

## Appendix. Supplementary data

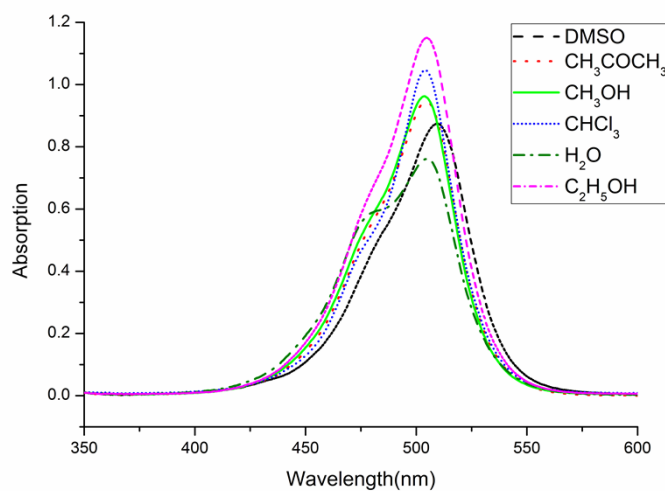
### 1. Synthesis of dyes D2 and D3

1,3-bis[4-(1-methyl-2-quinolinylidene-methyl) quinolinium]-2-propanol diiodide (**D2**): A mixture of 1,3-bis(4-methylquinolinium)-2-propanol dichloride (0.60 g, 1.4 mmol) and 1-methyl-2-methylthioquinolinium iodide (1.00 g, 3.2 mmol) was dissolved in anhydrous ethanol under reflux, and a few drops of triethylamine were added as a catalyst. The reaction mixtures were refluxed for 4 h. After cooling, the resulting precipitate was filtered off and purified by recrystallization from EtOH, to give pure green solid 0.54g. Yield: 44%, m.p.:210-211 °C. UV-Vis (MeOH)  $\lambda_{\text{max}}$ : 558.0 nm,  $\epsilon$ :  $1.02 \times 10^5 \text{ L} \cdot \text{mol}^{-1} \text{ cm}^{-1}$ .  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ )  $\delta$  (ppm): 4.03 (s, 6H, -NCH<sub>3</sub>), 4.29 (s, 1H, -OH), 4.34-4.50 (m, 2H, -NCH<sub>2</sub>), 5.04 (d,  $J=13.7$  Hz, 2H, -NCH<sub>2</sub>), 5.68-5.76 (m, 1H, -CH-), 6.53 (s, 2H, -CH=), 7.41 (d,  $J=7.2$  Hz, 2H, ArH), 7.53 (t,  $J=7.5$  Hz, 2H, ArH), 7.65 (t,  $J=7.5$  Hz, 2H, ArH), 7.82 (t,  $J=7.6$  Hz, 2H, ArH), 7.94 (t,  $J=7.6$  Hz, 2H, ArH), 7.97-8.02 (m, 5H, ArH), 8.08-8.15 (m, 5H, ArH), 8.28 (d,  $J=8.4$  Hz, 2H, ArH), 8.56 (d,  $J=8.4$  Hz, 2H, ArH). IR (KBr)  $\nu$ : 3440 (s,  $\nu_{\text{O-H}}$ ), 2920, 2855 (w,  $\nu_{\text{C-H}}$ ), 1598, 1500 (s,  $\nu_{\text{C=C}}$ ,  $\nu_{\text{C=N}}$ ), 1318 (s,  $\nu_{\text{C-O}}$ ), 1210, 1144, 1045 (s,  $\nu_{\text{C-N}}$ ,  $\delta_{\text{C-H}}$ ), 731 (s,  $\delta_{\text{C-H}}$ )  $\text{cm}^{-1}$ . HRMS (TOF MS ES-) calculated for  $\text{C}_{43}\text{H}_{38}\text{N}_4\text{O}^{2+}$ : 313.1517; found: 313.1536.

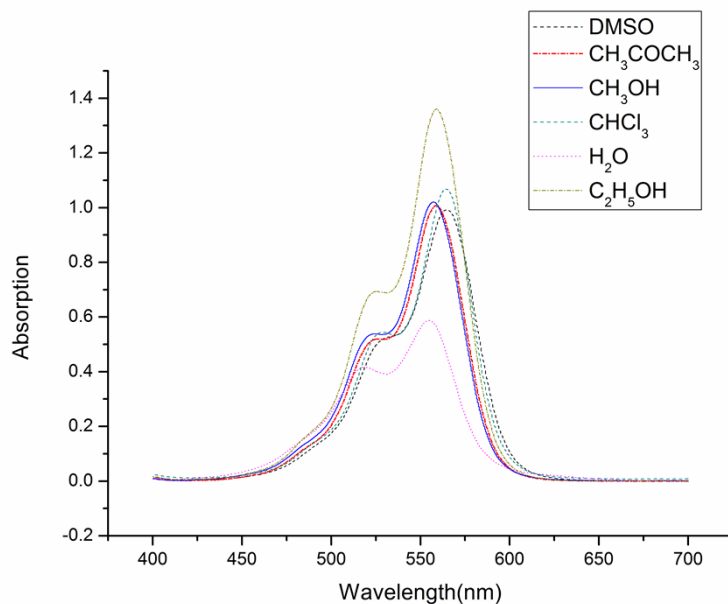
1,3-bis[4-(2-benzo[c, d]indolylidene-methyl) quinolinium]-2-propanol diiodide (**D3**): A mixture of 1,3-bis(4-methylquinolinium)-2-propanol dichloride (0.33 g, 0.8 mmol) and 2-methylthiobenzo[c,d]indolium iodide (0.58 g, 1.8 mmol) was dissolved in anhydrous ethanol under reflux, and a few drops of triethylamine were added as a catalyst. The reaction mixtures were refluxed for 6 h. After cooling, the resulting precipitate was filtered off and purified by recrystallization from EtOH, to give pure violet black solid 0.41 g. Yield: 57%, m.p.:256-257 °C. UV-Vis (MeOH)  $\lambda_{\text{max}}$ : 591.0 nm,  $\epsilon$ :  $1.07 \times 10^5 \text{ L} \cdot \text{mol}^{-1} \text{ cm}^{-1}$ .  $^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ )  $\delta$  (ppm): 3.85-3.95 (m, 4H, -NCH<sub>2</sub>), 4.17 (s, 1H, -OH), 4.55-4.61 (m, 1H, -CH-), 5.97 (s, 2H, -CH=), 7.30 (d,  $J=6.5$  Hz, 2H, ArH), 7.54-7.61 (m, 4H, ArH), 7.81-7.87 (m, 4H, ArH), 8.05-8.11 (m, 4H, ArH), 8.20 (d,  $J=6.9$  Hz, 2H, ArH), 8.32 (d,

$J=8.5$  Hz, 2H, ArH), 8.66 (d,  $J=7.2$  Hz, 2H, ArH), 8.78 (d,  $J=6.9$  Hz, 2H, ArH), 8.99 (d,  $J=8.5$  Hz, 2H, ArH). IR (KBr)  $\nu$ : 3440 (s,  $\nu_{\text{O-H}}$ ), 3242(w,  $\nu_{\text{N-H}}$ ), 1532 (s,  $\nu_{\text{C=C}}$ ,  $\nu_{\text{C=N}}$ ), 1301 (s,  $\nu_{\text{C-O}}$ ), 1219, 1161, 1069 (s,  $\nu_{\text{C-N}}$ ,  $\delta_{\text{C-H}}$ ), 813, 756 (s,  $\delta_{\text{C-H}}$ )  $\text{cm}^{-1}$ . HRMS (TOF MS ES-) calculated for  $\text{C}_{45}\text{H}_{34}\text{N}_4\text{O}^{2+}$ : 323.1361; found: 323.1429.

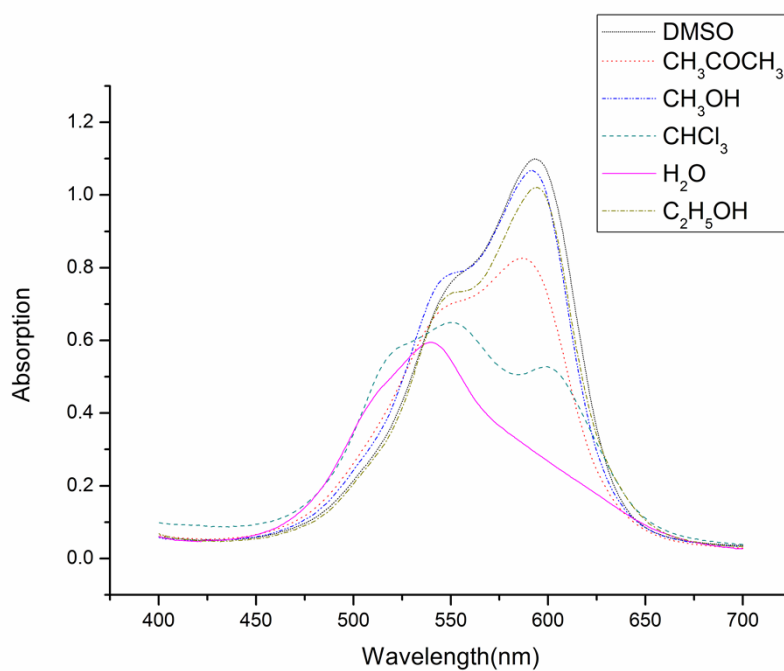
## 2. The absorption spectra of dyes D1, D2 and D3



**Fig. S1** Absorption spectra of dye **D1** in different solvents



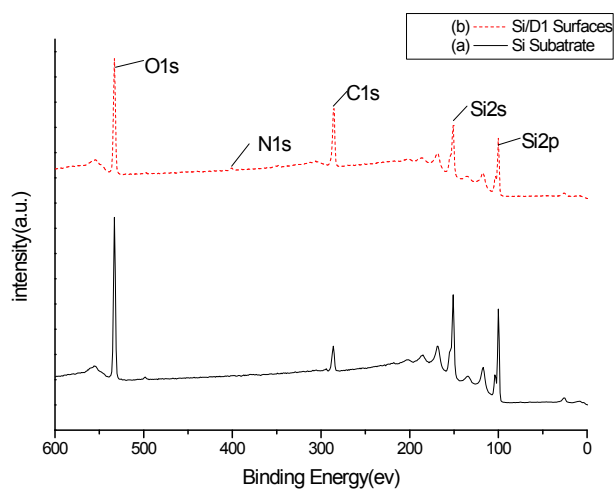
**Fig. S2** Absorption spectra of dye **D2** in different solvents



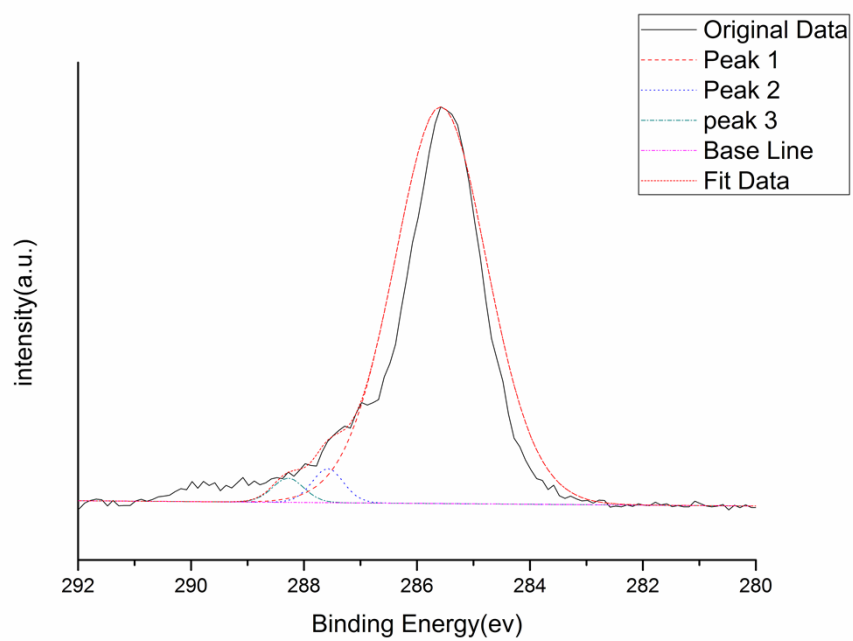
**Fig. S3** Absorption spectra of dye **D3** in different solvents

### 3. The X-ray photoelectron spectroscopy of Si/D2 and Si/D3

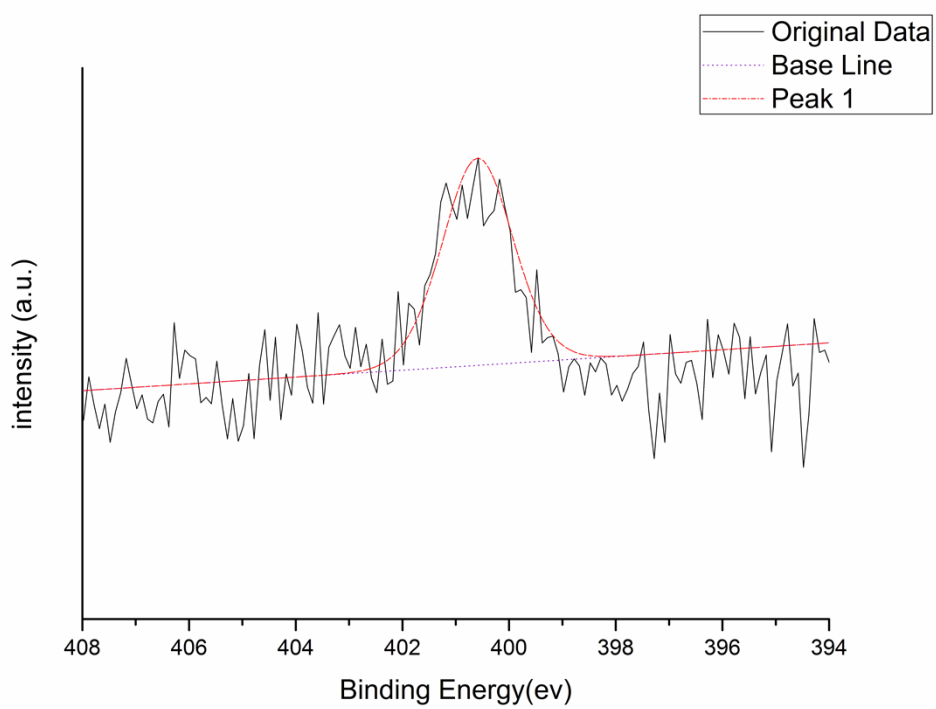
The  $C_{1s}$ ,  $O_{1s}$  and  $N_{1s}$  spectra of Si/**D2** and Si/**D3** were showed in Fig. S4-S11.



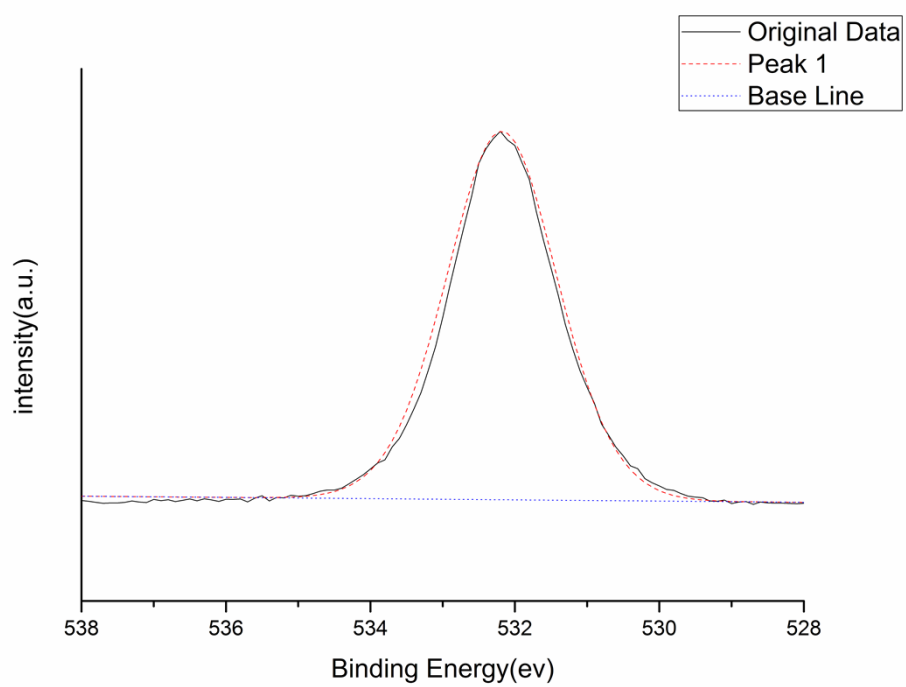
**Fig. S4.** The XPS scan spectra of Si substrate (a) and Si/**D2** (b) surfaces



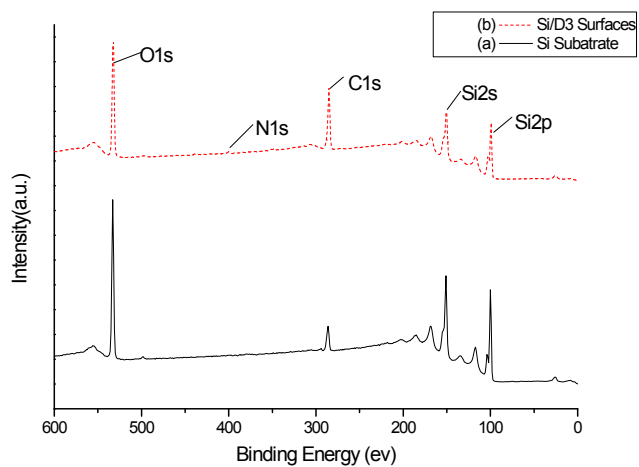
**Fig. S5.** The  $C_{1s}$  spectra of Si/D2 surface



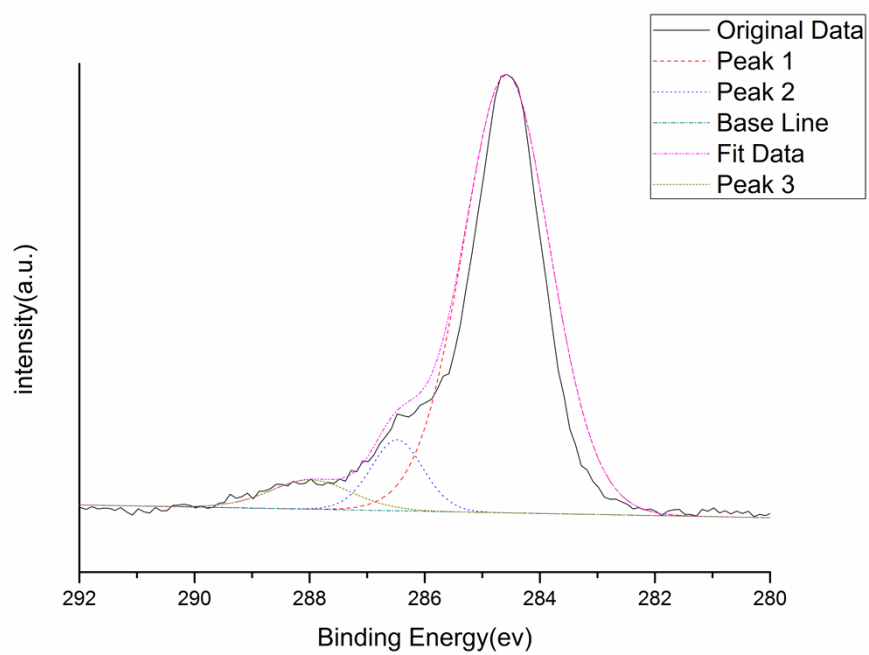
**Fig. S6.** The  $N_{1s}$  spectra of Si/D2 surface



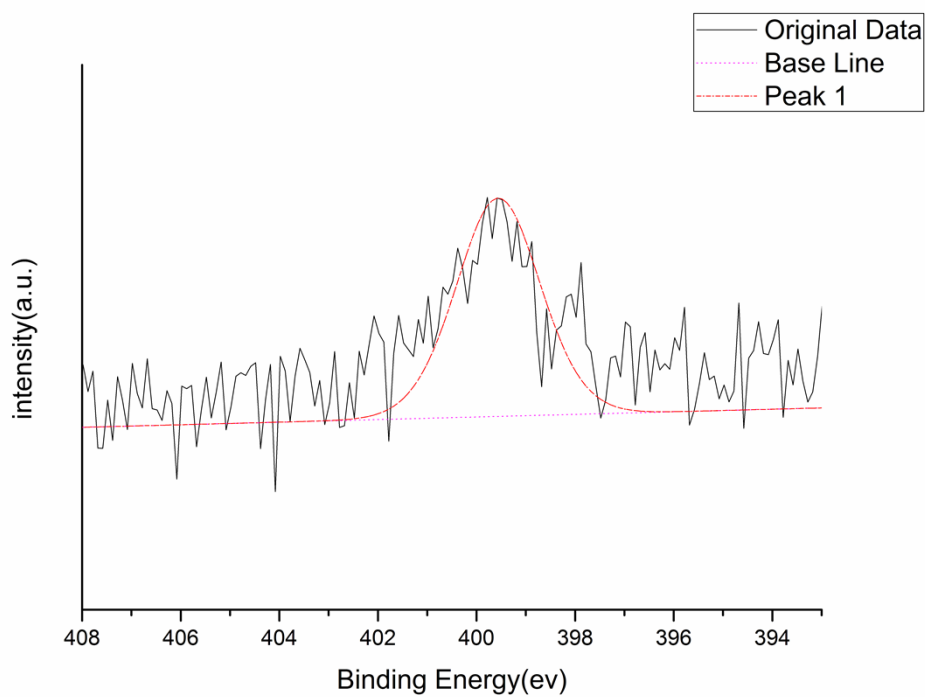
**Fig. S7.** The  $O_{1s}$  spectra of Si/D2 surface



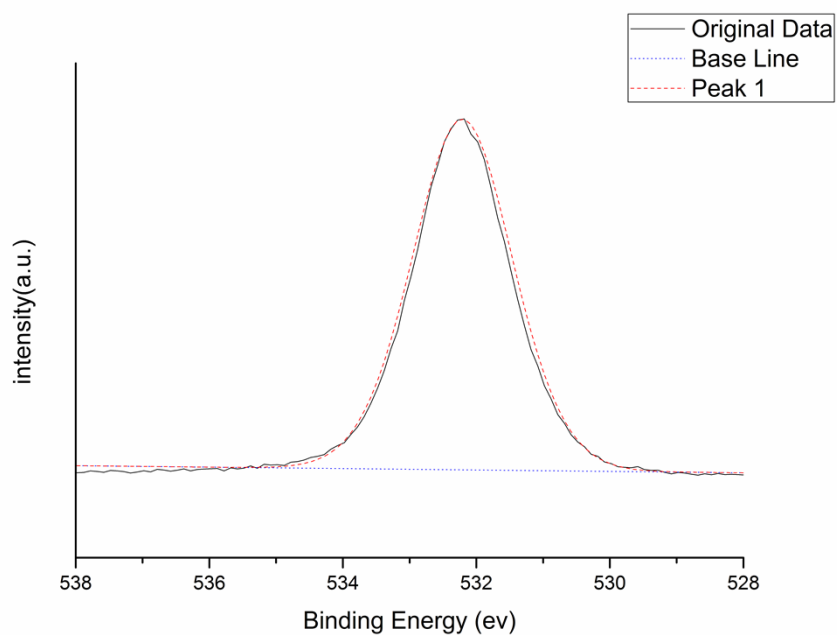
**Fig. S8.** The XPS scan spectra of Si/D3



**Fig. S9.** The  $C_{1s}$  spectra of Si/D3 surface



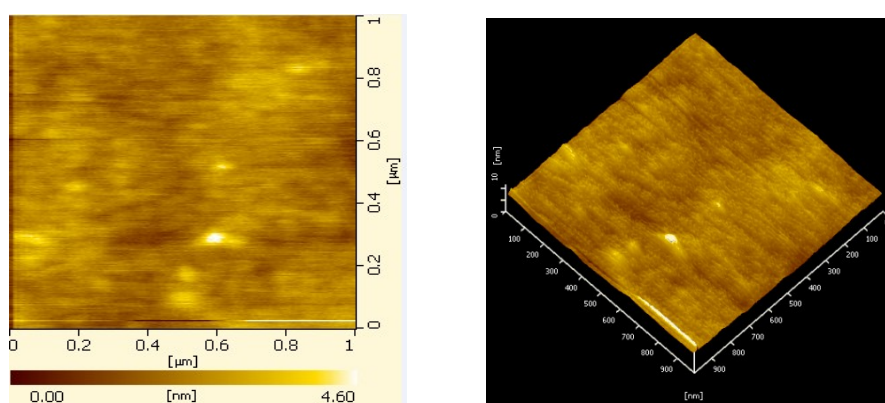
**Fig. S10.** The  $N_{1s}$  spectra of Si/D3 surface



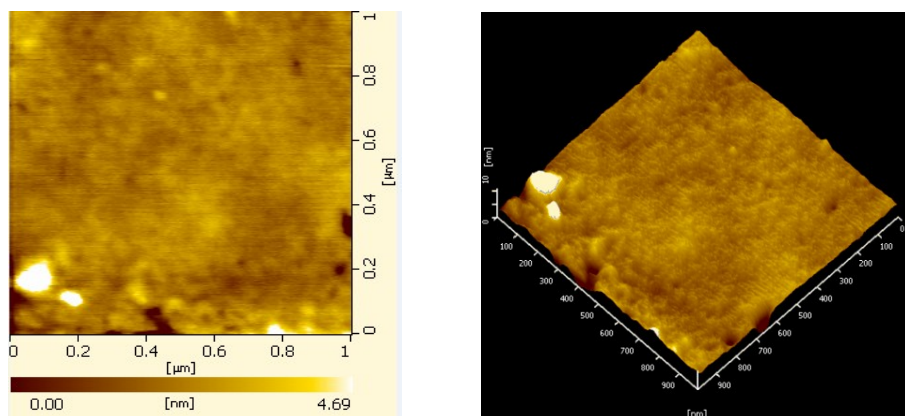
**Fig. S11.** The O<sub>1s</sub> spectra of Si/D3 surface

#### 4. 2D and 3D AFM images of Si substrate, Si/D2 and Si/D3

Fig. S12 and S13 showed two-dimensional and three-dimensional surface topography images of Si/D2 and Si/D3.



**Fig. S12.** 2D and 3D AFM images of Si/D2



**Fig. S13.** 2D and 3D AFM images of Si/D3