Supporting Information

Paper reinforced with regenerated cellulose: Sustainable and fascinating material with good mechanical performance, barrier property and shape-retaining in water

Weihua Zhang, Zhijiao Jing, Youna Shan, Xuesong Ge, Xindong Mu, Yijun Jiang, Hui Li and Pingwei Wu

Key Laboratory of Bio-based Materials, Qingdao Institute of Bioenergy and Bioprocess Technology, Chinese Academy of Sciences, Qingdao, 266101, China. *E-mail: jiangyj@qibebt.ac.cn

Institute of Materials Science and Engineering, Ocean University of China, Qingdao 266100, China. *E-mail: wupingwei@ouc.edu.cn

Figure S1. SEM images of the cross sections of the (a) origin paper, (b) RCP-4.
### Table S1. Mechanical properties of composite paper at 50 RH.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Toughness</th>
<th>Tensile strength</th>
<th>Elastic modulus</th>
<th>Elongation at break</th>
</tr>
</thead>
<tbody>
<tr>
<td>RCP-0</td>
<td>0.3±0.05</td>
<td>20±3</td>
<td>700±300</td>
<td>3±1</td>
</tr>
<tr>
<td>RCP-1</td>
<td>1.8±0.2</td>
<td>27±4</td>
<td>900±250</td>
<td>9±1</td>
</tr>
<tr>
<td>RCP-2</td>
<td>7±0.15</td>
<td>50±7</td>
<td>1000±300</td>
<td>18±3</td>
</tr>
<tr>
<td>RCP-3</td>
<td>5±0.3</td>
<td>60±8</td>
<td>1300±200</td>
<td>11±2</td>
</tr>
<tr>
<td>RCP-4</td>
<td>6.5±0.2</td>
<td>75±5</td>
<td>1900±100</td>
<td>10±2</td>
</tr>
</tbody>
</table>

### Table S2. Wet strength of the composite paper.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Toughness</th>
<th>Tensile strength</th>
<th>Elastic modulus</th>
<th>Elongation at break</th>
</tr>
</thead>
<tbody>
<tr>
<td>RCP-0</td>
<td>0.028±0.02</td>
<td>1±0.2</td>
<td>24±10</td>
<td>3±1</td>
</tr>
<tr>
<td>RCP-1</td>
<td>0.19±0.01</td>
<td>4±2</td>
<td>50±8</td>
<td>9±1</td>
</tr>
<tr>
<td>RCP-2</td>
<td>0.94±0.02</td>
<td>10±1</td>
<td>100±20</td>
<td>13±2</td>
</tr>
<tr>
<td>RCP-3</td>
<td>0.75±0.03</td>
<td>12±1</td>
<td>100±10</td>
<td>10±2</td>
</tr>
<tr>
<td>RCP-4</td>
<td>0.75±0.02</td>
<td>13±1</td>
<td>110±15</td>
<td>10±2</td>
</tr>
</tbody>
</table>
Table S3. Mechanical properties of composite paper at 50 RH.

<table>
<thead>
<tr>
<th>Sample</th>
<th>RC</th>
<th>Tensile strength</th>
<th>Elastic modulus</th>
<th>Elongation at break</th>
</tr>
</thead>
<tbody>
<tr>
<td>RP-0</td>
<td>0</td>
<td>20±3</td>
<td>700±300</td>
<td>3±1</td>
</tr>
<tr>
<td>RP-1</td>
<td>2.21</td>
<td>23±1</td>
<td>700±50</td>
<td>10±1</td>
</tr>
<tr>
<td>RP-2</td>
<td>9.38</td>
<td>39±1</td>
<td>800±100</td>
<td>14±3</td>
</tr>
<tr>
<td>RP-3</td>
<td>15.32</td>
<td>47±2</td>
<td>1400±80</td>
<td>15±1</td>
</tr>
<tr>
<td>RP-4</td>
<td>19.86</td>
<td>54±3</td>
<td>1700±100</td>
<td>12±2</td>
</tr>
</tbody>
</table>

Table S4. Wet strength of the composite paper without ECH.

<table>
<thead>
<tr>
<th>Sample</th>
<th>RC</th>
<th>Tensile strength</th>
<th>Elastic modulus</th>
<th>Elongation at break</th>
</tr>
</thead>
<tbody>
<tr>
<td>RP-0</td>
<td>0</td>
<td>1±0.2</td>
<td>24±10</td>
<td>3±1</td>
</tr>
<tr>
<td>RP-1</td>
<td>2.21</td>
<td>6±1</td>
<td>60±10</td>
<td>9±1</td>
</tr>
<tr>
<td>RP-2</td>
<td>9.38</td>
<td>8±1</td>
<td>90±10</td>
<td>11±3</td>
</tr>
<tr>
<td>RP-3</td>
<td>15.32</td>
<td>10±1</td>
<td>100±10</td>
<td>10±1</td>
</tr>
<tr>
<td>RP-4</td>
<td>19.86</td>
<td>11±1</td>
<td>110±10</td>
<td>10±2</td>
</tr>
</tbody>
</table>

Figure S2. a) Stress–strain curves of origin paper and cross-linked paper without ECH at 50 RH. b) Stress–strain curves of origin paper and cross-linked paper without ECH under wet conditions.
Figure S3. a) Stress-strain curves of regenerated cellulose cross-linked paper treated with 8 M H$_2$SO$_4$, 10 M NaOH and saturated NaCl for 10 min, respectively.

Figure S4. a) Young's modulus of the dry samples. b) Young's modulus of the wet samples.
Figure S5  a) Shape deformation of folded RCP-0 when immersed in water. b) Shape-retaining of folded RCP-4 when immersed in water. c) Shape deformation of wet paper (RCP-0). d) Shape-retaining of wet RCP-4.