Supporting Information Iron nanoparticle templates for constructing 3D graphene frame-work with enhanced performance in sodium-ion batteries

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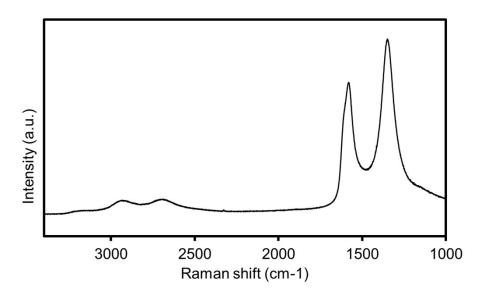
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1. Materials

Graphite (SP-10) was purchased from BAY CARBON Inc. KMnO4, H_2SO_4 , 30% aq. H_2O_2 , hydrazine hydrate, HCl were purchased from Wako Pure Chemical Industries, Ltd. Iron(III) Sulfate n-hydrate (Fe₂(SO₄)₃·nH₂O) was purchased from KANTO CHEMICAL CO., INC. 1 M sodium hexafluorophosphate (NaPF₆), dissolved in 1/1 (V/V) ethylene carbonate (EC)/diethyl carbonate (DEC) and 1 M lithium hexafluorophosphate (LiPF₆), dissolved in 1/1 (V/V) ethylene carbonate (EC)/diethyl carbonate (DEC) were purchased from Kishida Chemical Co., Ltd. Sodium was purchased from Sigma Aldrich. Lithium was purchased from Honjo Metal Co., Ltd. All reagents were used directly without further purification.

2. Characterization instrumentation and experimentation

Freeze-dried of GO was performed by ADVANTEC DRZ350WC. XPS was measured by JPS-9030 with a pass energy of 20 eV. The crystalline structure of samples was characterized by X-ray diffraction (XRD) using a PANalytical Co. X' part PRO using Cu K α radiation (l= 1,541 Å) in the 2 θ range of 5–75°. The operating tube current and voltage were 30 mA and 40 kV, respectively. The data was collected at the step size of 0.017° and the type of scan was continuous. The morphology was measured by transmission electron microscopy (TEM) JEOL JEM-2100F and atomic force microscopy (AFM) SHIMADZU SPM-9700HT. Heating treatment was conducted using a tube furnace KTF055N1 from KOYO THERMO SYSTEMS.



3. Additional figure

Figure S1. Raman of graphene oxide prepared by our modified Hummers' method.

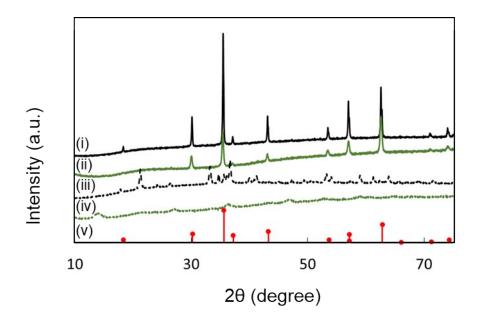


Figure S2. XRD patterns of (i) FeO_x from Urea, (ii) FeO_x from NH₃, (iii) Fe(OH)_x, (iv) Fe(OH)_x, (v) simulation of Fe₃O₄.

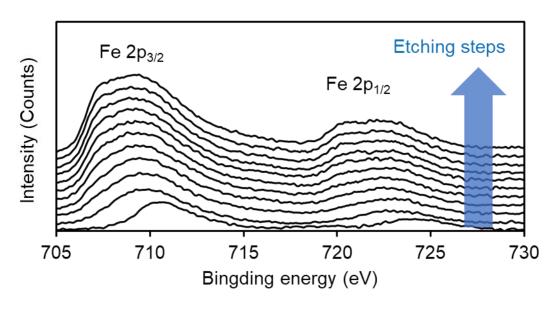


Figure S3. Iron sandwiched between GO layers was confirmed by XPS, with etching of surface layer by layer by Argon ion beam.

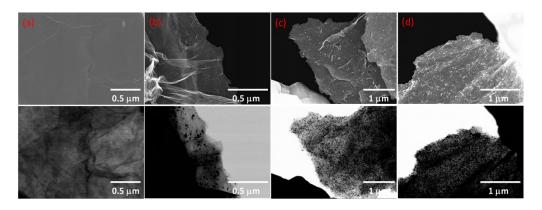


Figure S4. SEM (upper) and TEM (lower) images of rGO and iron oxide nano-particles with various amount of iron prepared in $600 \circ C$. (a) rGO-FeO_{0.5}, (b) rGO-Fe_{1.0}, (c) rGO-Fe_{1.5}, (d) rGO-Fe_{2.5}.

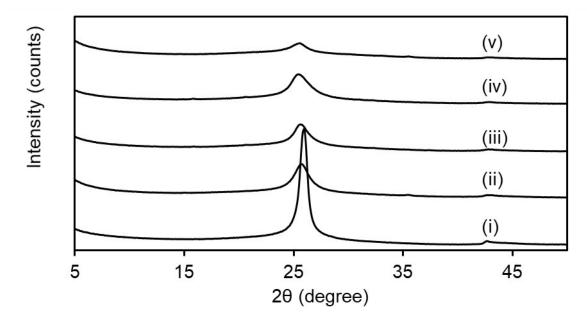
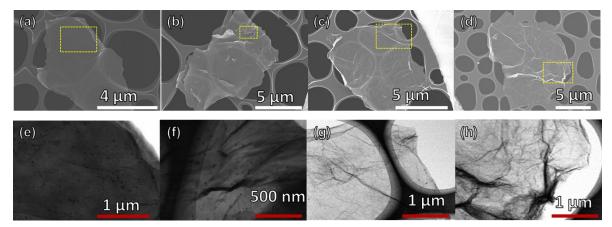


Figure S5. XRD of (i) rGO, (ii) $G_{0.5}$, (iii) $G_{1.0}$, (iv) $G_{1.5}$, (v) $G_{2.5}$.



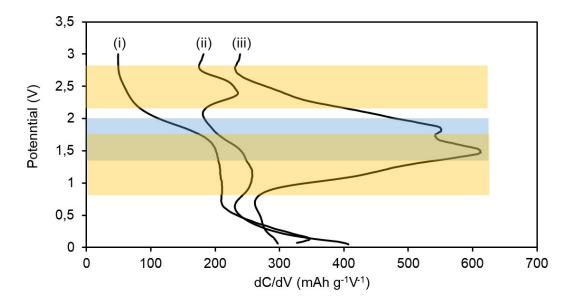


Figure S6. SEM (upper) and TEM (lower) images of G_x with various amount of iron. (a) G_{0.5}, (b) G_{1.0}, (c) G_{1.5}, (d) G_{2.5}.

Figure S7. Lithium-ion battery charge behavior after 5 cycle of (i) rGO, (ii) G_{2.5}, (iii) rGO-FeO_{2.5}. **Table S1.** SIBs performances of previously reported carbon host materials.

Sample name	Capacity	Current density	Electrode composition (wt.%)			References
	(mAh g⁻¹)	(mA g ⁻¹)	Active material	Carbon black	Binder	
S-doped porous carbon	440	50	80	10	10	1
S&N co-doped carbon	250	22	86	6	6	2
Porous graphene grids carbon	210	28	70	20	10	3
Porous graphene	195	50	100	-	-	4
CoFe2O4@3D- NG	260	50	80	10	10	5
Hard carbon-G	230	20	93	-	7	6
	289	20	80	10	10	
G _{2.5}	297	20	90	-	10	This work

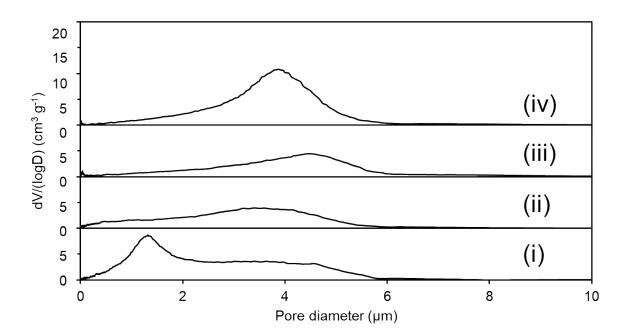


Figure S8. Mercury porosimeter results of (i) G_{0.5}, (i) G_{1.0}, (iii) G_{1.5}, (iv) G_{2.5}.

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