Iodine induced 1-D Lamellar Self Assembly in Organic Ionic Crystal for Solid State Dye Sensitized Solar Cell

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Electronic Supplementary Information

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XRD pattern of (a) PiHI (with 0.05M I2), (b) TiO2 + SK1 and (c) TiO2 + SK1 + PiHI (with 0.05M I2)

Assignments of the IR vibration modes of piperidine and PiHI.
Fig. S1 $^1$H-NMR spectra for PiHI

Fig. S2 FTIR spectra for PiHI
Fig. S3 Simulated XRD pattern for PiHI with 0.05M I$_2$. Line indicate fitted curve and point indicate experimental points.

Fig. S4 Structure optimization through Density Function Theory (DFT) with basis set, LanL2DZ.
Methods of introduction of electrolyte in device to achieve $\parallel$ and $\perp$ lamellar phases for EIS study:

Here, we have developed the method to arrange the material through gravitational sedimentation in slow evaporation method using modified device fabrication. **Fig. S5** shows the step wise procedure of formation of parallel arrangement represented with cross section of device. We have taken two platinized electrode with one electrolyte containing 0.8 mm two hole inside. Sandwiched device prepare using 60 $\mu$m sealant between them and filled 0.05 M I$_2$ doped PiHI solution inside in the device (Step-I). Put the device for slow solvent evaporation under vacuum at 60°C temperature results the solid film formation inside which will be in layer by layer fashion. Repeated this procedure for 5-6 time to achieve packed material contacted with both electrodes. Here, based on gravitational sediment forces, the layers are formed parallel to the electrode surface.

**Fig. S6** shows the procedure to prepare device which contain the materials layer perpendicular to the electrode surface by viewing the cross section of device. Here, similar procedure repeated with keeping device vertically during the all step. Here, material arrange itself perpendicular to the electrode surface during the solvent evaporation gravitationally.
**Fig. S6** Device fabrication for perpendicular arrangement

**Fig. S7** TEM image of (a) Pure PiHI, (b) 0.04M I₂, (c) 0.05M I₂ and (d) 0.06M I₂
**Fig. S8** Cross-section SEM image of the photoanode with solid ionic conductor PiHI

**Fig. S9** XRD pattern of (a) PiHI (with 0.05M I$_2$), (b) TiO$_2$ + SK1 and (c) TiO$_2$ + SK1 + PiHI (with 0.05M I$_2$), (* indicates Bragg reflections for anatase TiO$_2$)
Table S1 Assignments of the IR vibration modes of piperidine and PiHI.

<table>
<thead>
<tr>
<th>Piperidine</th>
<th>PiHI</th>
<th>Assignments</th>
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<tbody>
<tr>
<td>3271</td>
<td>3425</td>
<td>( \nu (N-H) )</td>
</tr>
<tr>
<td>2924</td>
<td>2847</td>
<td>( \nu (C-H) )</td>
</tr>
<tr>
<td>2854, 2800, 2731</td>
<td>2862, 2816, 2777, 2716</td>
<td>( \nu_{as} (C-H) )</td>
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<tr>
<td>1443</td>
<td>1450, 1420</td>
<td>( \nu (C-C) )</td>
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<tr>
<td>1650</td>
<td>1612</td>
<td>( \delta (N-H) )</td>
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<tr>
<td>1319, 1257</td>
<td>1304</td>
<td>Ring breathing bands</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Aromatic secondary amine</td>
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