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

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Fig. S1 <sup>1</sup>H-NMR spectra for PiHI



Fig. S2 FTIR spectra for PiHI



Fig. S3 Simulated XRD pattern for PiHI with 0.05M I<sub>2</sub>. 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 || and $\perp$ lamellar phases for EIS study:

Here, we have develop the method to arrange the material through gravitational sedimentation in slow evaporation method using modified device fabrication. **Fig. S5** shows the step wise



Fig. S5 Device fabrication for parallel arrangement

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<sub>2</sub> 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  $I_2,$  (c) 0.05M  $I_2$  and (d) 0.06M  $I_2$ 



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<sub>2</sub>), (b) TiO<sub>2</sub> + SK1 and (c) TiO<sub>2</sub> + SK1 + PiHI (with 0.05M I<sub>2</sub>), (\* indicates Bragg reflections for anatase TiO<sub>2</sub>)

Piperidine	PiHI	Assignments
3271	3425	v(N-H)
2924	2847	v (C-H)
2854, 2800, 2731	2862, 2816, 2777, 2716	v as(C-H)
1443	1450, 1420	v (C-C)
1650	1612	δ(N-H)
1319, 1257	1304	Ring breathing bands Aromatic secondary amine

**Table S1** Assignments of the IR vibration modes of piperidine and PiHI.