Electronic Supplementary Information

A highly sensitive, low-cost, wearable pressure sensor based on conductive hydrogel spheres [†]

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Fig. S1 Alginate-based hydrogels with different geometries. a) An alginate hydrogel film, b) a hydrogel thread made of cylinders 3 ± 0.2 mm in diameter, c) and d) hydrogel cylinders of different lengths cut from the hydrogel thread. Transparent samples are alginate hydrogels, while black samples are SWCNT/alginate hydrogels with a SWCNT concentration of 0.5 wt. %.



Fig. S2 I-V curves of the as-prepared pressure sensor with pure alginate sphere and SWCNT (0.5 wt. %)/alginate sphere (sensing sphere diameter is 1 mm).



Fig. S3 Total resistance variation as a function of external loading with different alginate cylinder lengths (1 mm, 2 mm, 3 mm). Gold-coated wafer served as the electrodes; the cylinder sample had a diameter of 3 mm.

S1: Analytical modeling of the device's piezoresistive response.

We propose elementary modeling elements of the device's piezoresistive response to explain the decrease in device sensitivity with increasing pressure with a power exponent close to (2/3).

To keep the modeling as simple as possible, we move sequentially through a set of assumptions that are validated at each step. We also keep the demonstration as general as possible, avoiding the details of each term, but rather trying to extract the trend of the change in resistance with pressure.



Fig. S4 A schematic of the system. p is the external applied pressure and P is the contact force at the sphere/electrode interface.

First, considering the very low load applied to the device under normal operating conditions, we can assume reversibility of the system and, more precisely, linear elasticity of all material behaviors. As a consequence, we assume the contact force, P (unit: N), at the sphere/electrode interface to be proportional to the external applied pressure, p (unit: N/m^2), such that (Fig. S4):

$$P = k_e * p \tag{S1}$$

where k_e (unit: m^2) is a proportionality factor that could be obtained by solving the full mechanical-structural problem over the whole device. Although it depends on many parameters (all the geometrical and material parameters of the different system components), we expect that it mainly depends on the deflexion of the electrode membrane. As a consequence, it would mainly depend on the geometrical parameters and on the boundary condition of the electrode membrane. The precise determination of k_e is not needed.

The actual total resistance of the system $(^{R_{T}})$ results from the combination in series of the actual resistances of the sensing sphere $(^{R_{S}})$ and of the sphere/electrode contact $(^{R_{C}})$, such that:

$$R_T = R_S + 2R_C \tag{S2}$$

Modeling R_C: contact resistance as function of the loading.



Fig. S5 Decomposition of the contact resistance.

The contact resistance, R_c , results from the initial resistance of the contact (as mounted before any loading, R_c^0) and the resistance of the additional contact surface resulting from the loading (R_c^+) such that (Fig. S5):

$$R_{C} = \frac{1}{\frac{1}{R_{C}^{0}} + \frac{1}{R_{C}^{+}}} = \frac{R_{C}^{0}R_{C}^{+}}{R_{C}^{0} + R_{C}^{+}} \approx R_{C}^{0} \left(1 - \frac{R_{C}^{0}}{R_{C}^{+}}\right)$$
(S3)

 R_c^0 is a constant resistance that corresponds to the contact resistance at zero pressure. R_c^+ is inversely proportional to the new contact surface resulting from the force, P, at the sphere/electrode contact such that:

$$R_{\mathcal{L}}^{+} = \beta/A \tag{S4}$$

 β is a material parameter (unit: $\Omega .m^2$) that quantifies the specific resistivity of the newly created interface. The second approximation in Eq. S3 is valid as $(R_c^+ \gg R_c^0)$, which is obviously true.

In Eq. S4, A is the contact surface (unit: m^2). Considering linear elasticity, we can assume a Hertzien contact between an elastic sphere (the sensing sphere) and an infinite plane (the electrode). Then we have, using classical results from Hertz's theory:

$$A = \pi a^2 \text{ with } a = \frac{3}{4} \left(\frac{\Phi P}{E^*}\right)^{1/3}$$
(S5)

 E^* is the reduced Hertz modulus, defined as: $\frac{1}{E^*} = \frac{1}{E_1} + \frac{1}{E_2} = \frac{1}{E_1} (E_1, v_1)$ and (E_2, v_2) are the Young's modulus and the Poisson's ratios of the constitutive materials of the sensing sphere and the electrode, respectively.

Based on Eqs. S1, S4 and S5, we derive the following approximation for R_c^+ :

$$R_{C}^{+} = \frac{\beta 16}{\pi 9} \left(\frac{E^{*}}{k_{e}}\right)^{2/3} (\Phi p)^{-2/3} = \gamma . \Phi^{-2/3} . p^{-2/3}$$
(S6)

 γ is a reduced constant coefficient that we introduce for simplicity (unit: $\Omega . m^{-2/3} . N^{2/3}$).

Modeling R_{S} : resistance of the sensing sphere.

As we can see from Fig. S3, the volume piezoresistance of the sphere material is limited to only a small percentage so it can be neglected. We then assume that the resistance of the sphere itself is constant and independent of the loading. We note it as R_s^0 to emphasize that we consider it as non-loading dependent.

Estimation of the change in resistance.

Eq. S2 defines the actual resistance of the system. If R_T^0 is the initial total resistance of the system, then R_T^0 is defined as:

$$R_T^0 = R_S^0 + 2R_C^0$$
(S7)

Using Eqs. S2, S3, S6 and S7, the change in resistance of the device with pressure can be written as:

$$\frac{\Delta R}{R_T^0} = \frac{R_T^0 - R_T}{R_T^0} = \left(\frac{2R_C^{02}\Phi^{2/3}}{R_T^0\gamma}\right) p^{2/3} = \zeta p^{2/3}$$
(S8)

Eq. S8 indicates that for our assumption of contact-induced piezoresistivity, the change in resistance varies as a (2/3) exponent of the external pressure. This is confirmed in Fig. S6 where we represent the change in resistance as a function of the pressure in a log scale, as well as the (2/3) exponent direction.



Fig. S6 Change in resistance as a function of pressure in log-log scale.

The dependency with the diameter of the sphere is more difficult to estimate. Indeed, γ , R_T^0 , R_C^0 in Eq. S8 can have a complex dependence on the diameter of the sphere. Rather than extrapolating a model that cannot be trusted here, we prefer for this correlation to rely on experimental results that indicate a slight increase in sensitivity while decreasing the diameter of the sensing sphere.

Reference

S1. K.K. Liu, D.R. Williams, and B.J. Briscoe, J. Phys. D: Appl. Phys., 1998, 31, 294.

S2. Y.R. Jeng, and P.Y. Wang, J. Tribol., 2003, 125, 232.