Supplementary Information

Original Design of Nitrogen-doped Carbon Aerogels from Sustainable Precursors:
Application as Metal-Free Oxygen Reduction Catalysts

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Materials

All chemicals were used as received without further purification. D-(+)-glucose 99% was purchased from Roth Chemicals. N-acetyl-D-glucosamine (> 99%), D-(+)-glucosamine hydrochloride (> 99%) and cyanuric acid were purchased from Sigma-Aldrich. Gum arabic powder and phloroglucinol 99% anhydrous were purchased from Acros Organic. Nafion® 117 solution (5 wt% in a mixture of lower aliphatic alcohols and water) was purchased from Aldrich. Potassium hydroxide solution (0.1 M) and methanol were purchased from Merck. Platinum catalyst (referred as to Pt@C in the text), nominally 20 wt% on carbon black, was purchased from Alfa Aesar.
Figure S 1. Scanning electron microscopy (SEM) micrographs of carbon aerogels obtained after further thermal treatment under inert atmosphere at 950°C. (a-b) 950-G1G1A1; (c-d) 950-AG2; and (e-f) 950-AG21CA2.
**Figure S 2.** C1s (left) and N1s (right) X-ray photoelectron spectra (black solid lines) and deconvolution curves (colored solid lines) obtained for the N-containing organic aerogels series. (a-b) HTC-G1GA1; (c-d) HTC-G1AG1; (e-f) HTC-AG2; and (g-h) HTC-AG21CA2. 

*C1, C2 and C3 deconvoluted peaks correspond to \([C-C / CH_n]\), \([C-O / C=N]\) and \([C=O / C=O-N]\) species respectively.*
Figure S 3. C1s X-ray photoelectron spectra (black solid lines) and deconvolution curves (colored solid lines) obtained for the N-doped carbon aerogels obtained after pyrolysis at 950°C. (a) 950-G1GA1; (b) 950-G1AG1; (c) 950-AG2; and (d) 950-AG21CA2. $C1$, $C2$ and $C3$ deconvoluted peaks correspond to $[C-C / CH_n]$, $[C-O / C=N]$ and $[C=O / C=O-N]$ species respectively.
Aerogel obtained from gum arabic

Synthesis. In a typical synthesis, 0.89 g of gum arabic (from acacia tree; Sigma) and 0.31 g of phloroglucinol were mixed into a hydroalcoholic mixture of 4.75 g of water and 4.75 g of absolute ethanol. After vigorous stirring for few minutes, the hydroalcoholic solution was then added in a glass inlet (30 mL volume) sealed in a Teflon lined autoclave (45 mL volume) and placed in a laboratory oven preheated to the desired temperature (e.g. 180°C) and left for the desired reaction time (e.g. 20 h at 180°C). The as-synthesized gum arabic gel was then removed from the autoclave, washed a first time with a mixture water/ethanol, and then several times with ethanol, till the gel was saturated in alcohol. Then, the gel was dried by extraction with supercritical CO₂ as described in the manuscript (see experimental section).

Figure S 4. (a) SEM and (b) TEM micrographs of the gum arabic-based organic aerogel. (c) TEM micrograph of the same aerogel after pyrolysis at 950°C.
Table S 1. Elemental analysis and nitrogen sorption data extracted from gum arabic aerogel before and after pyrolysis.

<table>
<thead>
<tr>
<th>Samples</th>
<th>Elemental analysis</th>
<th>BET surface area m²·g⁻¹</th>
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<tbody>
<tr>
<td></td>
<td>wt.% C N N/C</td>
<td></td>
</tr>
<tr>
<td>Gum arabic aerogel - HTC</td>
<td>57.9 2.3 4.0</td>
<td></td>
</tr>
<tr>
<td>Gum arabic aerogel - 950°C</td>
<td>81.9 1.1 1.3</td>
<td>340</td>
</tr>
</tbody>
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Figure S 5. (a) Nitrogen sorption isotherms and (b) pore size distribution (DFT method, QSDFT equilibrium model) of the gum arabic-based aerogel after pyrolysis at 950°C.