Supporting Information Contents

Heterotriangulene-based unsymmetrical squaraine dyes: synergistic effects of donor moiety and out-of-plane branched alkyl chains on dye cell performance

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Table S1. Selected dihedral angles of NSQs were calculated from the optimized ground state geometry

<table>
<thead>
<tr>
<th>NSQ Dyes</th>
<th>$\theta_1$</th>
<th>$\theta_2$</th>
<th>$\theta_3$</th>
<th>$\theta_4$</th>
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</thead>
<tbody>
<tr>
<td>NSQR</td>
<td>47.06</td>
<td>-0.33</td>
<td>-178.08</td>
<td>-</td>
</tr>
<tr>
<td>NSQ1</td>
<td>-0.05</td>
<td>0.05</td>
<td>-179.6</td>
<td>-</td>
</tr>
<tr>
<td>NSQ2</td>
<td>-0.43</td>
<td>0.57</td>
<td>-177.65</td>
<td>-</td>
</tr>
<tr>
<td>NSQ3</td>
<td>0.61</td>
<td>-0.24</td>
<td>-178.57</td>
<td>-176.66</td>
</tr>
</tbody>
</table>
**Figure S48.** Distance between sp$^3$-C (methyl group of HT) to –O atom of carboxylic acid, distance between the terminal carbon atomes of sp$^3$-branched alkyl chain and sp$^3$-C (indoline) to –O atom of carboxylic acid of NSQ3 calculated from the optimized ground state geometry using density functional theory (DFT) at B3LYP/6-31G** level with the Gaussian 09 program.

**Supplementary photovoltaic performance**
Figure S49. $J$–$V$ characteristics of NSQR and NSQ1-3 with deviation of 5 cells measured under simulated AM 1.5 G simulated sunlight (100 mW cm$^{-2}$).
Table S2. Photovoltaic performance of NSQ3 with different ratios of CDCA.

<table>
<thead>
<tr>
<th>Dye</th>
<th>$V_{oc}$ (V)</th>
<th>$J_{sc}$ (mA/cm$^2$)</th>
<th>$ff$ (%)</th>
<th>$\eta$ (%)</th>
<th>Amount of adsorbed dyes ($\times 10^{-7}$ mol cm$^{-2}$)$^a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>NSQ3/CDCA (1 eqv.)</td>
<td>0.541</td>
<td>20.11</td>
<td>65.6</td>
<td>7.14</td>
<td>0.94</td>
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<tr>
<td>NSQ3/CDCA (3 eqv.)</td>
<td>0.544</td>
<td>20.01</td>
<td>64.1</td>
<td>6.99</td>
<td>0.76</td>
</tr>
<tr>
<td>NSQ3/CDCA (5 eqv.)</td>
<td>0.541</td>
<td>19.51</td>
<td>63.2</td>
<td>6.67</td>
<td>0.64</td>
</tr>
<tr>
<td>NSQ3/CDCA (10 eqv.)</td>
<td>0.550</td>
<td>14.28</td>
<td>69.4</td>
<td>5.45</td>
<td>0.32</td>
</tr>
</tbody>
</table>

$^a$by dye desorption method, carried out in 2M ethanolic HCl.

Figure S50. UV-Vis absorption spectra of desorbed NSQR and NSQ1-3 dyes in 2 M HCl in EtOH.
Figure S51. (a) Bode plot of NSQ dye cells (with an applied potential of -0.5 V), and (b) $C_u$ as a function of voltage (with an applied potential of -0.3 V).

Supplementary references