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 Ni$^{2+}$ and 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 (Ni$^{2+}$ and Fe$^{3+}$) are self-assembled into layered Ni-Fe/graphene hybrid nanostructure due to the hydrophobic nature of graphene.

![Schematic illustration of the formation mechanism of Ni-Fe LDH/GNS.](image)

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 cm$^{-1}$ corresponds to the -OH stretching vibration of water molecules.
in the interlayer and H-bonded OH group, companied with the bending mode at 1630 cm\(^{-1}\)[1,2]. The intense peaks at 1360 cm\(^{-1}\) are ascribed to the \(\nu_3\) vibration and bending modes of \(\text{CO}_3^{2-}\). Other absorption bands at 720 and 485 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.

![FTIR spectrum of Ni-Fe LDH/GNS](image)

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