

## **A Highly Permeable Fluorinated Polymer Nanocomposite for Plasmonic Hydrogen Sensing**

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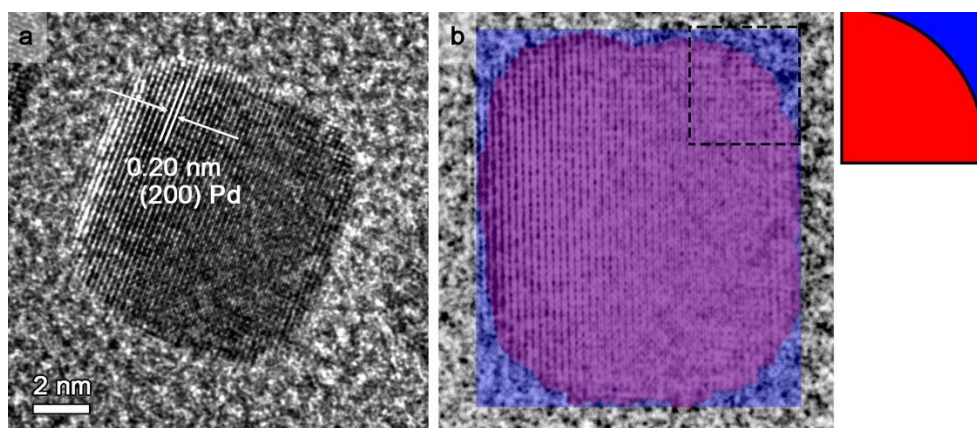
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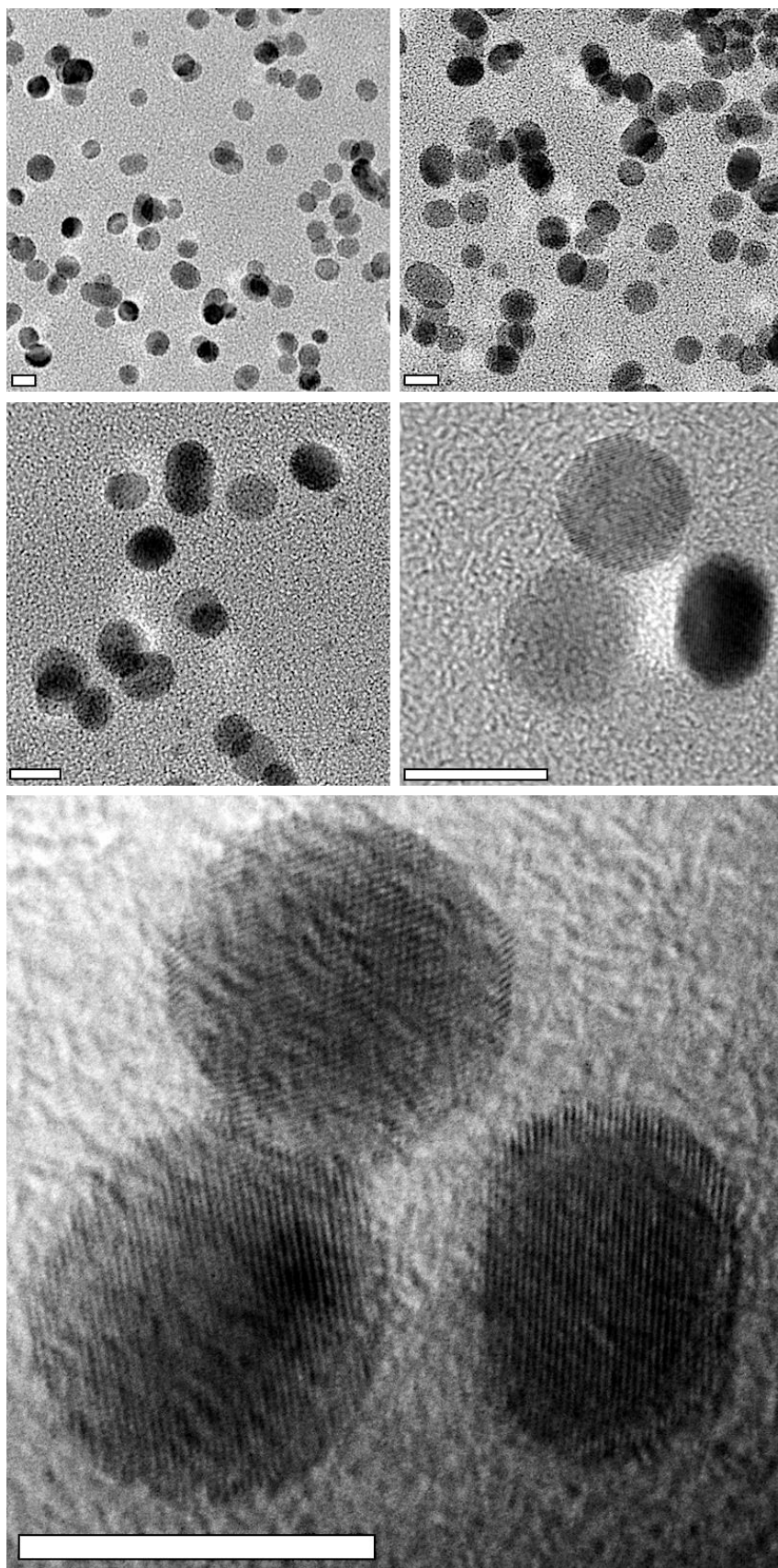


Blue Area = Expected area coverage for 100% sharp nanocube

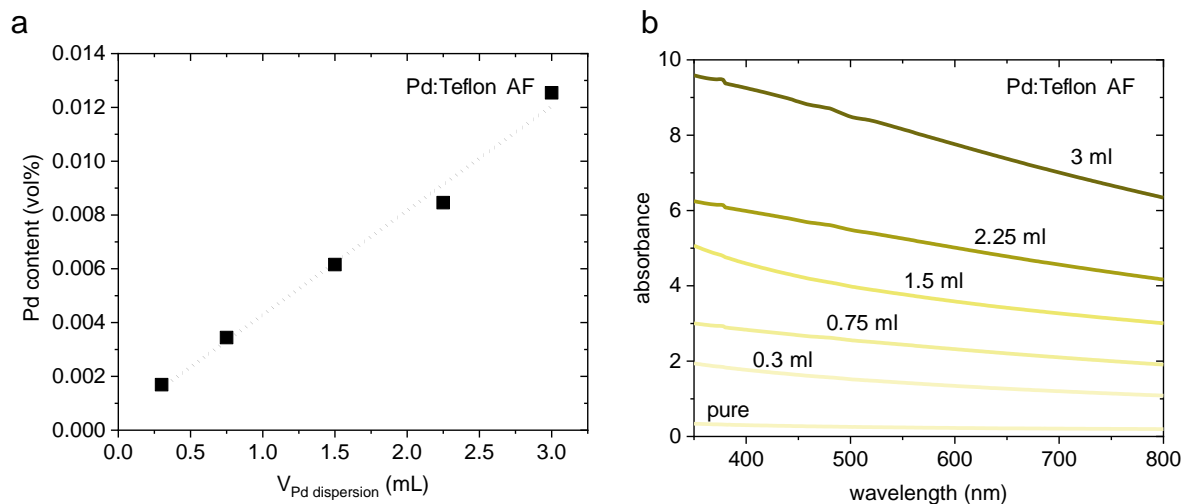
Red Area = Area covered by truncated nanocube

$$\% \text{ Truncation} = \frac{(\text{Blue Area} - \text{Red Area}) \times 100}{\text{Blue Area}}$$

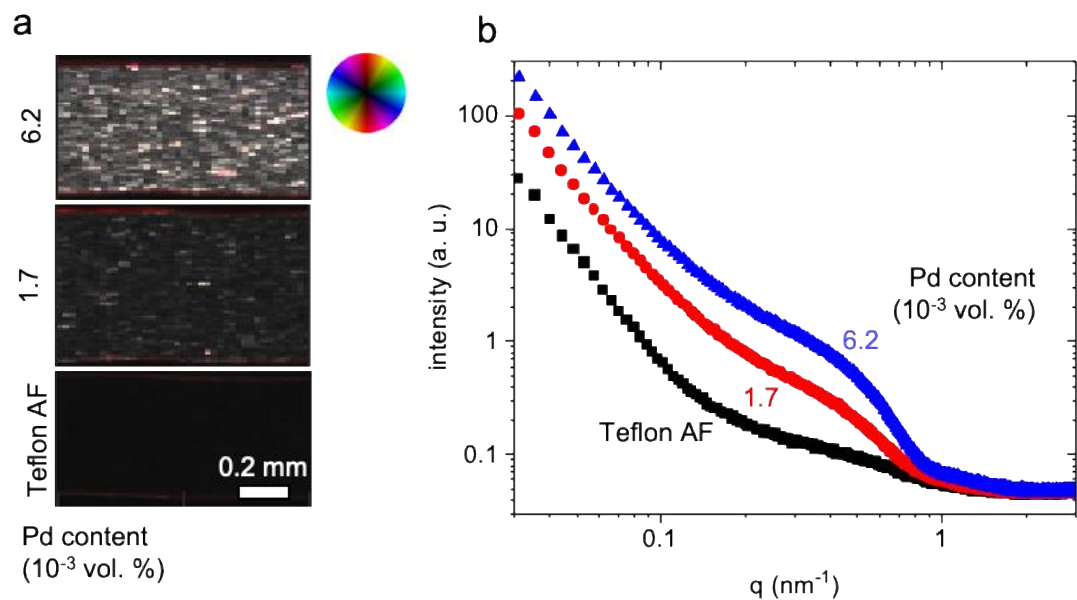
**Figure S1.** a) High-resolution TEM image of a single Pd nanocube, and b) calculation of edge-truncation degree of a single nanocube. The nanocubes showed ~15% of edge-truncation.



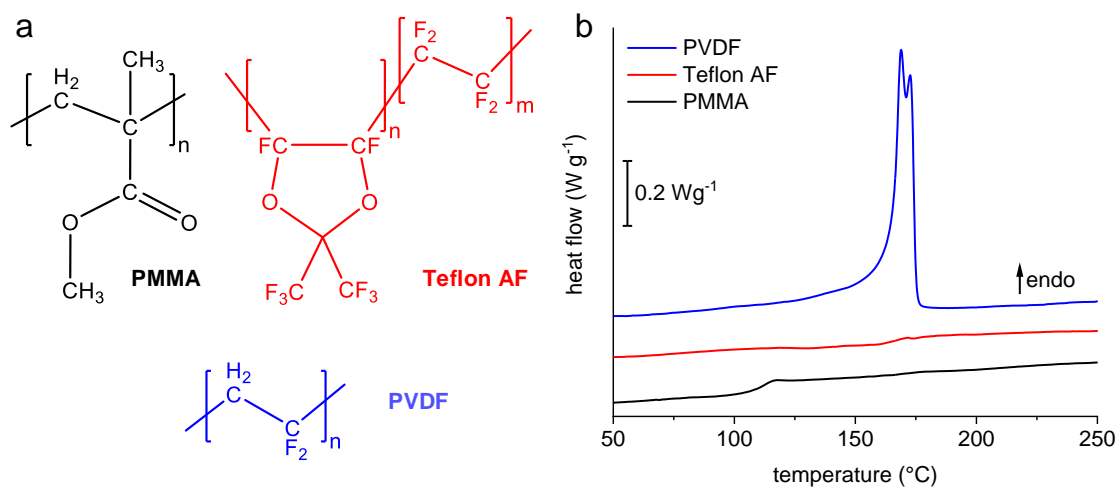
**Figure S2.** TEM images (different magnifications) taken from dispersed Pd nanoparticles in Teflon AF. Scale bars are 10 nm. Pd content  $\sim 8 \times 10^{-3}$  vol. %.



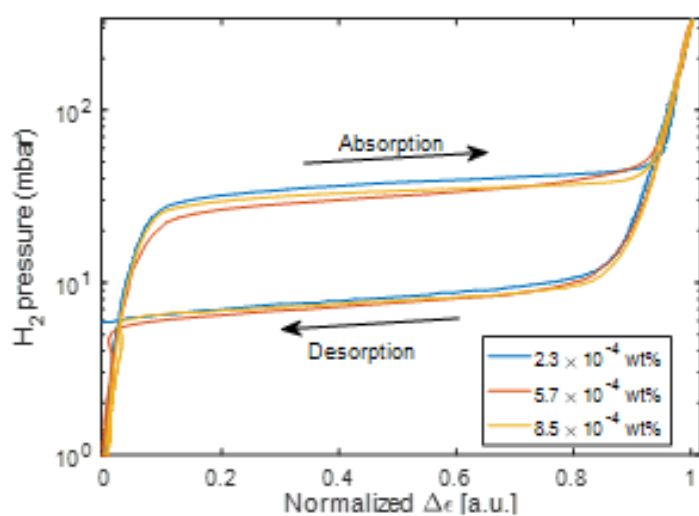
**Figure S3.** a) The Pd nanocube concentration in Pd:Teflon AF nanocomposites versus added volume of Pd nanocube suspension (isopropanol medium) after flow synthesis and medium exchange; b) UV-vis absorbance spectra of 100  $\mu$ m thick Pd:Teflon AF nanocomposite plates with different added volume of Pd nanocube suspension (isopropanol medium).



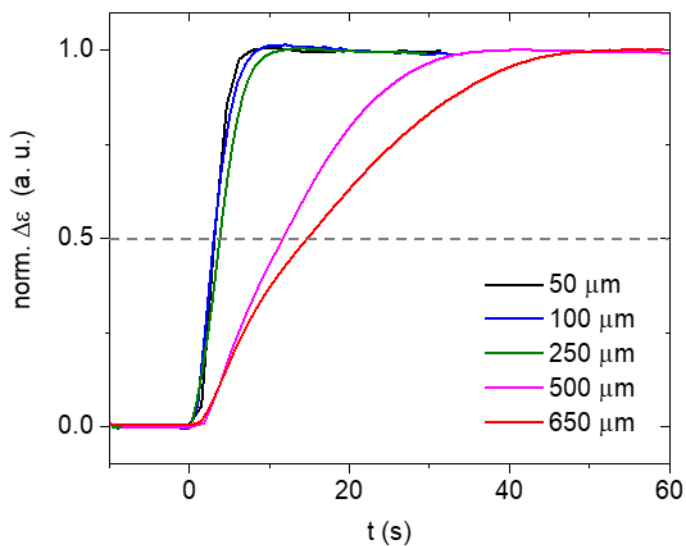
**Figure S4.** Small-angle X-ray scattering (SAXS) of Pd:Teflon AF nanocomposites. a) combined scanning-SAXS images of melt-pressed (combine the preferred orientation angle, the degree of orientation and the average scattering intensity in a hue-saturation-value representation according to the color wheel analyzed in a  $q$ -range of 0.2 to 0.69  $\text{nm}^{-1}$ ) and b) SAXS scattering curves of Pd:Teflon AF nanocomposites (raw data, i.e. before subtraction). Blue triangles correspond to a  $6.2 \times 10^{-3}$  vol % Pd particles and red circles to  $1.7 \times 10^{-3}$  vol % Pd particles.



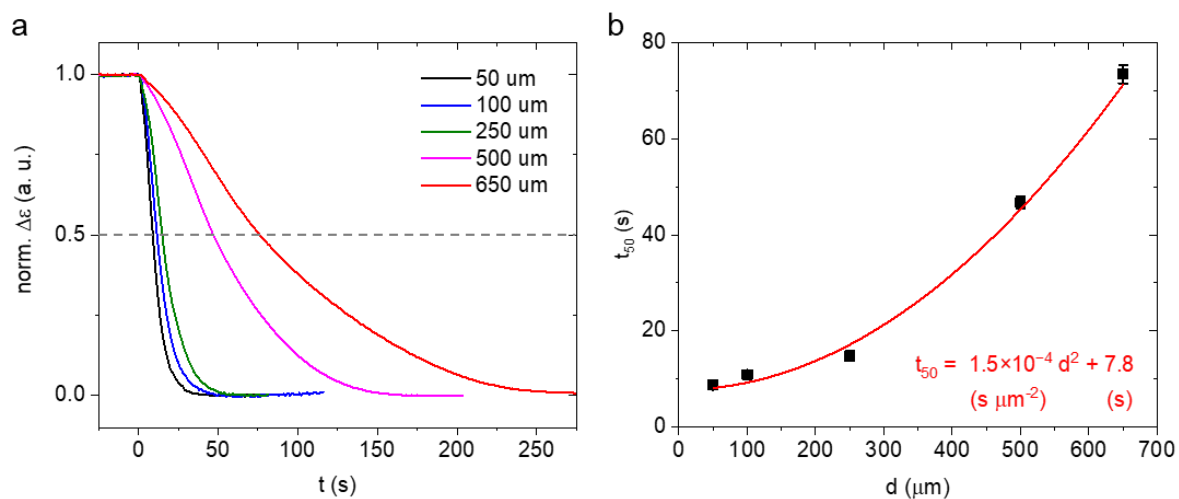
**Figure S5.** a) Chemical structure and b) heating DSC thermograms of PMMA, Teflon AF and PVDF.



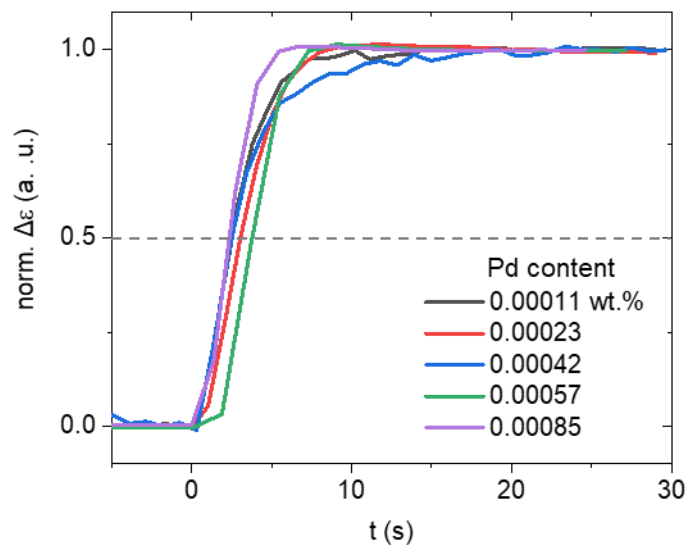
**Figure S6.** The pressure-composition isotherms of different Pd concentration in Teflon AF.



**Figure S7.** Normalized  $\Delta\varepsilon$  of melt-pressed Pd:Teflon AF ( $3.4 \times 10^{-3}$  vol. % Pd) plates upon a sudden increase in  $H_2$  pressure from 0 to 100 mbar  $H_2$  (the  $H_2$  valve opens at  $t = 0$ ).

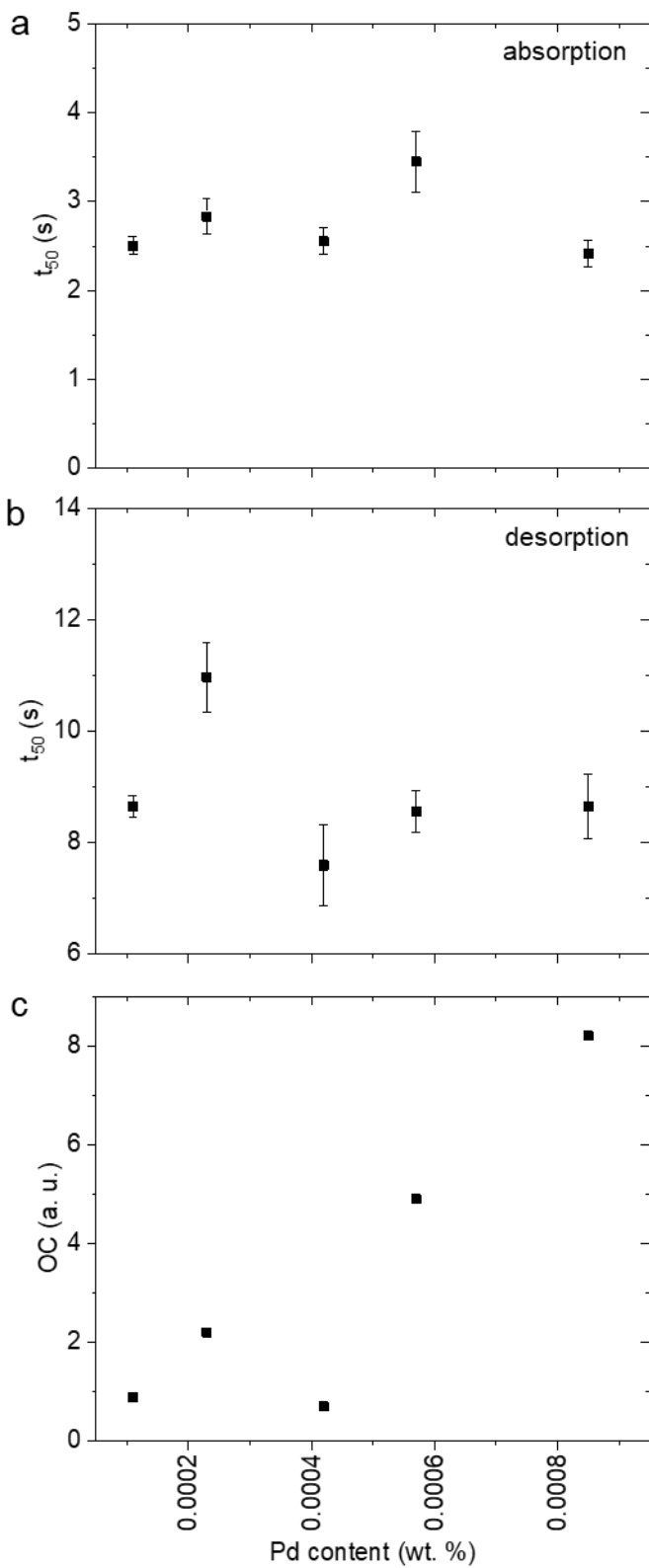


**Figure S8.** a) Normalized  $\Delta\varepsilon$  of melt-pressed Pd:Teflon AF ( $3.4 \times 10^{-3}$  vol. % Pd) plates during desorption; b) Corresponding sensor response time ( $H_2$  desorption time)  $t_{50}$ .

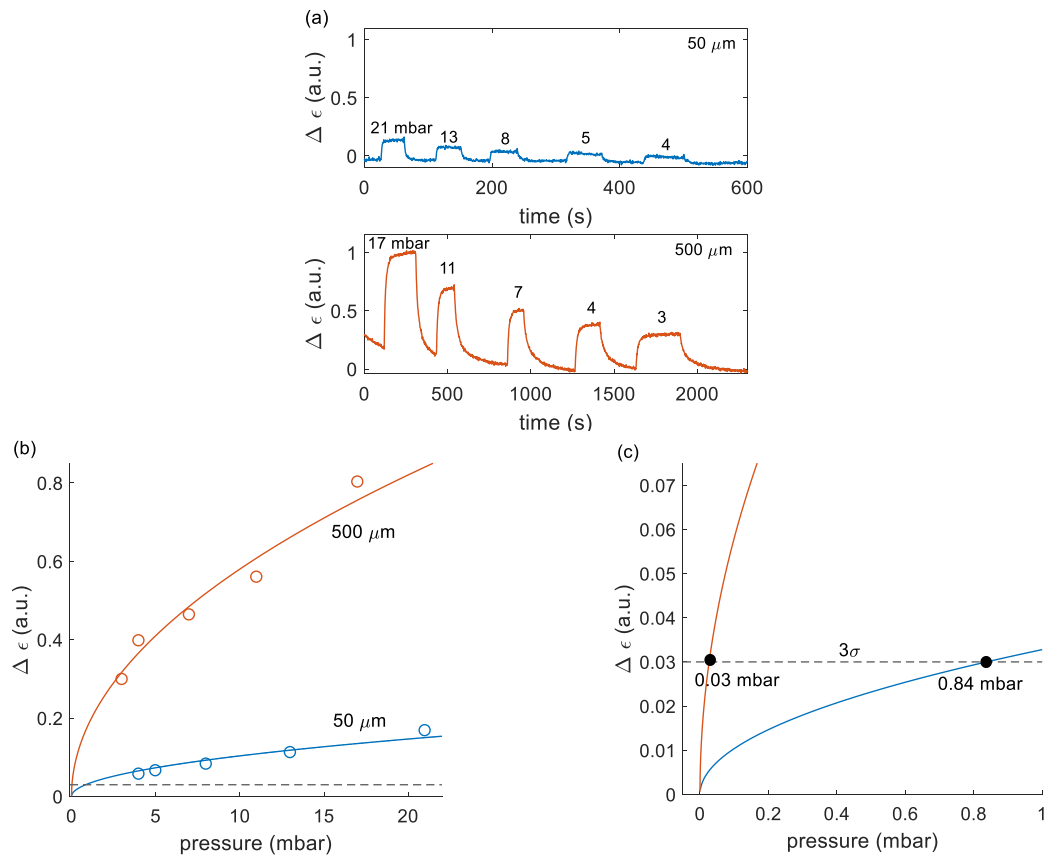


**Figure S9.** Normalized  $\Delta\varepsilon$  of 100  $\mu\text{m}$  thick melt-pressed Pd:Teflon AF plates with different Pd content.

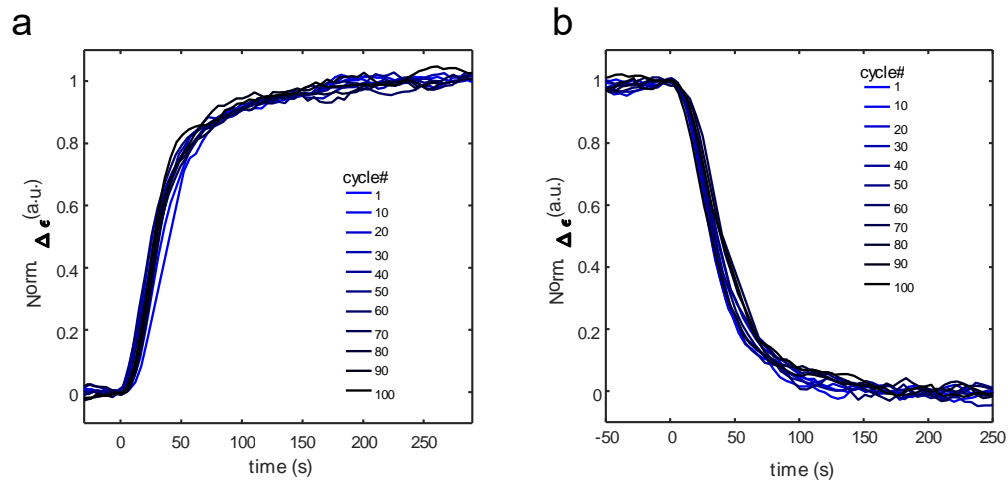




**Figure S10.** a) and b)  $H_2$  absorption and desorption time ( $t_{50}$ ), respectively and c) optical contrast of 100  $\mu\text{m}$  thick melt-pressed Pd:Teflon AF plates with different Pd content.



**Figure S11.** a) Sensor response of 50 and 500  $\mu\text{m}$  thick sensor at low hydrogen pressures, b) the optical responses ( $\Delta\epsilon$ ) vs hydrogen pressure and c) a magnification of b) at very low pressure.



**Figure S12.** Normalized  $\Delta\epsilon$  of optical fiber cap Pd:Teflon AF ( $3.4 \times 10^{-3}$  vol. % Pd) a) absorption and b) desorption at during cyclic exposure to 4 vol% hydrogen in synthetic air.