## **Supplementary Information**

## Distinct interface behaviors of Ni(II) on graphene oxide and oxidized carbon nanotubes triggered by their different topological aggregations

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Number of Pages: 5 (including the cover page) Number of Figures: 7 Number of Tables: 1 Table S1 The ligand exchange processes and free energy change ( $\Delta G$ , in kcal/mol) with solvent effect taken into accounted.

 $GO-X(H_2O) + Ni(OH)_{4-m}(H_2O)_m \rightarrow GO-X \cdot Ni(OH)_{4-m}(H_2O)_{m-n} + (n+1)H_2O$ (m=2,4;n=0-2)

Groups	Acidic		Alkaline	
	OSC	ISC	OSC	ISC
Edge carbonyl	-5.10	-9.76	-5.14	-14.25
Edge carboxylate	2.34	23.04	-13.75	-11.95
Edge hydroxyl	16.13	25.89	-1.57	-3.28
In-plane hydroxyl	-5.45	-3.03	-11.60	-15.60
In-plane epoxy	0.15	9.98	-1.62	3.16



Figure S1 DLS experiments of a) GO and b) OCNTs in the NaClO<sub>4</sub> solutions with variable concentrations.



Figure S2 AFM images of the GO, a) and c) initial GO, b) and d) GO with a loading of 30.4 mg/L Ni(II), in which light regions represent thick sheets, and dark regions represent the thin flake.



Figure S3 AFM measurements of the GO. a) and b) initial GO with an average thickness of 0.77 nm, c) and d) GO with a loading of 30.4 mg/g Ni(II).



Figure S4 SEM images of a) initial GO, b) GO with a loading of 30.4 mg/g Ni(II).



Figure S5 XPS analysis of C 1s in initial GO.



Figure S6 The correlation of GO hydrodynamic size and Ni(II) loadings at  $T = 298 \pm 1$  K, pH 6.5

 $\pm$  1, *m*/*V*=2.0 g/L, and *I* = 1.0 × 10<sup>-2</sup> mol/L NaClO<sub>4</sub>).



Figure S7 The sorption/release of Ni(II) on a) GO and b) OCNT ( $T = 298 \pm 1$  K, pH 6.5  $\pm 0.1$ , m/V

= 2.0 g/L, and  $I = 1.0 \times 10^{-2}$  mol/L NaClO<sub>4</sub>).