

# Supporting Information

## **Solution-processed organometallic quasi-two-dimensional nanosheets as hole buffer layer for organic light-emitting devices**

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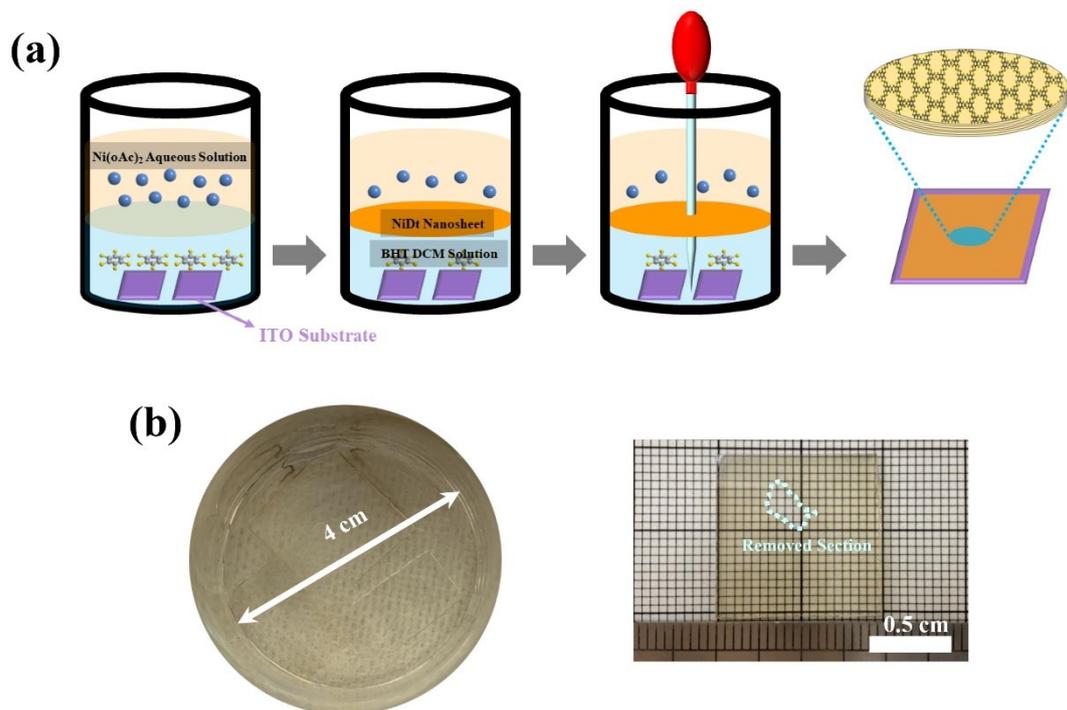
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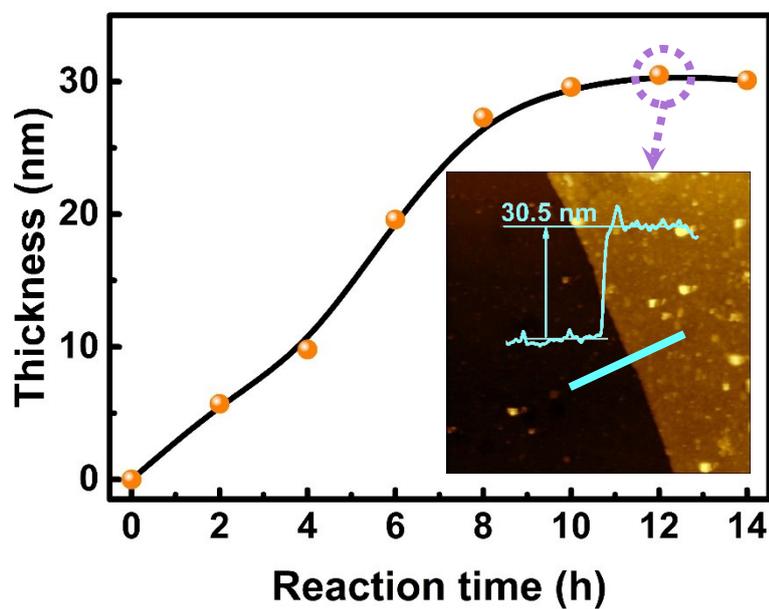
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**Fig. S1** (a) Schematic diagram for depositing NiDT organic nanosheet on ITO substrate in liquid/liquid interface fabrication process and (b) the obtained NiDT nanosheets on ITO substrate.



**Fig. S2** Thickness of NiDT nanosheets vs. reaction time; the inset is an atomic force microscopy image of the NiDT nanosheet obtained with a 12 h reaction.

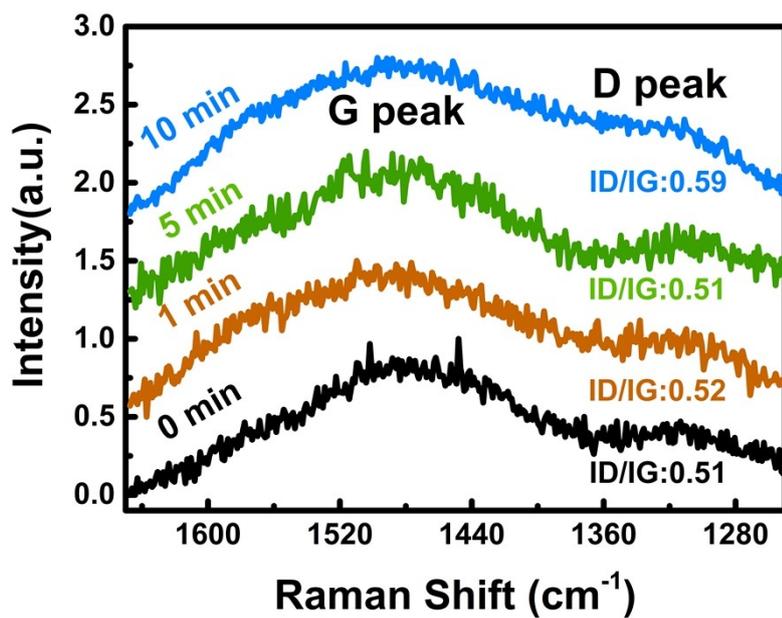


Fig. S3 Raman spectra of NiDT nanosheets after oxygen/plasma treatment

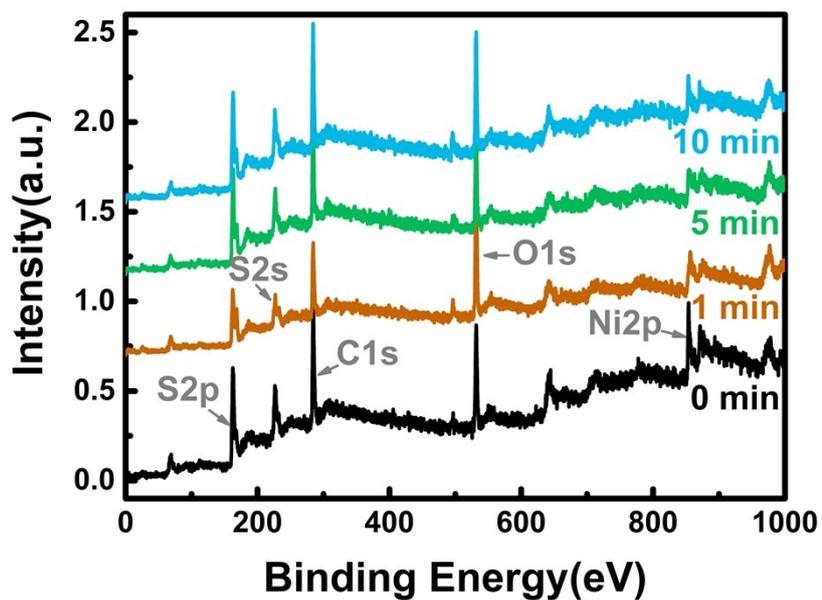
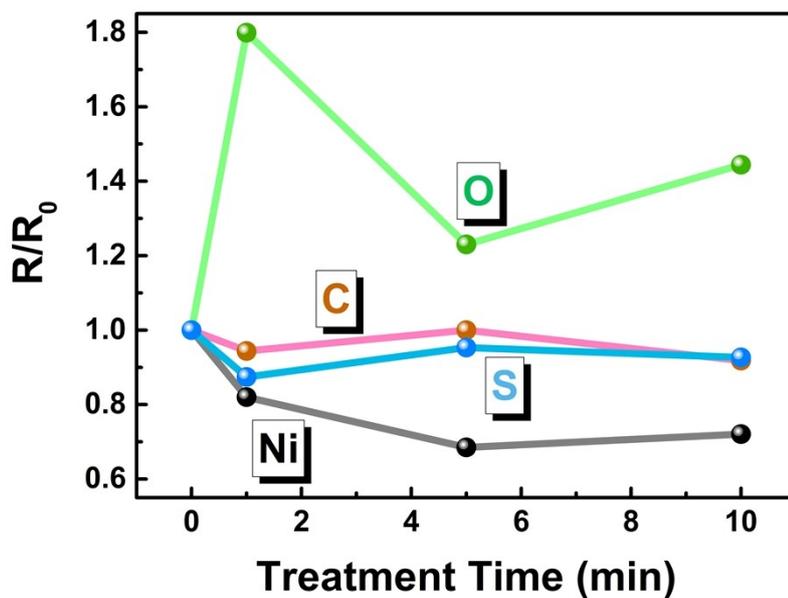
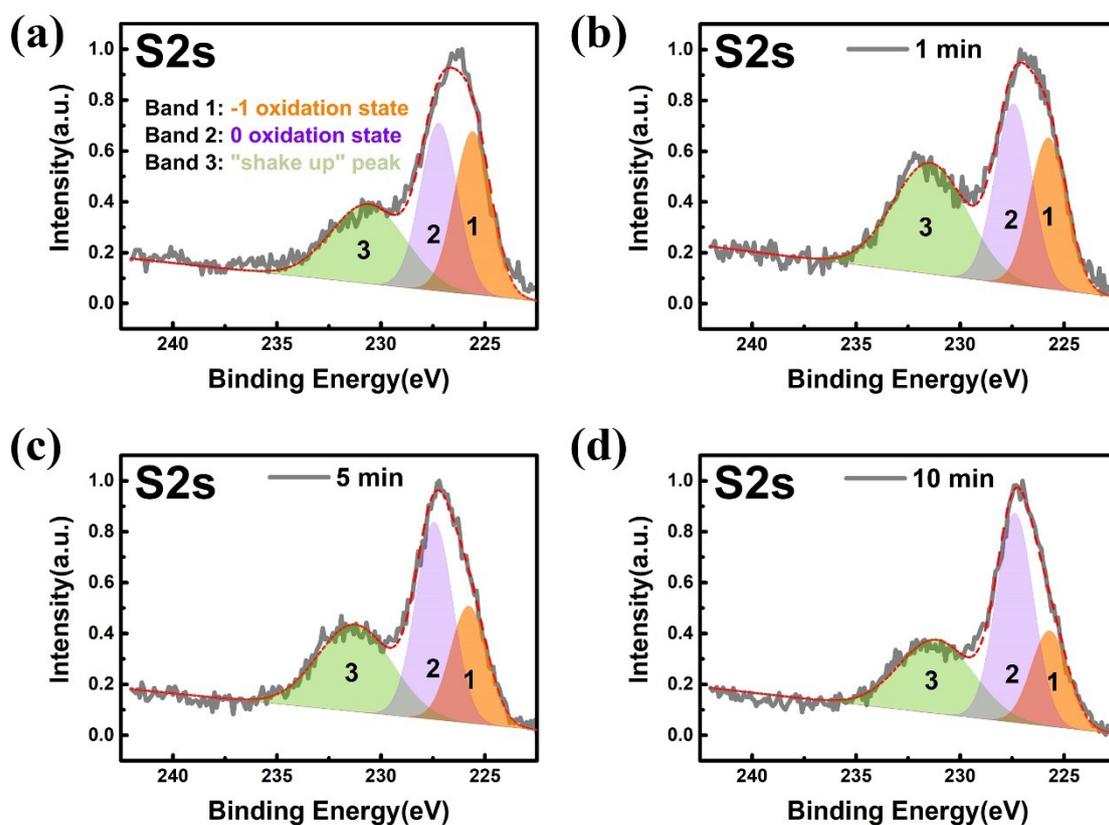


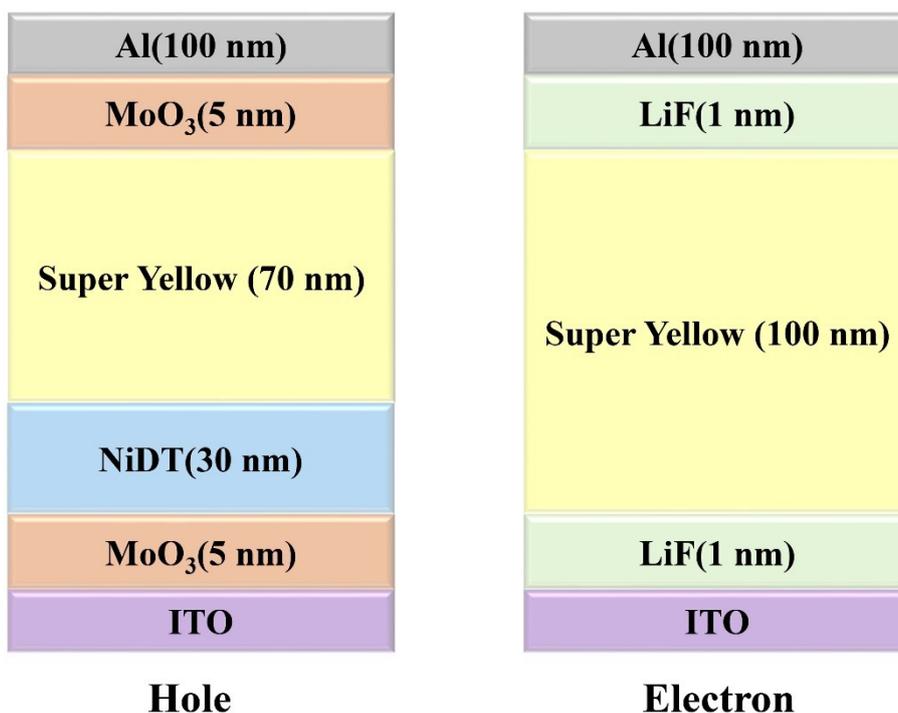
Fig. S4 XPS spectra of NiDT nanosheets after oxygen/plasma treatment



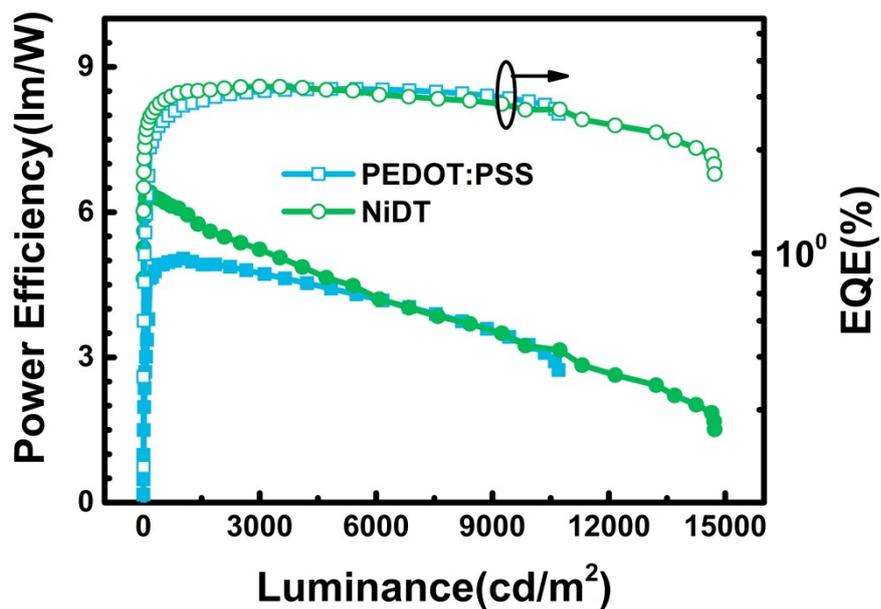
**Fig. S5** Normalized Atomic ratio  $R/R_0$  of Ni, C, O and S calculated from XPS spectra of NiDT nanosheets after oxygen/plasma treatment.  $R_0$  represents the atomic ratio of Ni, C, O and S for NiDT nanosheet without oxygen/plasma treatment.



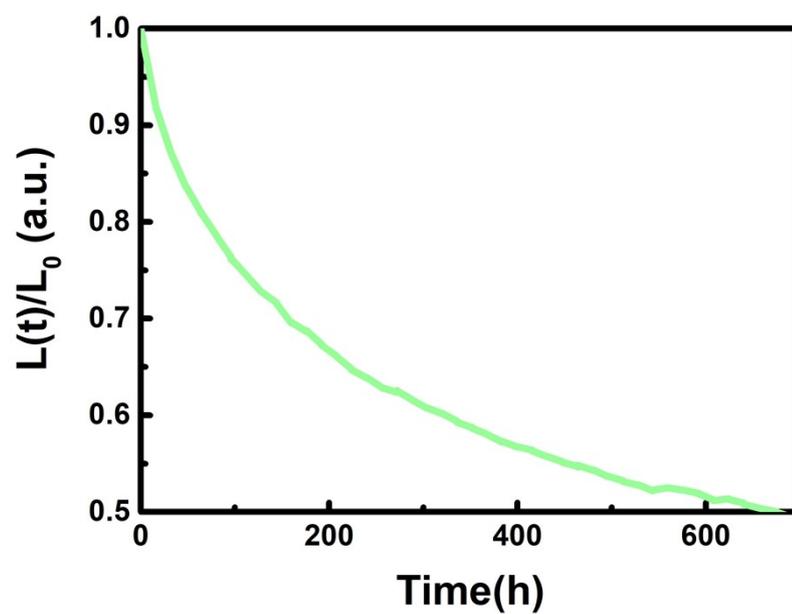
**Fig. S6** Deconvolution of S 2s peaks for NiDT nanosheet after oxygen/plasma treatment with (a) 0 min, (b) 1 min, (c) 5 min and (d) 10 min. Band 1 and 2 represents the -1 and 0 oxidation states, respectively, while band 3 is “shake-up” peak.



**Fig. S7** Device structure used for measuring hole and electron current density of yellow polymer OLEDs.



**Fig. S8** Power efficiency-luminance-external quantum efficiency (EQE) characteristics of NiDT and PEDOT:PSS devices with Super Yellow.



**Fig. S9** Normalized luminance degradation  $L(t)/L_0$  of the encapsulated NiDT employing  $\text{Ir}(\text{ppy})_3$  as emitter.  $L_0$  is the luminance of  $1000 \text{ cd/m}^2$ . The device structures are ITO/NiDT nanosheet/ CBP:10 wt%  $\text{Ir}(\text{ppy})_3$ /TPBi/LiF/Al.