Supporting Information

Vacuum dried flexible hydrophobic aerogels using bridged methylsiloxane as reinforcement: performance regulation with alkylorthosilicate or alkyltrimethoxysilane co-precursors

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Table S1. The detailed formulas of the products.

<table>
<thead>
<tr>
<th>sample</th>
<th>APDEMS (mmol)</th>
<th>TPAL (mmol)</th>
<th>TMOS (mmol)</th>
<th>TEOS (mmol)</th>
<th>MTMS (mmol)</th>
<th>PTMS (mmol)</th>
<th>HTMS (mmol)</th>
<th>EtOH (ml)</th>
<th>Water (ml)</th>
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<tbody>
<tr>
<td>DT</td>
<td>12.81</td>
<td>6.39</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>18</td>
<td>2</td>
</tr>
<tr>
<td>DT-Q-M</td>
<td>4.27</td>
<td>2.13</td>
<td>8.54</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>18</td>
<td>2</td>
</tr>
<tr>
<td>DT-Q-E</td>
<td>4.27</td>
<td>2.13</td>
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<td>8.54</td>
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<td>0</td>
<td>0</td>
<td>18</td>
<td>2</td>
</tr>
<tr>
<td>DT-T-M</td>
<td>4.27</td>
<td>2.13</td>
<td>0</td>
<td>0</td>
<td>8.54</td>
<td>0</td>
<td>0</td>
<td>18</td>
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</tr>
<tr>
<td>DT-T-P</td>
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<td>0</td>
<td>18</td>
<td>2</td>
</tr>
<tr>
<td>DT-T-H</td>
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<td>0</td>
<td>0</td>
<td>0</td>
<td>8.54</td>
<td>18</td>
<td>2</td>
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</table>

Figure S1. DT-Q-M in drying process (a), DT-T-H in sol-gel process (b).
Figure S2. The FT-IR spectrum of the BMSQ precursor, in the synthesis of the aerogel, the mixture of ethanol, TPAL and ATDEMS was measured by the FT-IR before water was added, the generation of the imine and the disappearance of the aldehyde group means the precursor was synthesized.

Figure S3. The FT-IR spectra of the start monomers.
Figure S4. The Solid-state $^{13}$C NMR spectra of the aerogels.

Figure S5. The $^{13}$C NMR spectra of the start monomers.
Figure S6. The SEM images of the aerogels with 40000 times magnification.

Figure S7. Photos of the sol-gel process of the samples in every 2 hours.
Figure S8. Pore size distribution of the aerogels, a) DT, b) DT-Q-E, c) DT-T-M, d) DT-T-P.

Figure S9. The optical images of aerogels, a) DT-T-3M, b) DT-Q-3E.

To improve the specific surface areas, the content of MTMS and TEOS was increased by 3 times as the content in DT-T-M and DT-Q-E individually, the two kind aerogels were named DT-T-3M and DT-Q-3E. DT-T-3M and DT-Q-3E both suffered larger shrinkage and even fragmentation, the specific surface area of DT-T-3M and DT-Q-3E are 69.88 and 265.41 m$^2$/g, and the shrinkage of DT-T-3M and DT-Q-3E are 33.7% and 27.9% individually.
Figure S10. The Nitrogen adsorption and desorption isotherms of a) DT-T-3M and b) DT-Q-3E.

Figure S11. Pore size distribution of the aerogels, a) DT-T-3M, b) DT-Q-3E.

Table R1. A comparison of the removal rate and separation efficiency of aerogel in this work and the rate of materials reported in previous works.

<table>
<thead>
<tr>
<th>Material</th>
<th>Removal rate (L·m⁻²·s⁻¹)</th>
<th>Separation efficiency</th>
<th>Reference</th>
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<tbody>
<tr>
<td>mesh</td>
<td>20</td>
<td>93%</td>
<td>1</td>
</tr>
<tr>
<td>mesh</td>
<td>15</td>
<td>96%</td>
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<tr>
<td>membrane</td>
<td>0.13</td>
<td>99%</td>
<td>3</td>
</tr>
<tr>
<td>membrane</td>
<td>0.09</td>
<td>100%</td>
<td>4</td>
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<tr>
<td>Aerogel (monolith)</td>
<td>0.18</td>
<td>99%</td>
<td>This work</td>
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