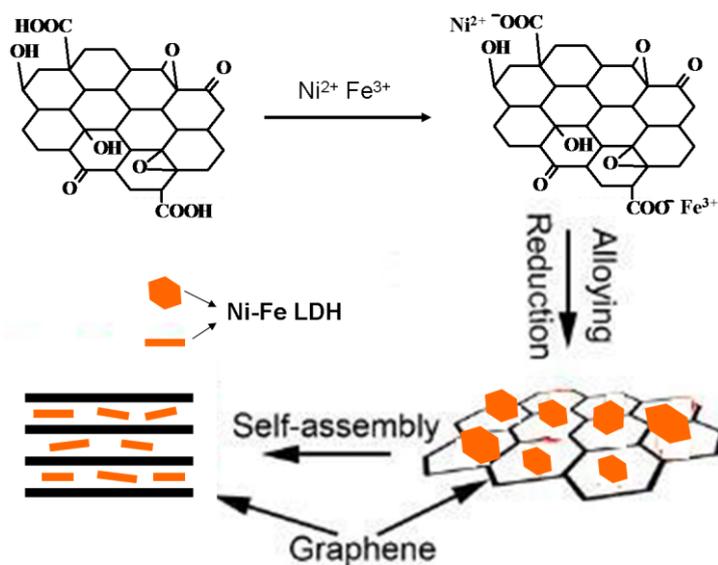


## Supplementary Information

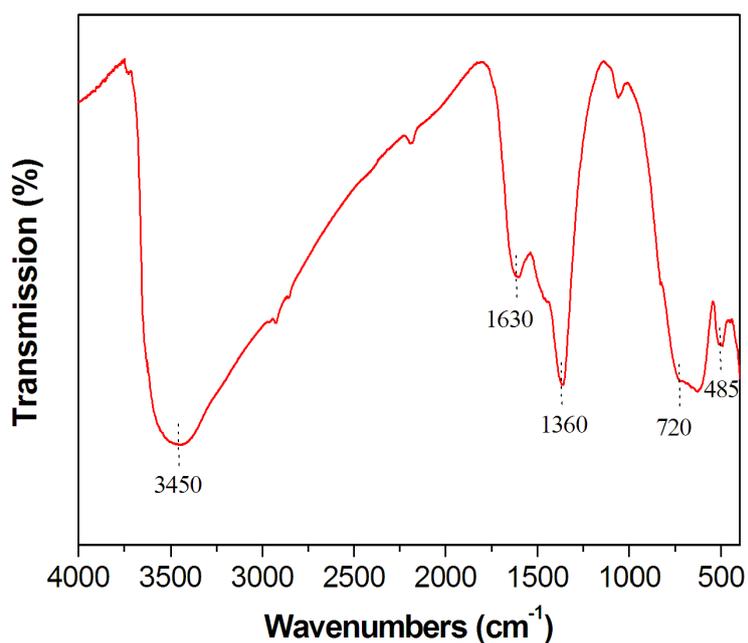
The formation mechanism of Ni-Fe layered double hydroxide/graphene hybrids is schematically illustrated in Fig. S1. The formation process is depicted as follows: first, the graphite oxide is sufficiently exfoliated into graphene oxide by sonication; second, the  $\text{Ni}^{2+}$  and  $\text{Fe}^{3+}$  are attached onto the negatively charged graphene oxide by electrostatic attraction; third, the graphene oxide is reduced to graphene; finally, graphene sheets loaded with the metal ions ( $\text{Ni}^{2+}$  and  $\text{Fe}^{3+}$ ) are self-assembled into layered Ni-Fe/graphene hybrid nanostructure due to the hydrophobic nature of graphene.



**Fig. S1.** Schematic illustration of the formation mechanism of Ni-Fe LDH/GNS.

To further confirm the metal hydroxide (M-OH) bonds, FTIR spectrum of Ni-Fe LDH/GNS was measured, as shown in Fig. S2. It can be observed that the broad peak centered at  $3450\text{ cm}^{-1}$  corresponds to the -OH stretching vibration of water molecules

in the interlayer and H-bonded OH group, accompanied with the bending mode at 1630  $\text{cm}^{-1}$ [1,2]. The intense peaks at 1360  $\text{cm}^{-1}$  are ascribed to the  $\nu_3$  vibration and bending modes of  $\text{CO}_3^{2-}$ . Other absorption bands at 720 and 485  $\text{cm}^{-1}$  are attributed to metal hydroxide (M-OH) stretching and bending modes in the brucite-like lattice [2]. All above observations confirm that Ni-Fe LDH has been made.



**Fig. S2.** FTIR spectrum of Ni-Fe LDH/GNS.

[1] G. Abellan, E. Coronado, C. Marti-Gastaldo, E. Pinilla-Cienfuegos, and A. Ribera, *J. Mater. Chem.*, 2010, **20**, 7451.

[2] L.J. Zhang, X.G. Zhanga, L.F. Shen, B. Gao, L. Hao, X.J. Lu, F. Zhang, B. Ding, and C.Z. Yuan, *J. Power Sources*, 2012, **199**, 395.