## Ultrafine Nickel Nanocatalyst-engineering Organic Layered Double Hydroxide towards Super-efficiently Fire-safe Epoxy Resin via Interfacial Catalysis

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Fig S1 XRD spectra of LDH-DBS and LDH-DBS@Ni(OH)<sub>2</sub> at small angle



**Fig S2** (a) and (b) TEM images of LDH-DBS at different magnification; (c) Mg mapping of LDH-DBS and (d) HAADF image with S mapping of LDH-DBS



Fig S3 (a) and (b) SEM images of LDH-DBS and LDH-DBS@Ni(OH)<sub>2</sub>; (c) EDS spectra with inset element percentage



Fig S4 (a) TG and (b) DTG curves of LDH-DBS and LDH-DBS@Ni(OH) $_2$ 



**Fig S5** (a) XRD patterns of EP and EP nanocomposites; (b) schematic illustration of intercalation process



**Fig S6** (a) THR; (b) SPR; (c) COP and (d) CO2P profile of EP, EP/3LDH-NO<sub>3</sub>, EP/3LDH-DBS and EP/3LDH-DBS@Ni(OH)<sub>2</sub>



**Fig S7** (a-1) front view and (a-2) top view of EP; (b-1) front view and (b-2) top view of EP/3LDH-NO<sub>3</sub>; SEM images of (a-3) interior surface and (a-4) exterior surface of EP; SEM images of (b-3) interior surface and (b-4) exterior surface of EP/3LDH-NO<sub>3</sub>



**Fig S8** (a-1) front view and (a-2) top view of EP/3LDH-DBS; (b-1) front view and (b-2) top view of EP/3LDH-DBS@Ni(OH)<sub>2</sub>; SEM images of (a-3) interior surface and (a-4) exterior surface of EP/3LDH-DBS; SEM images of (b-3) interior surface and (b-4) exterior surface of EP/3LDH-DBS@Ni(OH)<sub>2</sub>



Fig S9 Vertical section of char from (a) EP/3LDH-DBS and (b) EP/3LDH-DBS@Ni(OH)<sub>2</sub>; transverse section of char from (c) EP/3LDH-DBS and (d) EP/3LDH-DBS@Ni(OH)<sub>2</sub>



**Fig S10** High-magnification SEM image of interior char from (a) EP/LDH-DBS and (b) EP/3LDH-DBS@Ni(OH)<sub>2</sub>



 Fig
 S11
 (a)
 TG
 curves
 of
 EP,
 EP/3LDH-DBS,
 EP/3LDH-DBS@Ni(OH)2
 and

 calculated
 EP/3LDH-DBS@Ni(OH)2;
 (b)
 difference
 between
 experimental
 and

 calculated
 value



Fig S12 (a) top view and (b) front view of char from neat EP after CCT at 157s



**Fig S13** SEM images of interior surface of char from (a), (c) EP/3LDH-DBS and (b) (d) EP/3LDH-DBS@Ni(OH)<sub>2</sub>; exterior surface of char from (e), (g) EP/3LDH-DBS and (f) (g) EP/3LDH-DBS@Ni(OH)<sub>2</sub>



**Fig S14** Digital images of vertical section of char in CCT at 157s of (a) EP/3LDH-DBS and EP/3LDH-DBS@Ni(OH)<sub>2</sub>



Fig S15 Top, middle and bottom temperatures of EP at CCT



**Fig S16** FTIR spectra of evolved volatiles at the maximum degradation rate of (a) EP, (b) EP/3LDH-DBS and (c) EP/3LDH-DBS@Ni(OH)<sub>2</sub>



Fig S17 Evolution of (a) phenol compounds, (b) carbonyl compounds, (c) doublebond compounds



Fig S18 MS results of EP and EP/3LDH-DBS@Ni(OH)<sub>2</sub> at 15.7min



**Fig S19** (a) GC results of EP and EP/10LDH-DBS@Ni(OH)<sub>2</sub>; (b) MS results at  $35min \text{ of } EP/10LDH-DBS@Ni(OH)_2$