Supplementary Information

Intercalation of alkylamines in layered MoO$_3$ and \textit{in-situ} carbonization for high-performance asymmetric supercapacitor

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**Fig. S1** SEM images of MoO$_3$ at different magnifications.

**Fig. S2** (a) EDX spectrum and (b) SEM image of the interlayered carbon obtained by dissolving MoO$_3$/C$_{HDA}$ in 2 mol·L$^{-1}$ NaOH solution to eliminate host MoO$_3$ layers.
As seen from Fig. S3a and b, the raw MoO$_3$ presents the well-defined (010) crystal planes with a narrow interlayer spacing about 0.69 nm, showing the typical layered structure. After introduction of DA, the interlayer spacing of (010) crystal planes is significantly increased to ~2.65 nm, further demonstrating the successful intercalation of alkylamines (Fig. S3c and d). A final calcination of MoO$_3$/DA leads to the formation of sandwich-like MoO$_3$/C$_{DA}$ hybrid nanostructures (Fig. S3e and f).

**Fig. S3** HRTEM images of MoO$_3$ (a, b), MoO$_3$/DA (c, d) and MoO$_3$/C$_{DA}$ (e, f).

According to the Eq.1 as following:

$$i = av^b$$

(1)

where $i$ is the current, $v$ is the scan rate, and both $a$ and $b$ are the constant parameters, the $b$ value can be determined as a slope of the linear plot between log $i$ vs. log $v$.

Typically, the $b$ value is equal to 1.0 for non-diffusion-controlled surface capacitive and 0.5 for diffusion-controlled redox reaction.

As shown in Fig. S4a, the fitting results show that the $b$ values in this work are 0.89 and 0.93 at the potentials of 0.08 and 0.49 V, respectively. It further confirms that the MoO$_3$/C$_{DA}$ has both EDLC and surface redox reactions.

In addition, the contribution of the intercalation capacitance can be calculated based on Eq. 2:

$$i(V) = k_1v + k_2v^{1/2}$$

(2)

where $k_1v$ and $k_2v^{1/2}$ correspond to the surface capacitive current and diffusion-
controlled current, respectively. By plotting $i/\sqrt{\nu}$ vs. $\nu^{0.5}$, $k_1$ and $k_2$ can be determined as the slope and intercept, respectively (Fig. S4b).

Fig. S4c shows the percentage of intercalation capacitive at different scan rates and it can be found that the intercalation capacitance decreases as the scan rate is increased due to the diffusion limit of the electrolytes.

Fig. S4 (a) log $i$ vs. log $\nu$, (b) $i/\sqrt{\nu}$ vs. $\nu^{0.5}$, and (c) a bar chart of the diffusion-controlled intercalation capacitance vs. scan rate of MoO$_3$/CDA.

Fig. S5 (a) Cyclic voltammetry curves of MoO$_3$/CDA//EG asymmetric supercapacitor at increasing voltage window from 1.0 V to 1.6 V (all acquired at 100 mV·s$^{-1}$) and (b) Corresponding galvanostatic charge-discharge curves at a current density of 1.0 A·g$^{-1}$ from 1.0 V to 1.6 V.
Table S1 XPS peak fitting results for MoO$_3$/C$_{PA}$ (a), MoO$_3$/C$_{DA}$ (b) and MoO$_3$/C$_{HDA}$ (c) in the Mo 3d region.

<table>
<thead>
<tr>
<th>Peak</th>
<th>Position (eV)</th>
<th>Relative peak area (%)</th>
<th>FWHM (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mo$^{4+}$(3d$_{5/2}$)</td>
<td>228.8</td>
<td>24.2</td>
<td>1.6</td>
</tr>
<tr>
<td>Mo$^{4+}$(3d$_{3/2}$)</td>
<td>233.1</td>
<td>4.3</td>
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<tr>
<td>Mo$^{6+}$(3d$_{5/2}$)</td>
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<td>16.3</td>
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<table>
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<th>FWHM (eV)</th>
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<td>22.5</td>
<td>1.5</td>
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Table S2 XPS peak fitting results for MoO$_3$/C$_{PA}$ (a), MoO$_3$/C$_{DA}$ (b) and MoO$_3$/C$_{HDA}$ (c) in the C 1s region.

<table>
<thead>
<tr>
<th>Peak</th>
<th>Position (eV)</th>
<th>Relative peak area (%)</th>
<th>FWHM (eV)</th>
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<tbody>
<tr>
<td>Mo–C</td>
<td>283.3</td>
<td>18.7</td>
<td>1.0</td>
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<td>C–H</td>
<td>283.8</td>
<td>59.8</td>
<td>1.4</td>
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<td>Intercalated C</td>
<td>285.3</td>
<td>21.5</td>
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<th>FWHM (eV)</th>
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<tr>
<td>Mo–C</td>
<td>283.5</td>
<td>21.2</td>
<td>1.1</td>
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<tr>
<td>C–H</td>
<td>284.4</td>
<td>49.0</td>
<td>1.1</td>
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<tr>
<td>Intercalated C</td>
<td>285.3</td>
<td>29.8</td>
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<th>FWHM (eV)</th>
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<td>Mo–C</td>
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<td>C–H</td>
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<td>61.3</td>
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<tr>
<td>Intercalated C</td>
<td>286.1</td>
<td>28.2</td>
<td>1.9</td>
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Table S3 A comparison of specific capacitance, rate capability and cycle stability of the present work with those reported MoO₃-based asymmetric supercapacitor.

<table>
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<tr>
<th>ASC</th>
<th>Capacitance</th>
<th>Rate capability</th>
<th>Cycling performance</th>
<th>Ref</th>
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</thead>
<tbody>
<tr>
<td>MoO₃/CDA//EG</td>
<td>88 F·g⁻¹ (1 A·g⁻¹)</td>
<td>69.3% (1 to 10 A·g⁻¹)</td>
<td>86.5% (1 A·g⁻¹ for 5000 cycles)</td>
<td>This work</td>
</tr>
<tr>
<td>MoO₃//AC</td>
<td>68 F·g⁻¹ (1 A·g⁻¹)</td>
<td>20.7% (0.5 to 10 A·g⁻¹)</td>
<td>113% (2 A·g⁻¹ for 10000 cycles)</td>
<td>1</td>
</tr>
<tr>
<td>MoO₃–PPy//CNTs–MnO₂</td>
<td>54 F·g⁻¹ (0.25 A·g⁻¹)</td>
<td>/</td>
<td>76% (5 A·g⁻¹ for 10000 cycles)</td>
<td>2</td>
</tr>
<tr>
<td>WO₃–x/MoO₃–x//PANI/carbon</td>
<td>216 mF·cm⁻² (2 mA·cm⁻²)</td>
<td>60.2% (2 to 20 mA·cm⁻²)</td>
<td>75% (5 mA·cm⁻² for 10000 cycles)</td>
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<tr>
<td>CF/MnO₂//CF/MoO₃</td>
<td>4.86 mF·cm⁻² (0.5 mA·cm⁻²)</td>
<td>65.8% (0.5 to 5 mA·cm⁻²)</td>
<td>89% (5 mA·cm⁻² for 3000 cycles)</td>
<td>4</td>
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<tr>
<td>MnO₂@TiN//N-MoO₃–x</td>
<td>10.3 mF·cm⁻¹ (0.25 mA·cm⁻¹)</td>
<td>/</td>
<td>80.3% (100 mV·s⁻¹ for 5000 cycles)</td>
<td>5</td>
</tr>
</tbody>
</table>

3 Reference