

Supporting Information

# A Highly Permeable Fluorinated Polymer Nanocomposite for Plasmonic Hydrogen Sensing

I. Östergren,<sup>1+</sup> A. M. Pourrahimi,<sup>1+</sup> I. Darmadi,<sup>2+</sup> R. R. da Silva,<sup>1</sup> A. Stolaś,<sup>1</sup> S. Lerch,<sup>1</sup> B. Berke,<sup>2</sup> M. Guizar-Sicairos,<sup>3</sup> M. Liebi,<sup>2</sup> G. Foli,<sup>4</sup> V. Palermo,<sup>4,5</sup> M. Minelli,<sup>6</sup> K. Moth-Poulsen,<sup>1\*</sup> C. Langhammer,<sup>2\*</sup> C. Müller<sup>1\*</sup>

<sup>1</sup>Department of Chemistry and Chemical Engineering, Chalmers University of Technology, 412 96 Göteborg, Sweden

<sup>2</sup>Department of Physics, Chalmers University of Technology, 412 96 Göteborg, Sweden

<sup>3</sup>Paul Scherrer Institut, PSI-Villigen, Switzerland

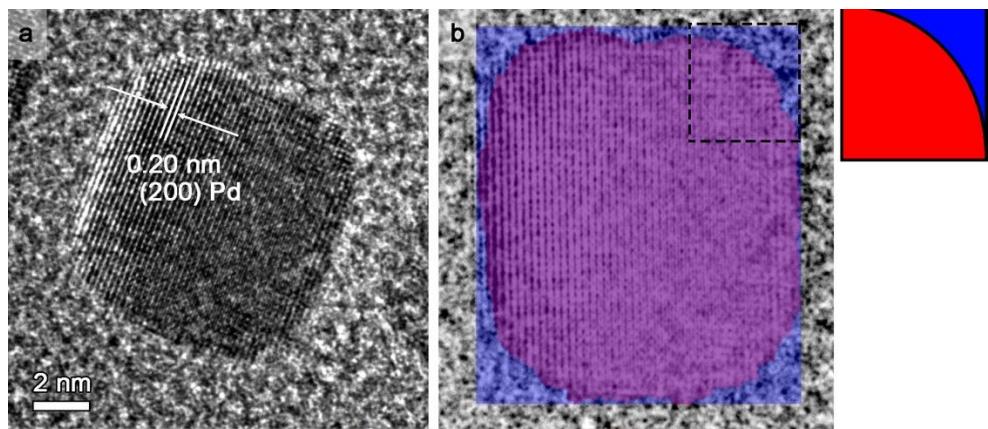
<sup>4</sup> Institute of Organic Synthesis and Photoreactivity, National Research Council, 40129, Bologna, Italy

<sup>5</sup> Department of Industrial and Materials Science, Chalmers University of Technology, 412 96 Göteborg, Sweden

<sup>6</sup>Department of Civil, Chemical, Environmental and Materials Engineering, Alma Mater Studiorum - University of Bologna, 40131 Bologna, Italy

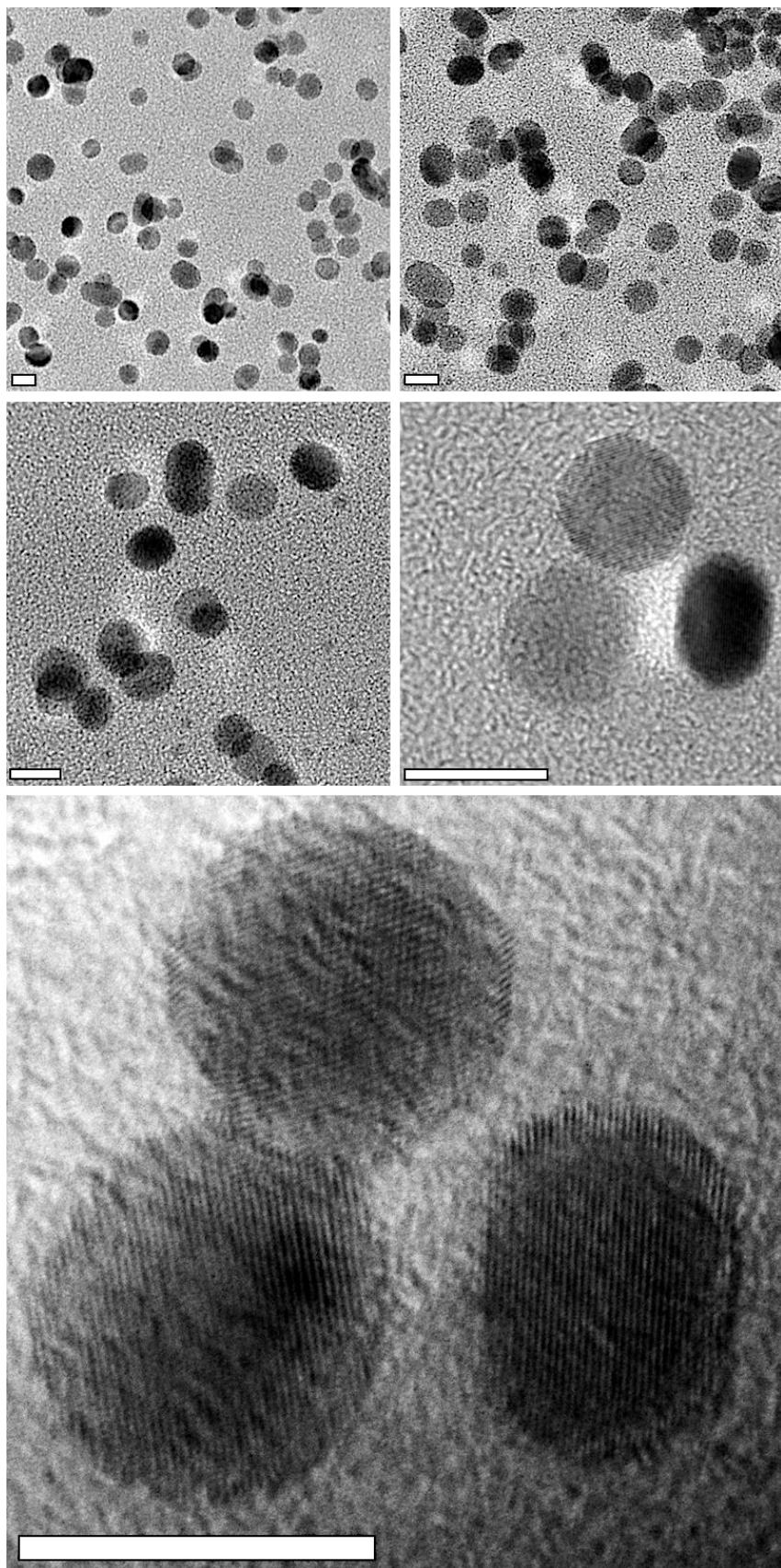
<sup>+</sup>equal contribution

\*Correspondence to: moth-poulsen@chalmers.se, christian.muller@chalmers.se, clangham@chalmers.se

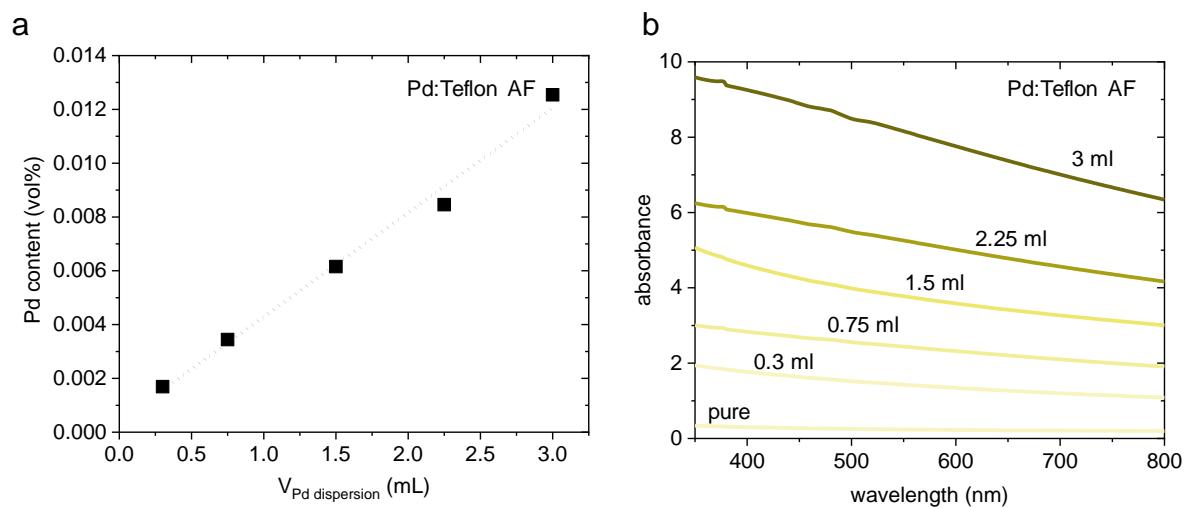


Blue Area = Expected area coverage for 100% sharp nanocube  
Red Area = Area covered by truncated nanocube  
$$\% \text{ Truncation} = \frac{\text{Blue Area} - \text{Red Area}}{\text{Blue Area}} \times 100$$

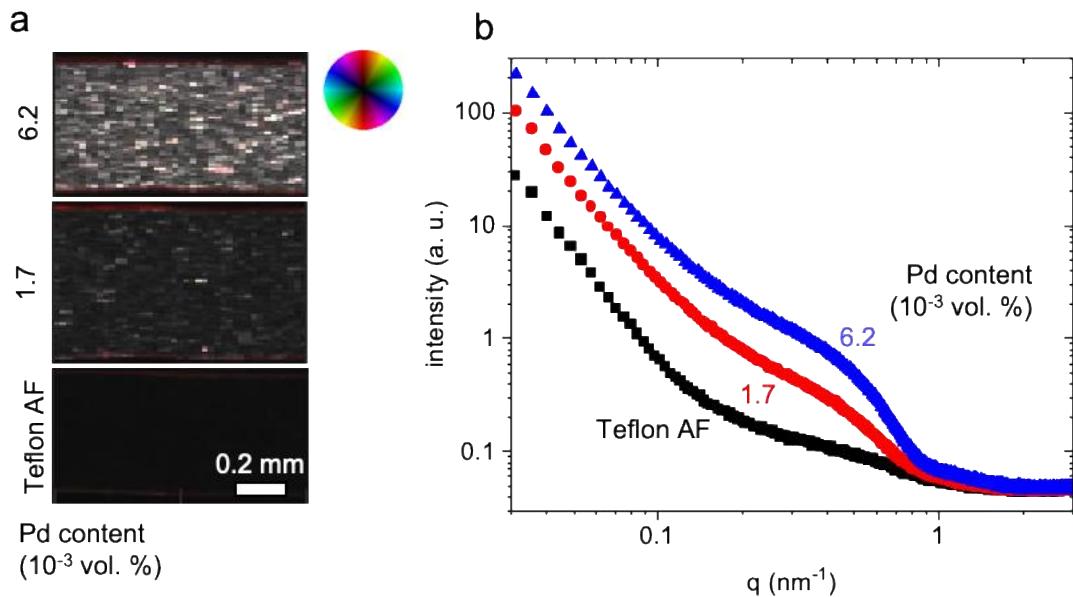
**Figure S1.** a) High-resolution TEM image of a single Pd nanocube, and b) calculation of edge-truncation degree of a single nanocube. The nanocubes showed ~15% of edge-truncation.



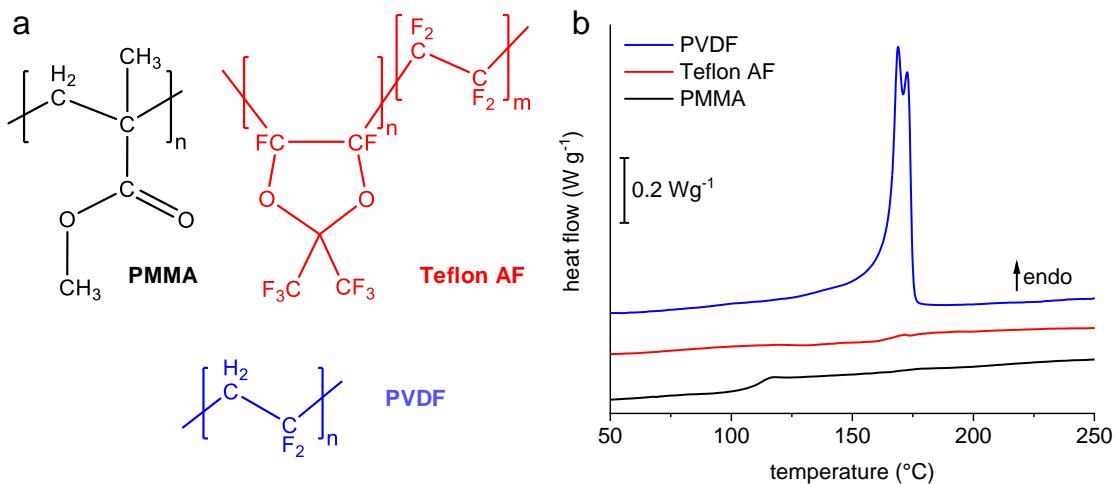
**Figure S2.** TEM images (different magnifications) taken from dispersed Pd nanoparticles in Teflon AF. Scale bars are 10 nm. Pd content  $\sim 8 \times 10^{-3}$  vol. %.



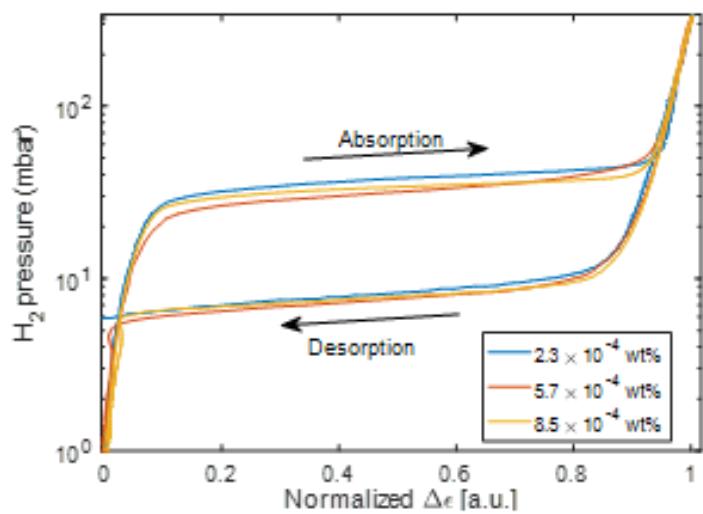
**Figure S3.** a) The Pd nanocube concentration in Pd:Teflon AF nanocomposites versus added volume of Pd nanocube suspension (isopropanol medium) after flow synthesis and medium exchange; b) UV-vis absorbance spectra of 100  $\mu\text{m}$  thick Pd:Teflon AF nanocomposite plates with different added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



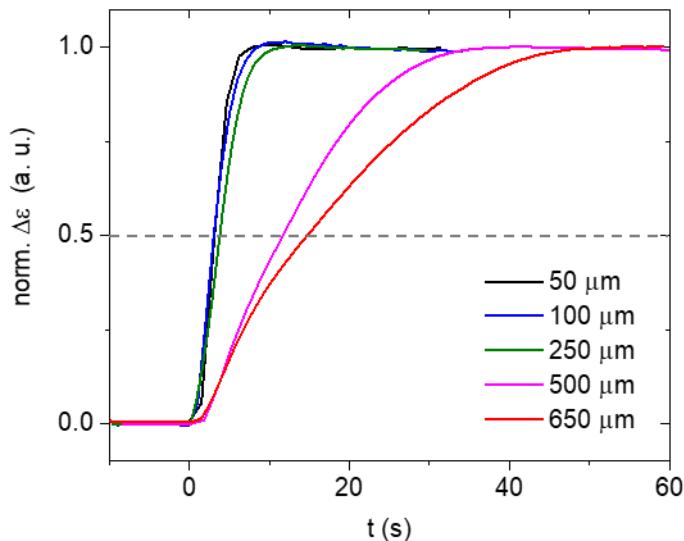
**Figure S4.** Small-angle X-ray scattering (SAXS) of Pd:Teflon AF nanocomposites. a) combined scanning-SAXS images of melt-pressed (combine the preferred orientation angle, the degree of orientation and the average scattering intensity in a hue-saturation-value representation according to the color 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.



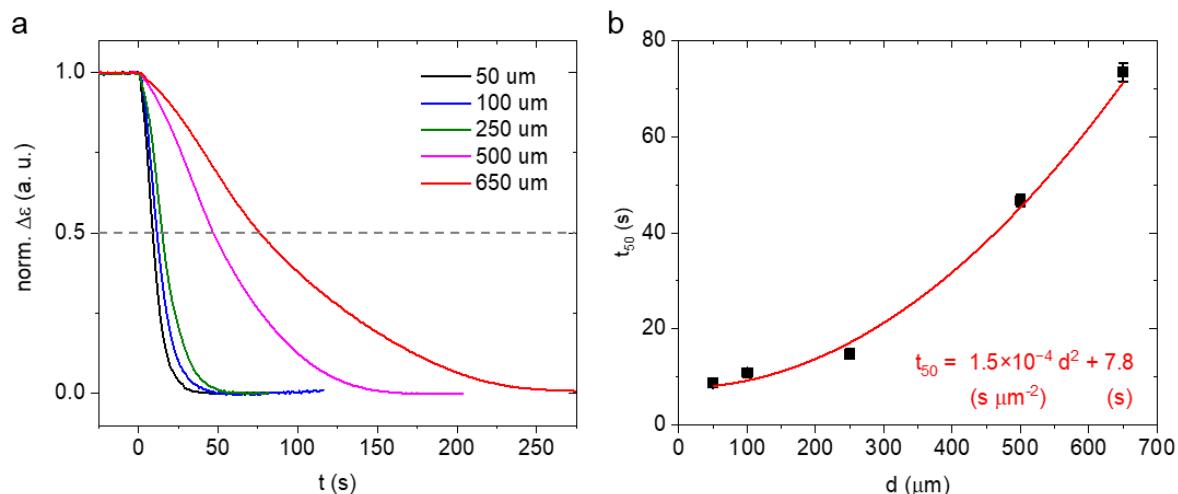
**Figure S5.** a) Chemical structure and b) heating DSC thermograms of PMMA, Teflon AF and PVDF.



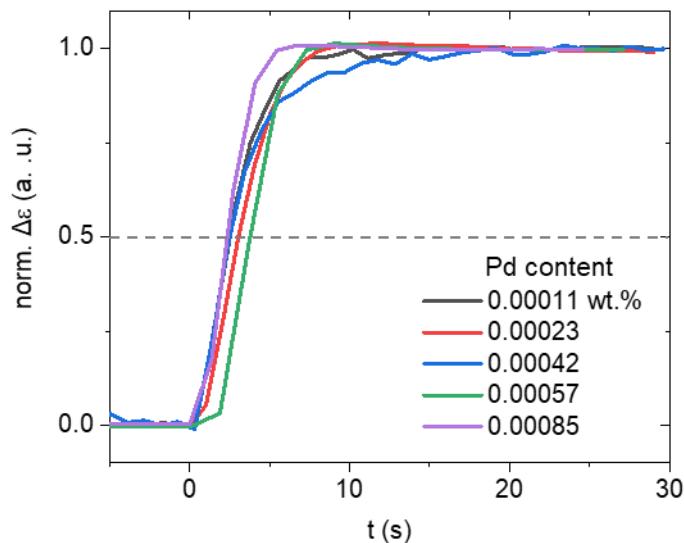
**Figure S6.** The pressure-composition isotherms of different Pd concentration in Teflon AF.



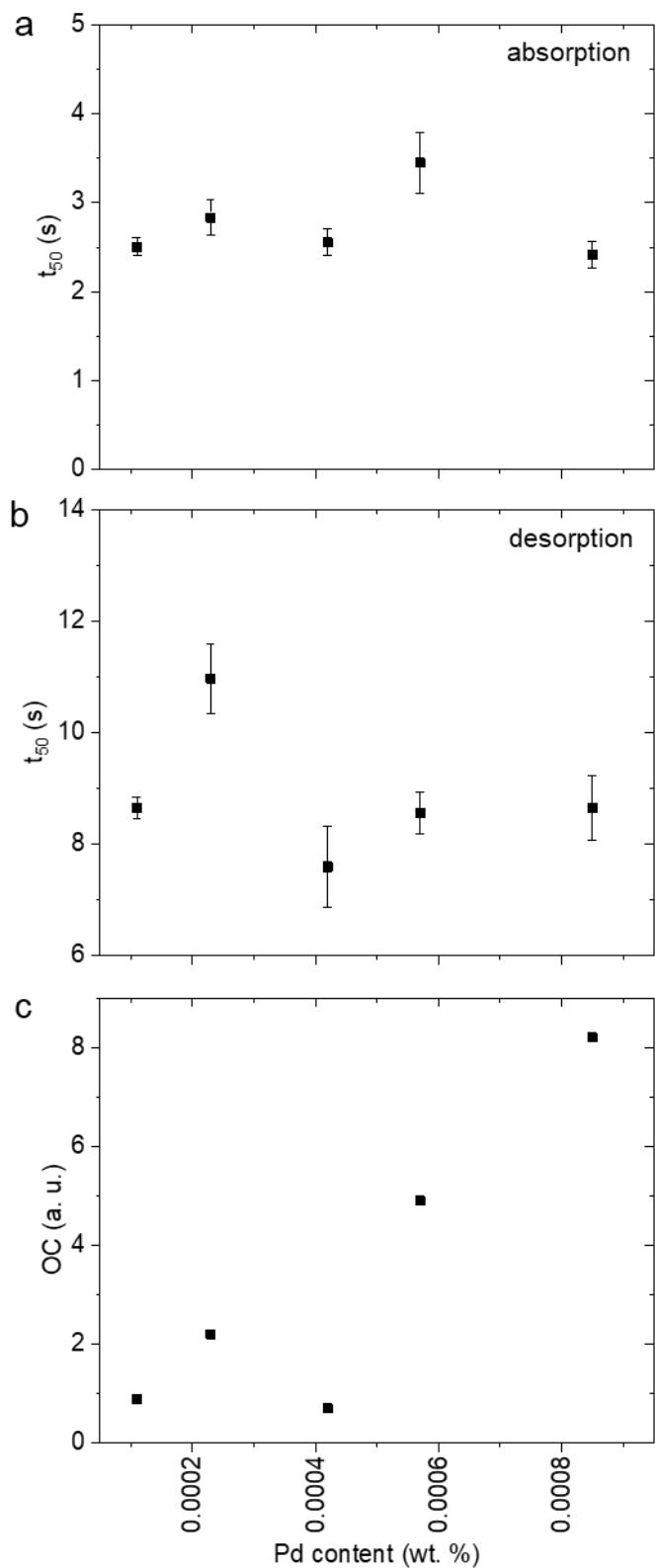
**Figure S7.** Normalized  $\Delta\epsilon$  of melt-pressed Pd:Teflon AF ( $3.4 \times 10^{-3}$  vol. % Pd) plates upon a sudden increase in  $\text{H}_2$  pressure from 0 to 100 mbar  $\text{H}_2$  (the  $\text{H}_2$  valve opens at  $t = 0$ ).



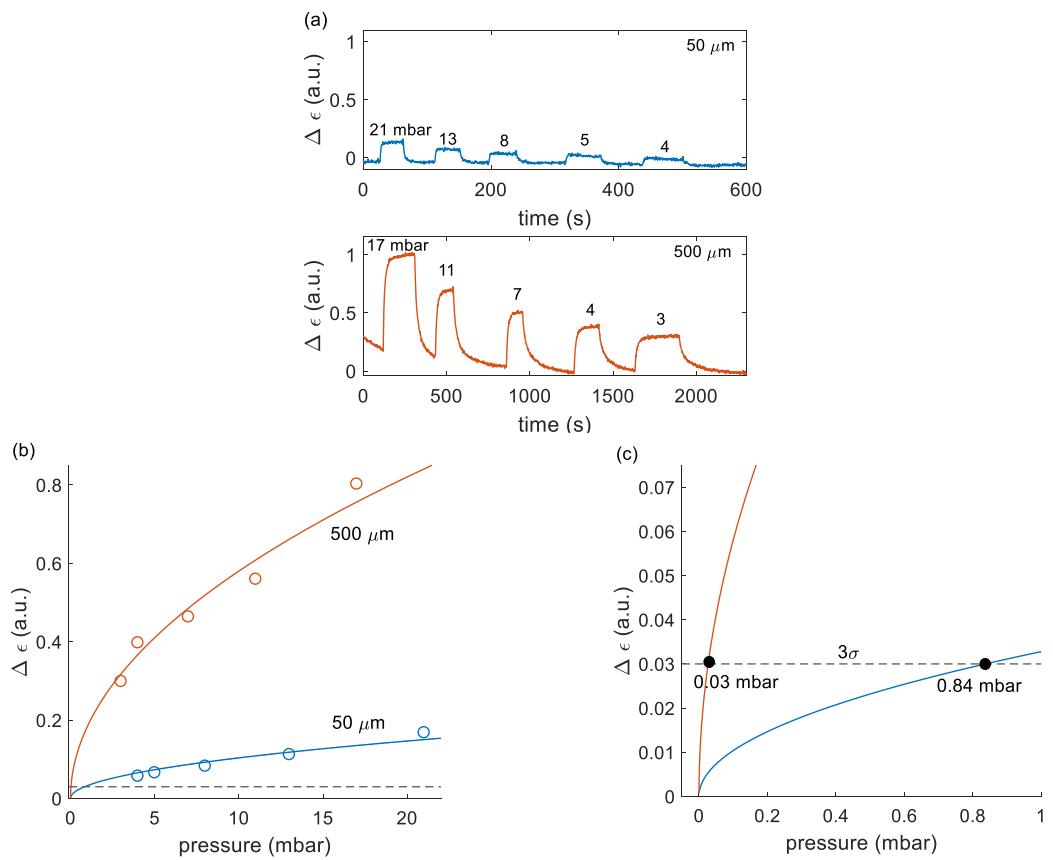
**Figure S8.** a) Normalized  $\Delta\epsilon$  of melt-pressed Pd:Teflon AF ( $3.4 \times 10^{-3}$  vol. % Pd) plates during desorption; b) Corresponding sensor response time ( $\text{H}_2$  desorption time)  $t_{50}$ .



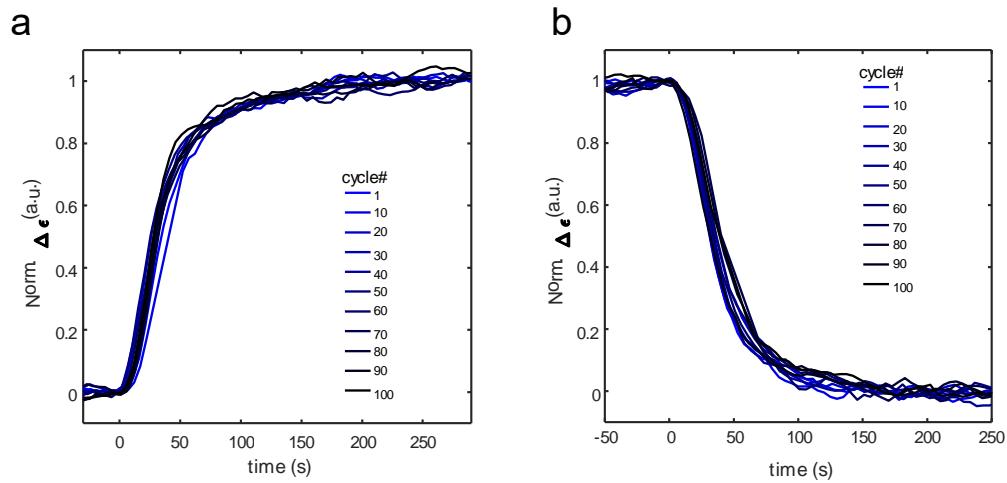
**Figure S9.** Normalized  $\Delta\epsilon$  of 100  $\mu\text{m}$  thick melt-pressed Pd:Teflon AF plates with different Pd content.



**Figure S10.** a) and b)  $\text{H}_2$  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 during cyclic exposure to 4 vol% hydrogen in synthetic air.