 4).

Determining poroelastic relaxation dynamics by tracking the overall circularity of the hydrogel. In a given experiment, we binarize our images of the hydrogel using a given threshold value and track the overall circularity Ψ over time t. We then fit $\Psi(t)$ with the stretched exponential function $\Psi(t) = \Psi_f - (\Psi_f - \Psi_i) e^{-(t/\tau)^\beta}$, where Ψ_f , Ψ_i , β , and τ are fitting parameters; this function simply serves as a phenomenological fitting function that provides a good fit to the data as shown in Fig. 3b. For all of our measurements, the exponent of the stretched exponential $\beta = 1.5 \pm 0.8$, in close agreement with previous measurements of poroelastic relaxation (Refs. 63, 71-73) that reported $0.4 \le \beta \le 2.4$. Our fits yield $\Psi_f = 0.894 \pm 0.008$ and $\Psi_i = 0.85 \pm 0.03$. However, it is important to note that the absolute values of Ψ_i and Ψ_f depend on the resolution and magnification of the images used—a well known consequence of pixelization, as described in Ref. 70. To assess the uncertainty in τ , we re-fit the data obtained upon varying this threshold value by $\pm 5\%$, for which the binarized images still closely approximate the shape of the imaged hydrogel. The resulting standard deviation in the values of τ thus obtained yields the reported uncertainty in τ . The uncertainty associated with determining τ from the least-squares fitting procedure itself is two orders of magnitude smaller; we therefore neglect it from our analysis.