Electronic Supporting Information

Graphene oxide conformally wrapped FeOOH/graphite composite anode for a lithium-ion battery

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1.1. Material characterizations

X-ray diffraction (XRD) measurements were carried out for the structural analysis using PANalytical X'pert Pro MPD diffractometer equipped with a $Cu\,K\alpha$ radiation source (wavelength Kα1 = 1.540598 Å and Kα2 = 1.544426 Å) operated at 40 kV and 30 mA at a scan rate of 0.0334225° 2 θ s^{-1} with a 2θ angle of 20–70° (Center for University-wide Research Facilities (CURF) at Jeonbuk National University). The chemical compositions of the anode were analyzed via X-ray photoelectron spectroscopy (XPS), which were performed on a PHI Quantera II spectrometer using a monochromatic AlKα X-ray source (Junbuk National University) and the binding energy was calibrated by the adventitious carbon peak of C 1s at 284.8 eV. Fourier transform infrared (FT-IR) spectra were obtained using an FT-IR spectrometer (CURF at Jeonbuk National University) in the range of 400–4000 cm^{-1}. The morphological features were observed using high-resolution field-emission scanning electron microscopy (HR FE-SEM, Hitachi SU8230, Korean Basic Science Institute). The microstructure images and elemental distribution results were collected using high-resolution transmission electron microscopy (HR-TEM) and scanning transmission electron microscopy-energy dispersive X-ray analysis (STEM-EDS, CURF at Jeonbuk National University). Semi-quantitative analysis (SQX) was conducted at Chungbuk National University.

1.2. Cell assembly

The electrochemical performance of the prepared anode material was tested by the CR2032-type coin cell. The slurry for each electrode was prepared with a weight ratio of 7:2:1 and was composed of an active material, super P and polyvinylidene fluoride in NMP. The slurry was cast on Cu foil. Each electrode had a loading level of 1.47–1.67 mg/cm^2. All cast electrodes were dried at 100 °C for 4 h. Subsequently, the electrodes were punched into a disk shape with a diameter of 11 mm and then subjected to drying in a vacuum oven at 100 ºC for 12 h. The half-cells were then fabricated in a dry glove box filled with a high-purity Ar gas environment. A polypropylene (celgard 2400) separator was used along with lithium metal as the counter/reference electrode. The electrolyte consists of a 1 M $LiPF_6$ in a 1:1 volume ratio of ethylene carbonate (EC) and diethyl carbonate (DEC).

1.3. Electrochemical measurements

The cycling performance of the cells was conducted via the galvanostatic discharge–charge cycling between 0.01–3.0 V (*vs.* Li/Li^+) using battery cycling test equipment

(WBCS3000L, WonATech, Korea). The cyclic voltammetry (CV) of the cells was obtained varying the voltage sweep rate from 0.1 to 1.0 mV/s in the voltage range between 2.0 V–5 mV. Electrochemical impedance spectroscopy (EIS) was performed at an amplitude of 10 mV in the frequency range of 1 MHz to 10 mHz using impedance analyzer (Ivium CompactStat potentiostat) at open-circuit voltage. All the electrochemical tests were performed at room temperature.

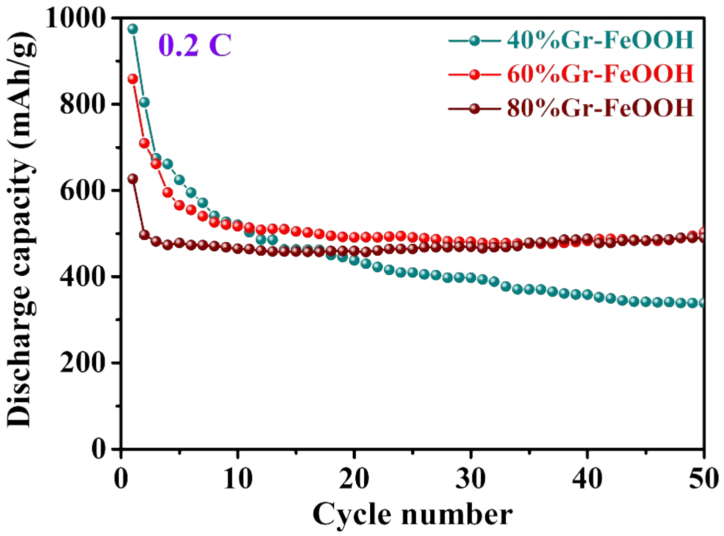


Fig. S1. Cycle performance at 0.2 C of 40%Gr-FeOOH, 60%Gr-FeOOH and 80%Gr-FeOOH samples.

We synthesized Gr-FeOOH with varying amounts of graphite. Initially, 0.9 g (40 wt.%), 2.0 g (60 wt.%) and 5.3 g (80 wt.%) of graphite powder were dispersed in deionized water separately. Then, the hydrothermal was conducted as described in the experimental section. The corresponding Gr-FeOOH variants are named as 40%Gr-FeOOH, 60%Gr-FeOOH and 80%Gr-FeOOH, respectively. Based on the optimized parameter of 60%Gr-FeOOH, it was subsequently referred to as Gr-FeOOH for convenience.

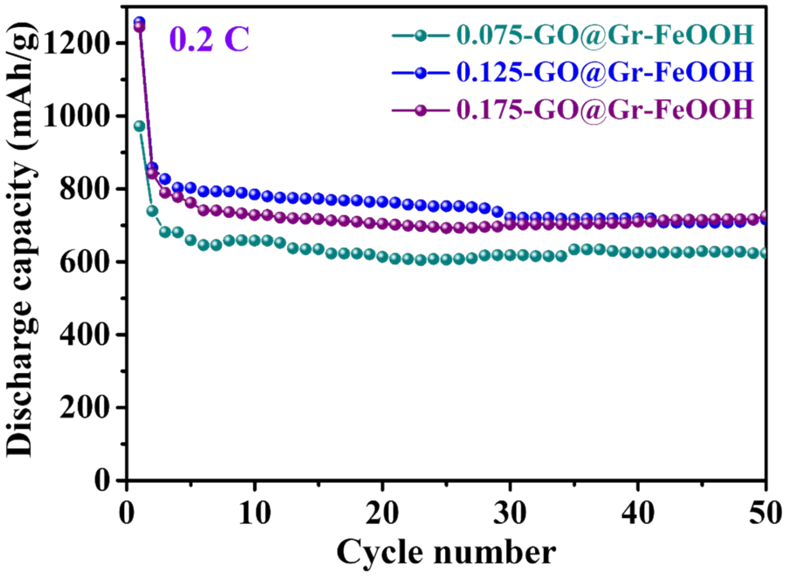


Fig. S2. Cycle performance at 0.2 C of 0.075-GO@Gr-FeOOH, 0.125-GO@Gr-FeOOH and 0.175-GO@Gr-FeOOH samples.

The obtained Gr-FeOOH sample was mixed with various quantities of GO to optimize the GO. 0.075 g GO, 0.125 g GO and 0.175 g of GO were individually mixed with Gr-FeOOH. The resulting final products were designated as 0.075-GO@Gr-FeOOH, 0.125-GO@Gr-FeOOH and 0.175-GO@Gr-FeOOH, respectively. The 0.125-GO@Gr-FeOOH sample showed enhanced capacity; thus, it was further denoted as GO@Gr-FeOOH.

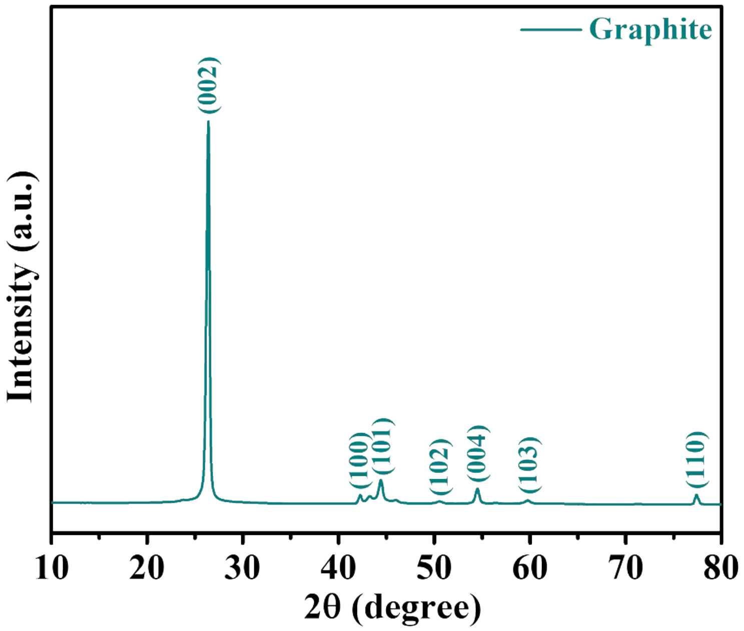


Fig. S3. XRD pattern of commercial graphite sample.

As depicted in Fig. S3, graphite exhibited XRD peaks at 26.4, 42.2, 44.4, 50.4, 54.5, 59.9 and 77.7° corresponding to (002), (100), (101), (102), (004), (103) and (110) planes, respectively (JCPDS No. 34-1266). The small peak appeared at 43.2° related with turbostratic band of disordered carbon materials [1].

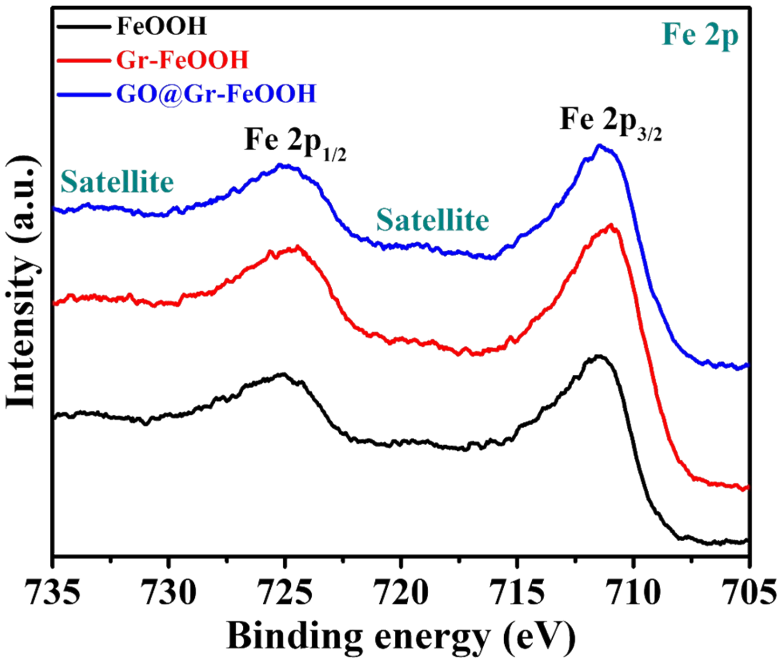


Fig. S4. XPS spectra of Fe 2p of FeOOH, Gr-FeOOH and GO@Gr-FeOOH samples.

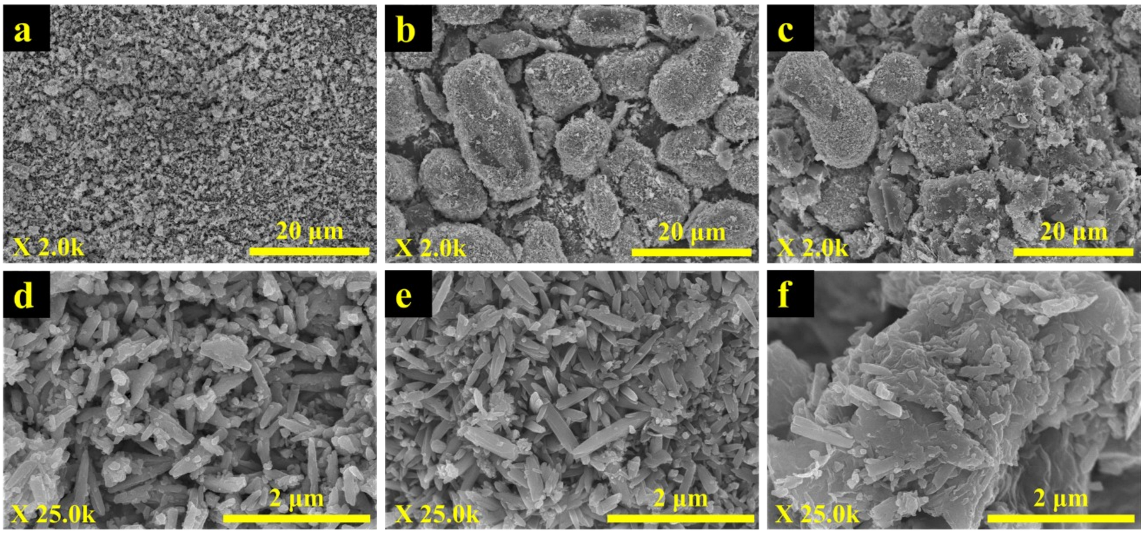


Fig. S5. HR FE-SEM top view images of (a and d) FeOOH, (b and e) Gr-FeOOH and (c and f) GO@Gr-FeOOH samples.

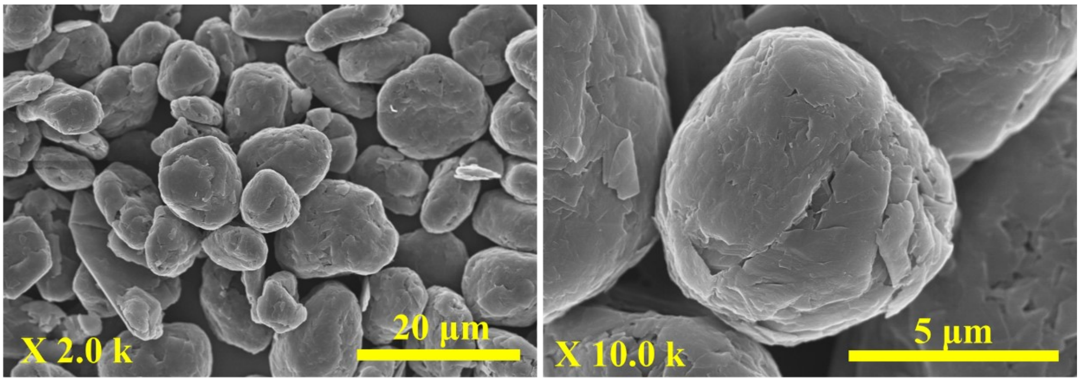


Fig. S6. HR FE-SEM top view images of commercial graphite sample.

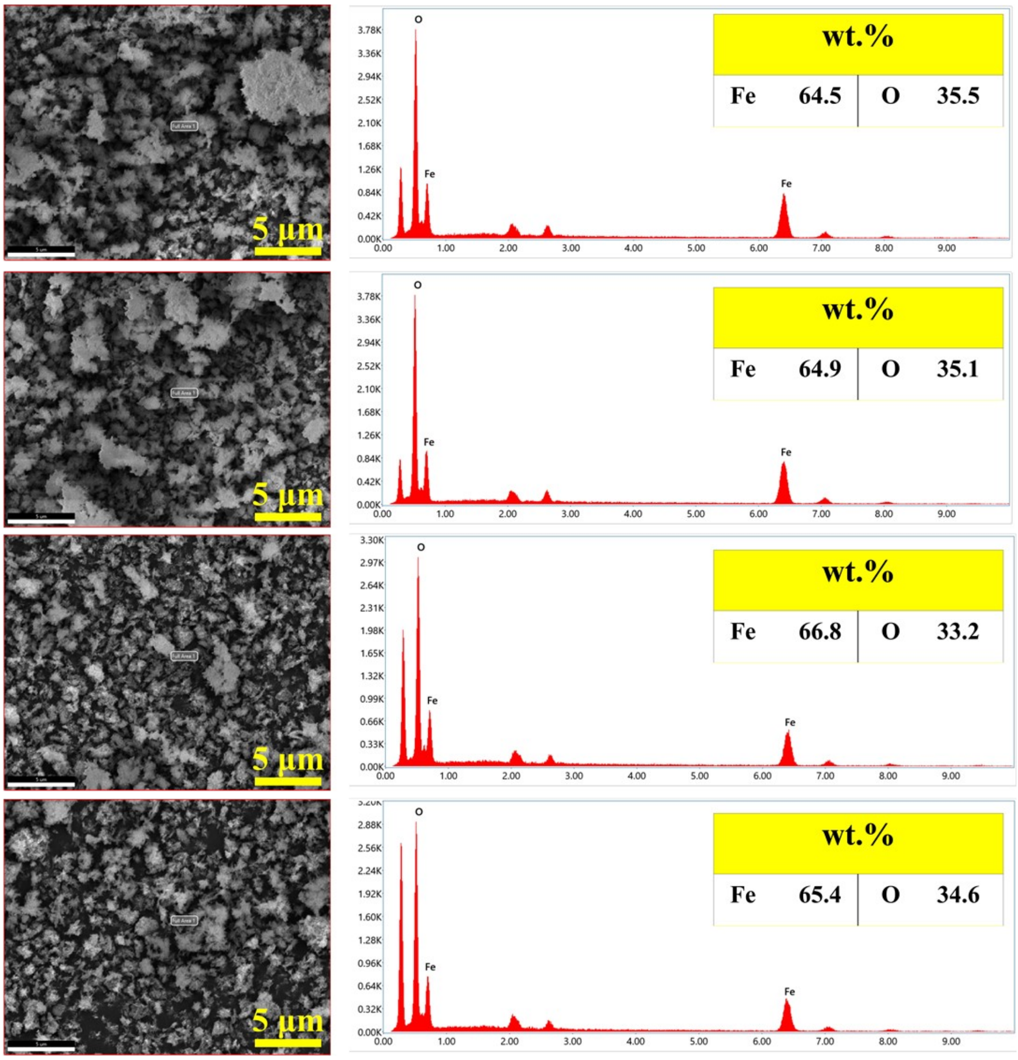
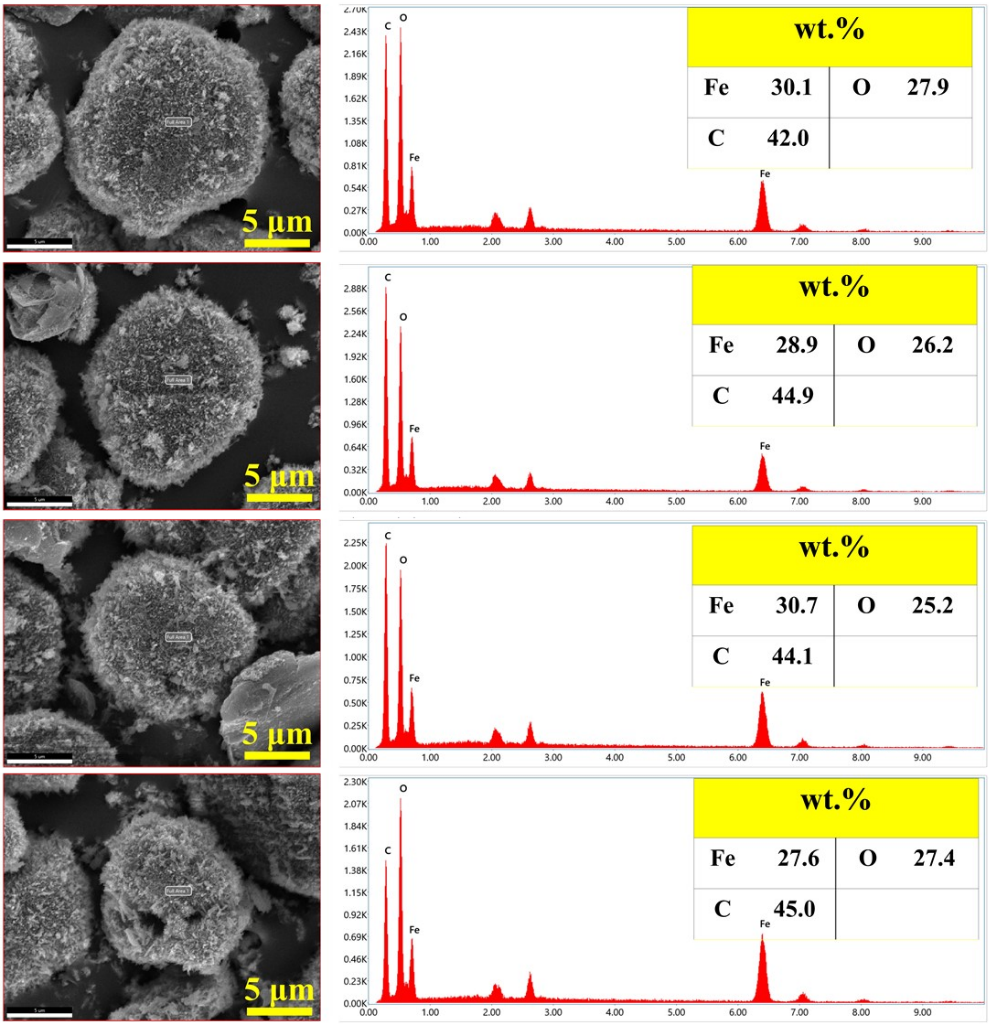


Fig. S7. SEM-EDS spectrum of FeOOH sample.



wt.%			
Fe	30.1	O	27.9
C	42.0		

wt.%			
Fe	28.9	O	26.2
C	44.9		

wt.%			
Fe	30.7	O	25.2
C	44.1		

wt.%			
Fe	27.6	O	27.4
C	45.0		

Fig. S8. SEM-EDS spectrum of Gr-FeOOH sample.

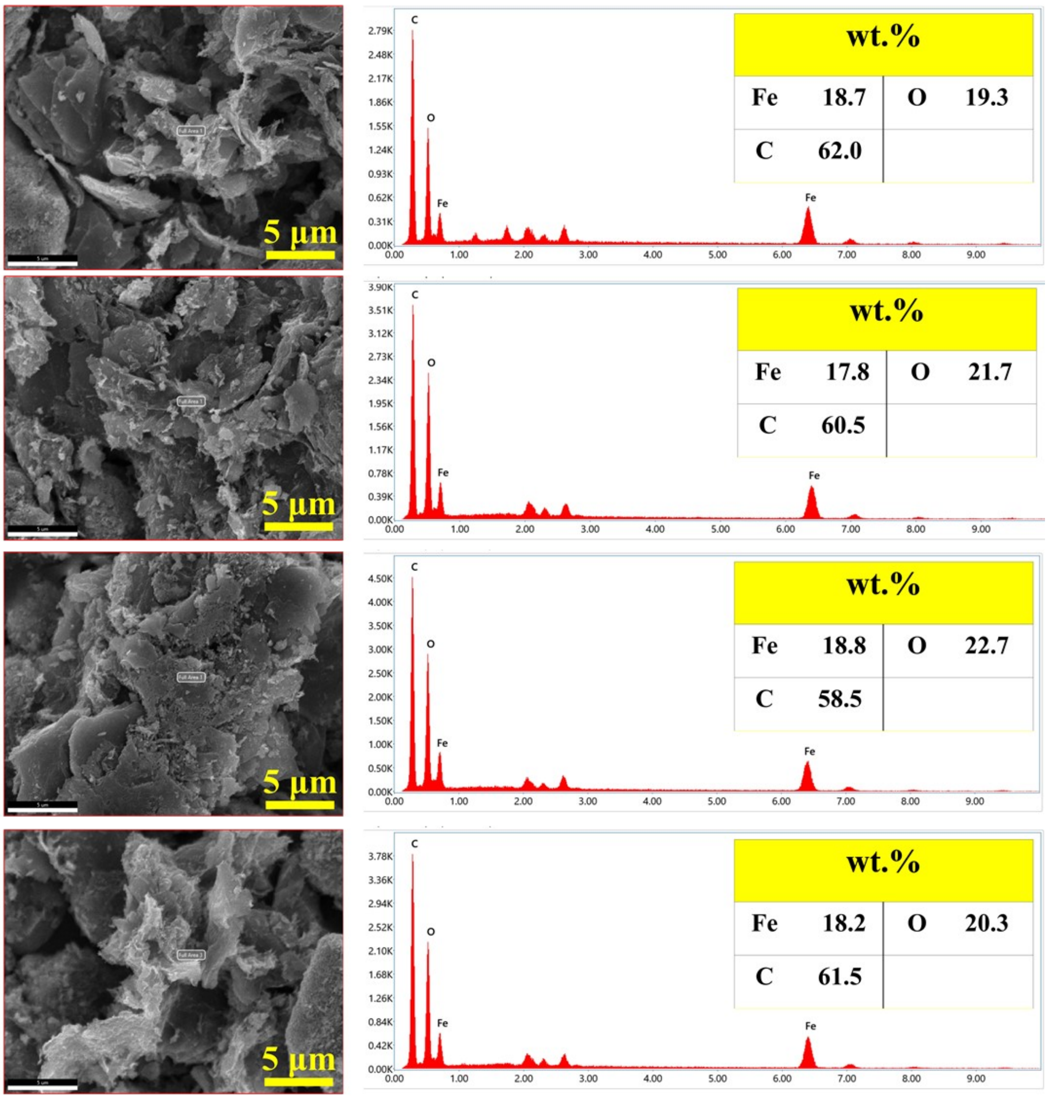


Fig. S9. SEM-EDS spectrum of GO@Gr-FeOOH sample.

The SEM-EDS analysis was performed at various regions and the average weight percent (wt.%) was determined (Table S1).

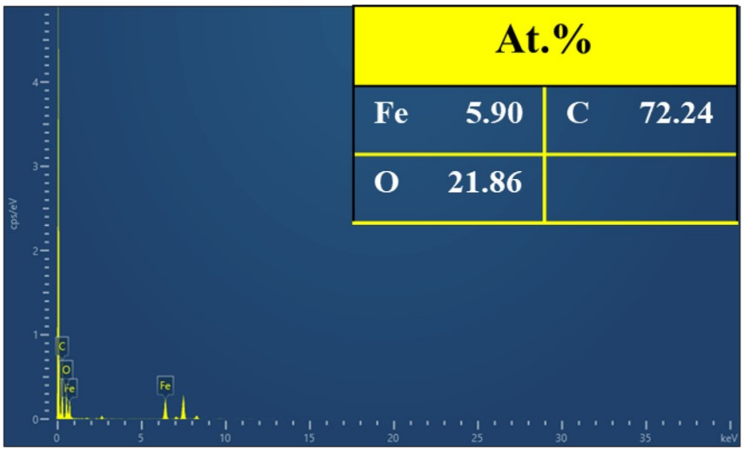
Table S1. The SEM-EDS average elemental compositions of FeOOH, Gr-FeOOH and GO@Gr-FeOOH samples.

Sample	C (wt.%)	Fe (wt.%)	O (wt.%)
FeOOH	-	65.4	34.6
Gr-FeOOH	44.0	29.3	26.7
GO@Gr-FeOOH	60.6	18.4	21.0

Table S2. SQX calculation results of FeOOH, Gr-FeOOH and GO@Gr-FeOOH samples.

Sample	C (wt.%)	Fe (wt.%)	O (wt.%)
FeOOH	-	71.7	28.3
Gr-FeOOH	38.9	35.5	25.5
GO@Gr-FeOOH	58.9	22.1	19.0

Na and Cl concentrations were below the detection limit of the instrument.



At.%			
Fe	5.90	C	72.24
O	21.86		

Fig. S10. EDS elemental spectrum of GO@Gr-FeOOH samples.

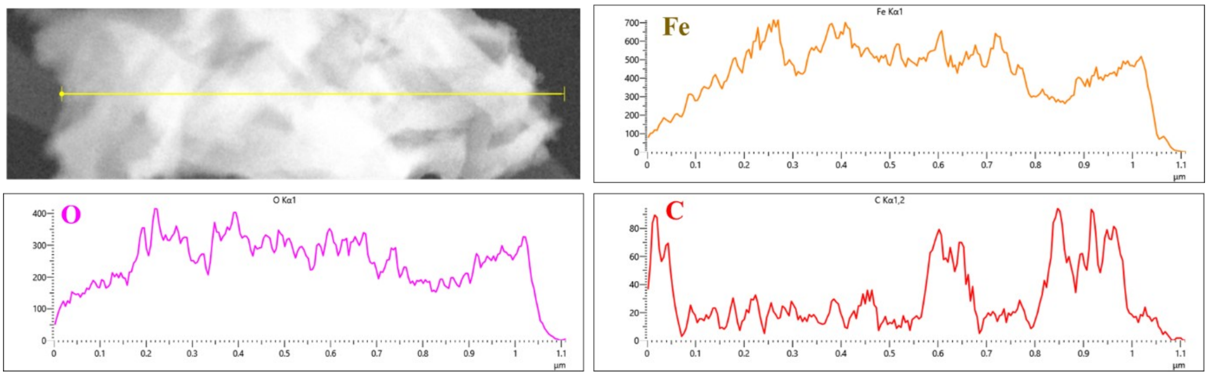
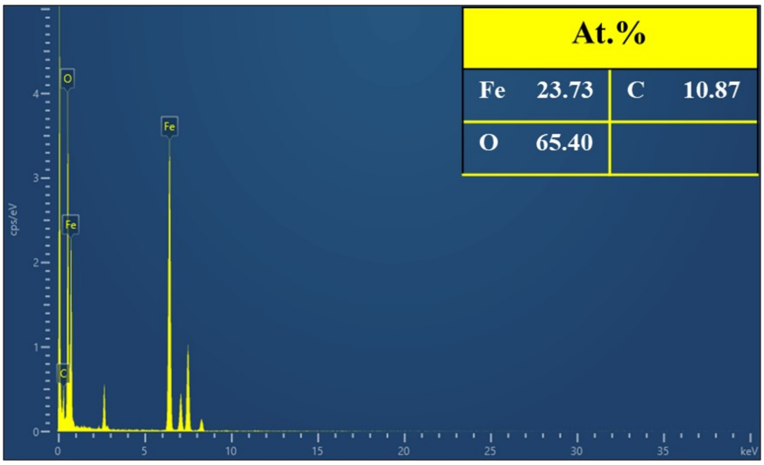


Fig. S11. EDS line scanning of GO@Gr-FeOOH samples.



At.%			
Fe	23.73	C	10.87
O	65.40		

Fig. S12. EDS line scanning elemental spectrum of GO@Gr-FeOOH samples.

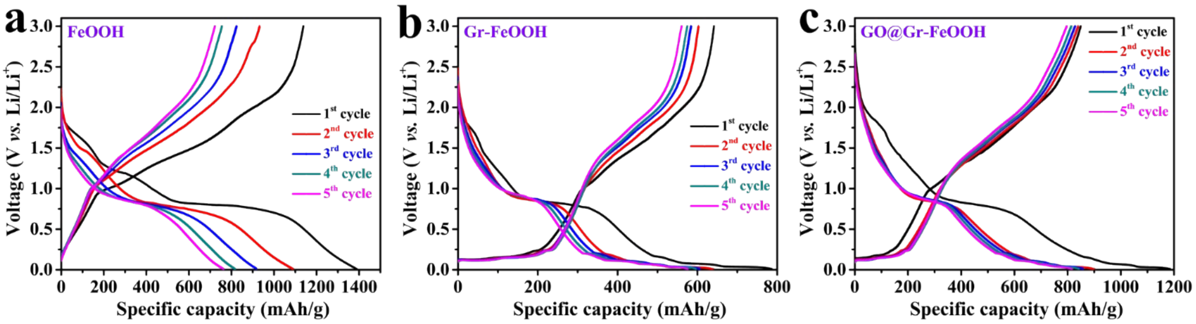


Fig. S13. Discharge-charge profiles of initial five cycles at 0.1 C for (a) FeOOH, (b) Gr-FeOOH and (c) GO@Gr-FeOOH samples.

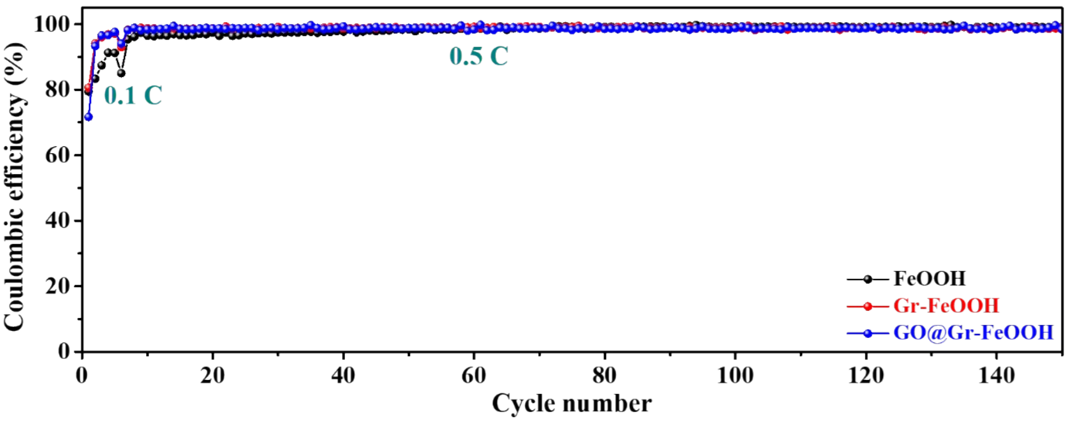


Fig. S14. Coulombic efficiencies of FeOOH, Gr-FeOOH and GO@Gr-FeOOH samples.

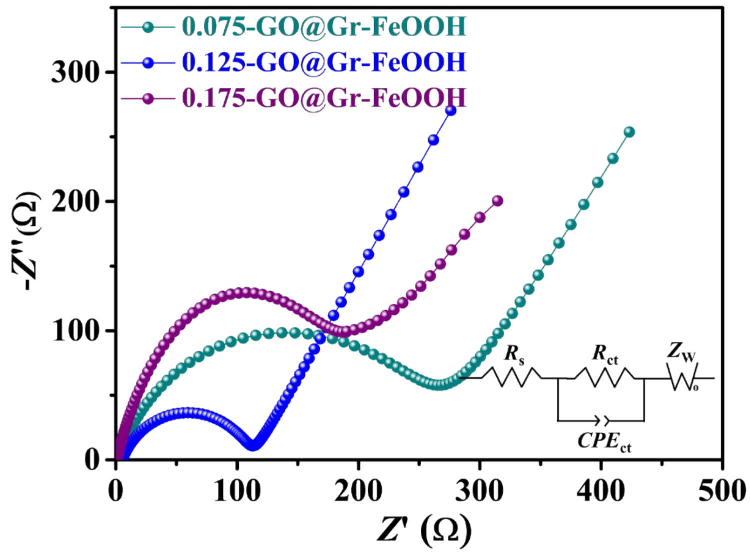


Fig. S15. EIS spectra before cycling of 0.075-GO@Gr-FeOOH, 0.125-GO@Gr-FeOOH and 0.175-GO@Gr-FeOOH samples (inset: corresponding equivalent circuit).

Table S3. Electrochemical impedance spectroscopy fitting parameters for the 0.075-GO@Gr-FeOOH, 0.125-GO@Gr-FeOOH and 0.175-GO@Gr-FeOOH.

Sample	R_S (Ω)	R_{ct} (Ω)
0.075-GO@Gr-FeOOH	2.6	273.0
0.125-GO@Gr-FeOOH	5.3	91.0
0.175-GO@Gr-FeOOH	2.7	156.8

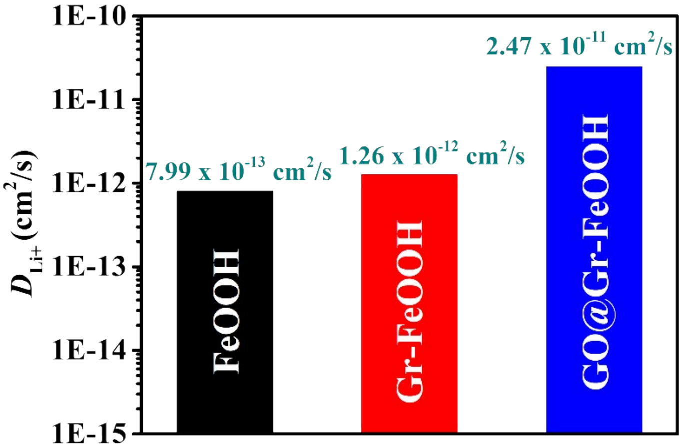


Fig. S16. Li$^+$ diffusion coefficients of FeOOH, Gr-FeOOH and GO@Gr-FeOOH samples.

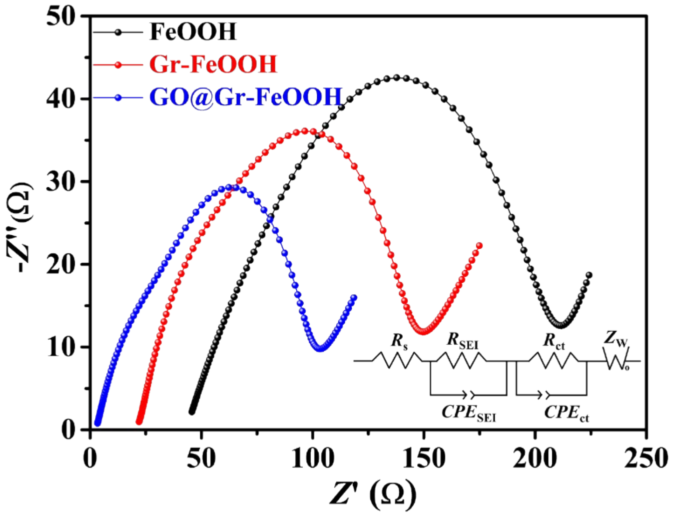


Fig. S17. EIS spectra of FeOOH, Gr-FeOOH and GO@Gr-FeOOH after 150-cycle stability test (inset: corresponding equivalent circuit).

Table S4. Electrochemical impedance spectroscopy fitting parameters for the FeOOH, Gr-FeOOH and GO@Gr-FeOOH after 150-cycle stability test.

Sample	R_S (Ω)	R_{SEI} (Ω)	R_{ct} (Ω)
FeOOH	43.0	83.6	80.1
Gr-FeOOH	29.7	53.0	57.8
GO@Gr-FeOOH	12.1	31.3	39.2

Reference

1. N. M. S. Hidayah, W. W. Liu, C. W. Lai, N. Z. Noriman, C. S. Khe, U. Hashim and H. C. Lee, *AIP Conf. Proc.*, 2017, **1892,** 150002.