Integrated approach for preventing hydrogen leakage from metallic vessels: barrier and scavenging

SUPPORTING INFORMATION
Yonatan Luzzatto, † Amer Alatawna, † Eli Peretz, † Orit Mendelson, ‡ Svetlana Pevzner, ‡ and Oren Regev ‡

The rate of 1,4-bis(phenylethynyl)benzene (PEB) hydrogenation is:

\[
\frac{d[PEB]}{dt} = k(T)[PEB]^x[H_2]^y
\]  
(S1)

Where [PEB] is unsaturated concentration, x and y are the reaction orders, and k(T) is the temperature-dependent reaction constant. At a constant temperature and hydrogen pressure:

\[ k_{obs} = k[H_2]^y \]  
(S2)

Since the hydrogen pressure is constant during the experiment, and assuming a first-order reaction (x=1, approved experimentally, see Fig. S1), eqn S2 yields:

\[ \ln[PEB] = \ln[PEB]_0 - k_{obs}t \]  
(S3)

According to eqn S3, ln[PEB] shows a linear behavior with time, where the slope in the equation is the reaction constant (k_{obs}).

Fig. S1 (a) Reaction curves of getter-PDMS (orange) and getter-GNP-PDMS (green, 5 µm, 1 wt%). Dashed lines is a guid for the eye. The determination of \( t_{R.D} \) was determined by following the change in the curve of \( R^2 \) vs. \( t \). \( t_{R.D} \) is the time at which the maximum \( R^2 \) is obtained, as shown for the getter-PDMS (b) and getter-GNP-PDMS systems (c).
Fig. S2 EDS mapping of aggerates formed after hydrogenation at 296 K in a getter-PDMS system. A large aggregate was chosen (much above the average size) to simplify the analysis.

<table>
<thead>
<tr>
<th>Element</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbon</td>
<td>81.7</td>
</tr>
<tr>
<td>Silicone</td>
<td>18.0</td>
</tr>
<tr>
<td>Palladium</td>
<td>0.1</td>
</tr>
<tr>
<td>Other</td>
<td>0.2</td>
</tr>
</tbody>
</table>

Table S1. Elemental analysis of the aggerates based on EDS (see Fig. S2).
Fig. S3 A SEM micrograph of cross-sections in the getter-GNP-PDMS (5 µm GNP) system after hydrogenation at (a) 296 K, (b) 313 K: the aggregates were rather small (31 ± 11 µm and 29 ± 13 in size, n=50) and uniformly dispersed. (c) At 333 K, larger aggregates (112 ± 40 µm in size, n=50) were found at the bottom of the sample, most probably due to PEB liquefaction followed by precipitation (delineated by a dashed line. Insets: size distributions of the aggregates. (d-f) higher magnification of the aggregates after hydrogenation at (d) 296 K, (e) 313 K, and (f) 333 K. In figures (a-c), the scalebar is 500 µm, and in (d-f), it is 200 µm.
Fig. S4 Cross-sections of (a) getter-PDMS and (b) getter-GNP-PDMS system after hydrogenation at 333 K. The scale bar refers to both (a) and (b).
**Fig. S5** A composite-based coating (polymer-GNP) created a tortuous path, decreasing hydrogen diffusion and, consequently, the composite permeability.

**Fig. S6** A composite-based coating (polymer-PEB) that absorbed hydrogen, where unreacted and partially and fully hydrogenated PEB are shown. The red arrow symbols the reaction between hydrogen and the triple bond of a PEB molecule.