Supplementary Information

Catalytic Graphitization of Pyrolysis Oil for Anode Application in Lithium-ion Batteries

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Table S1. Crystallite size (L_c, L_a), interlayer separation (d_{002}), and Raman intensity ratio (I_D/I_G) of biographites prepared with different iron loading.

Iron loading	L_c (Å)	L_a (Å)	d_{002} (nm)	I_D/I_G
0.00 × FC	27	122	0.3422	0.98
0.25 × FC	48	238	0.3384	0.98
0.50 × FC	65	375	0.3359	0.99
0.75 × FC	89	428	0.3350	0.81
1.00 × FC	160	403	0.3364	0.62
1.25 × FC	134	532	0.3359	0.72
1.50 × FC	180	644	0.3353	0.41
1.75 × FC	179	706	0.3345	0.42
2.00 × FC	179	615	0.3352	0.38
2.25 × FC	240	370	0.3354	0.52
2.50 × FC	233	390	0.3347	0.20
2.75 × FC	218	416	0.3350	0.34
3.00 × FC	177	418	0.3355	0.43

Table S2. Distribution of elements in raw pyrolysis oil and biographite (2.50×FC iron loading) using XRF technique.

Element (%)	Pyrolysis oil	Biographite
C	99.817 (organics)	99.861
Al	0.009	0.027
Si	1.5 ppm	0.012
P	0.031	0.018
S	0.115	0.009
Cl	ND*	0.009
Ca	0.009	0.007
Ti	ND	5.5 (ppm)
Fe	1.7 (ppm)	0.049
Ni	ND	1.1 (ppm)
Cu	2.8 (ppm)	1.2 (ppm)
Zn	ND	0.3 (ppm)
Br	ND	5.1 (ppm)
Sn	0.002	0.003
Te	0.003	0.004
K	0.012	ND
Sb	0.001	ND
Hf	1.7 (ppm)	ND

*ND: Not Detected

Table S3. The characteristic (002) peak position of biographite and corresponding peak intensity at different temperatures (heating and cooling) calculated from the *in-situ* XRD patterns.

Temperature (°C)	(002) peak position (degrees)	(002) peak intensity (counts)***
H-600 °C*	-	-
H-700 °C	-	-
H-800 °C	-	-
H-900 °C	-	-
H-1000 °C	-	-
H-1100 °C	25.95	4584
H-1200 °C	26.03	5718
C-1100 C**	26.14	5979
C-1000 °C	26.19	6128
C-900 °C	26.22	6587
C-800 °C	26.27	6423
C-700 °C	26.27	6485

C-600 °C	26.32	6719
C-25 °C	26.37	6834

*H stands for heating stage
**C stands for cooling stage
***The (002) peak intensity values reported here included the background signal. Therefore, the actual intensities of these peaks were much smaller. The smaller peak intensity of biographite formed at 1100 °C during heating was clearly understood from **Figure S4d**.

Table S4. Biographite (2.50×FC iron loading) crystal parameters as a function of aging time (month).

No. of Exp.	Time (month)	L_c (Å)	d_{002} (nm)	I_{002}×1000* (counts)
1.	0	233	0.3347	330
2.	6	203	0.3350	18
3.	8	193	0.3349	25
4.	9	190	0.3353	21

*Intensity of characteristic (002) peak.

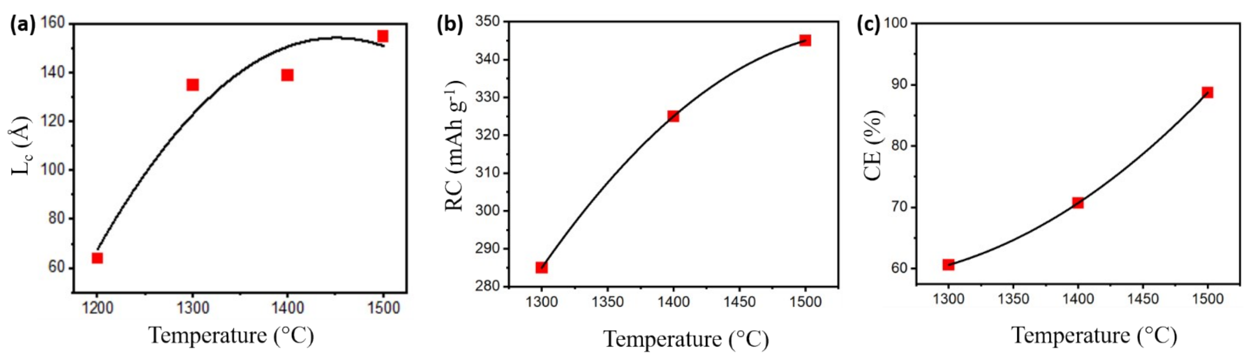

Figure S1. Effect of graphitization temperature on (a) crystallite size, (b) reversible capacity, and (c) Coulombic efficiency.

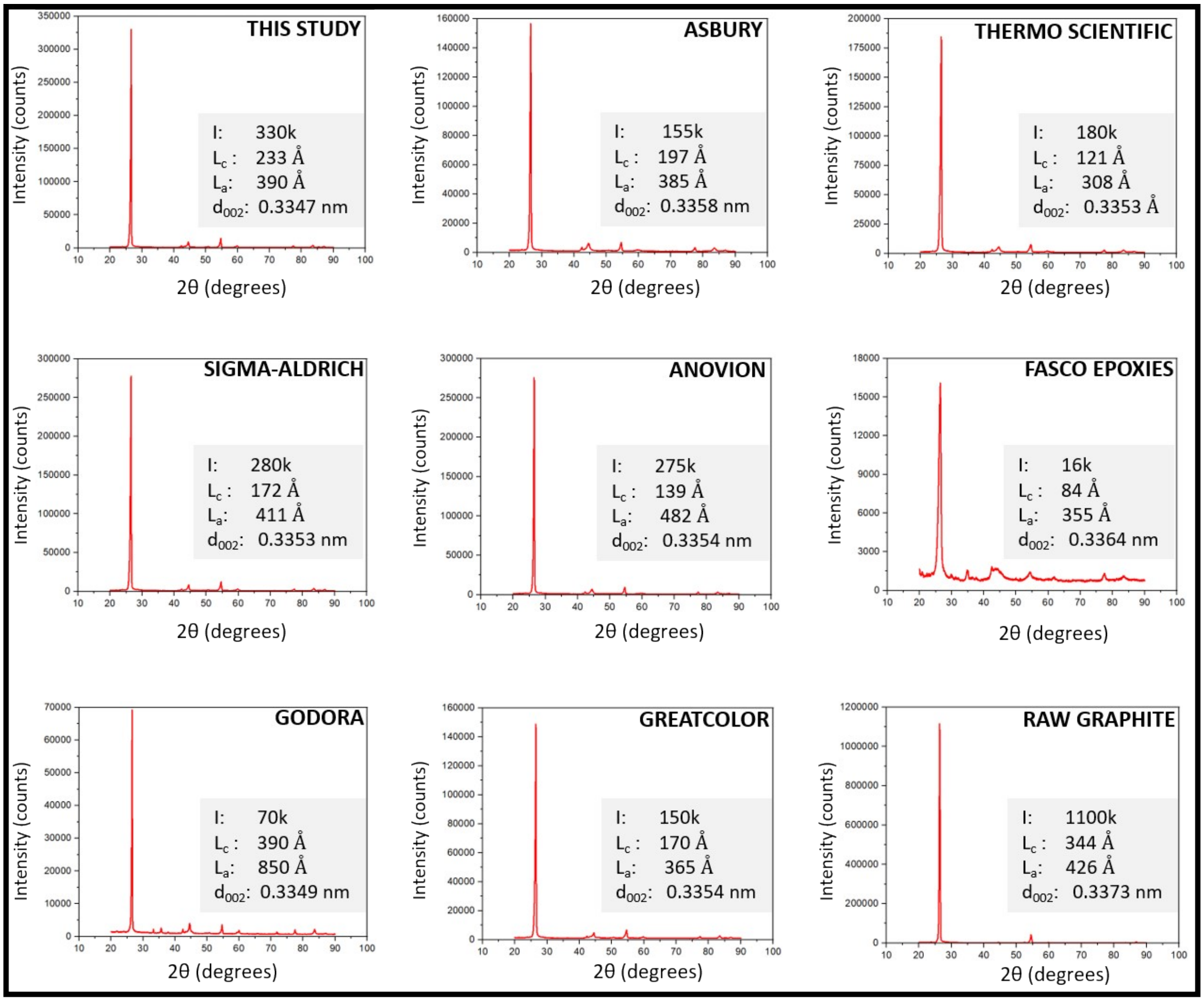

Figure S2. Comparison of the XRD pattern of optimal biographite (this study) with some commercial graphite samples. Intensity (I) of the characteristic (002) peak is highlighted (insets) along with other crystal parameters (L_c, L_a, and d_{002}) of graphite.

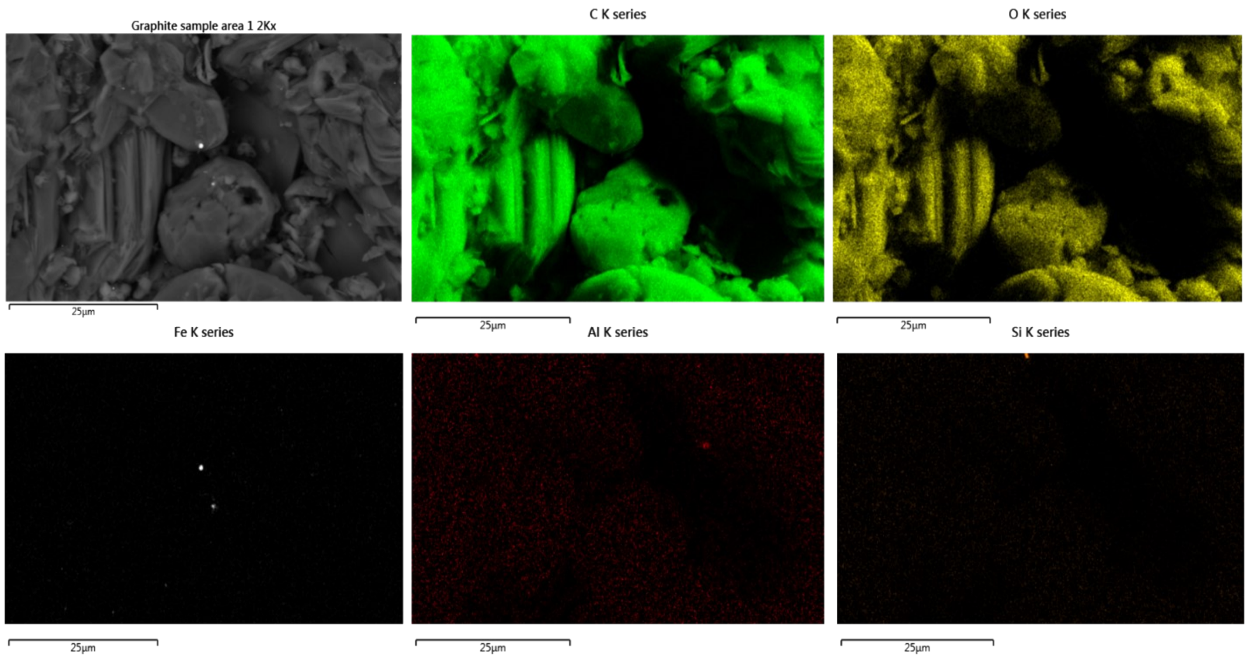

Figure S3. EDS elemental mapping of optimal biographite showing the presence of carbon (C), oxygen (O), iron (Fe), aluminum (Al), and silicon (Si).

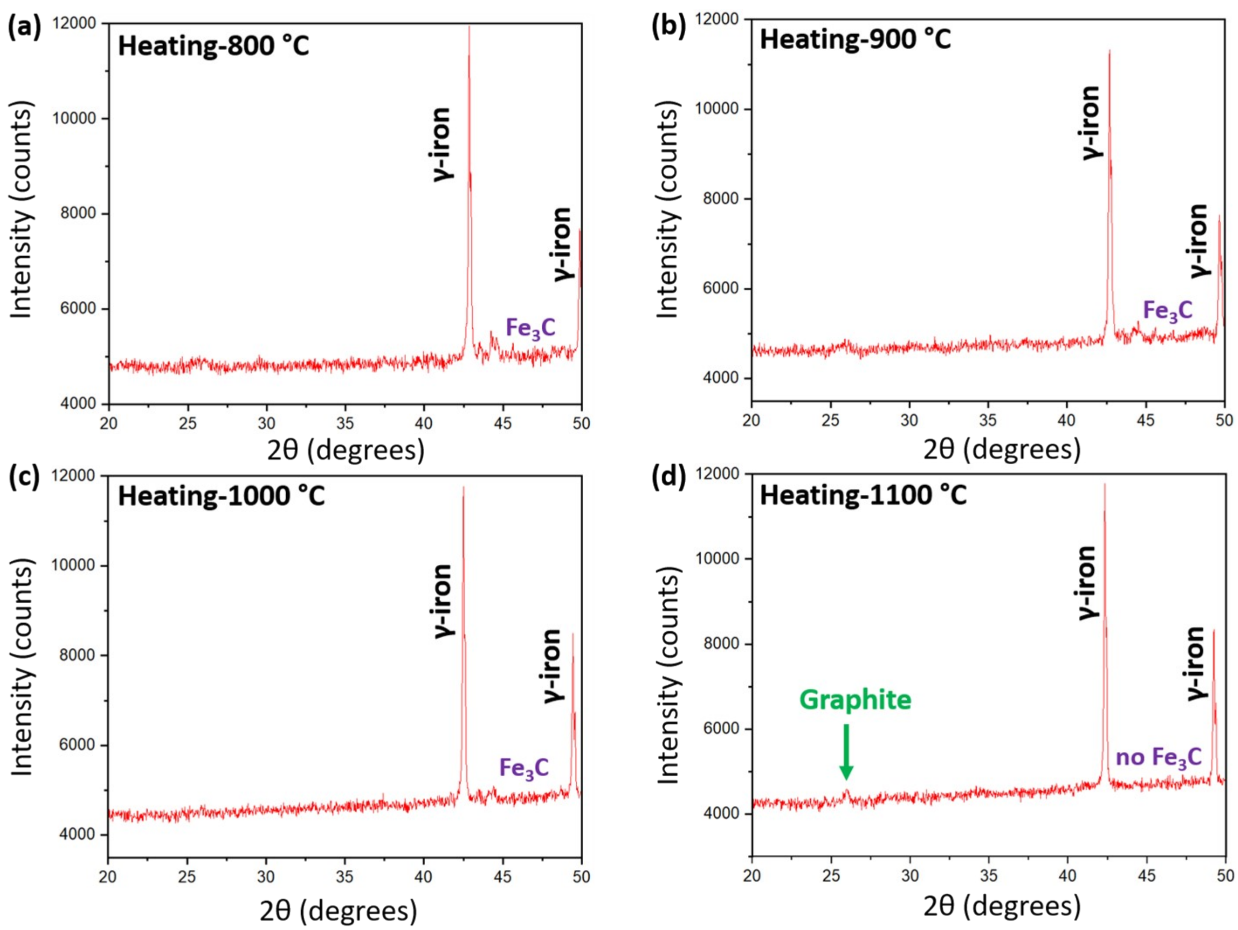

Figure S4. The *in-situ* XRD patterns recorded during heating at (a) 800, (b) 900, (c) 1000, and (d) 1100 °C respectively.

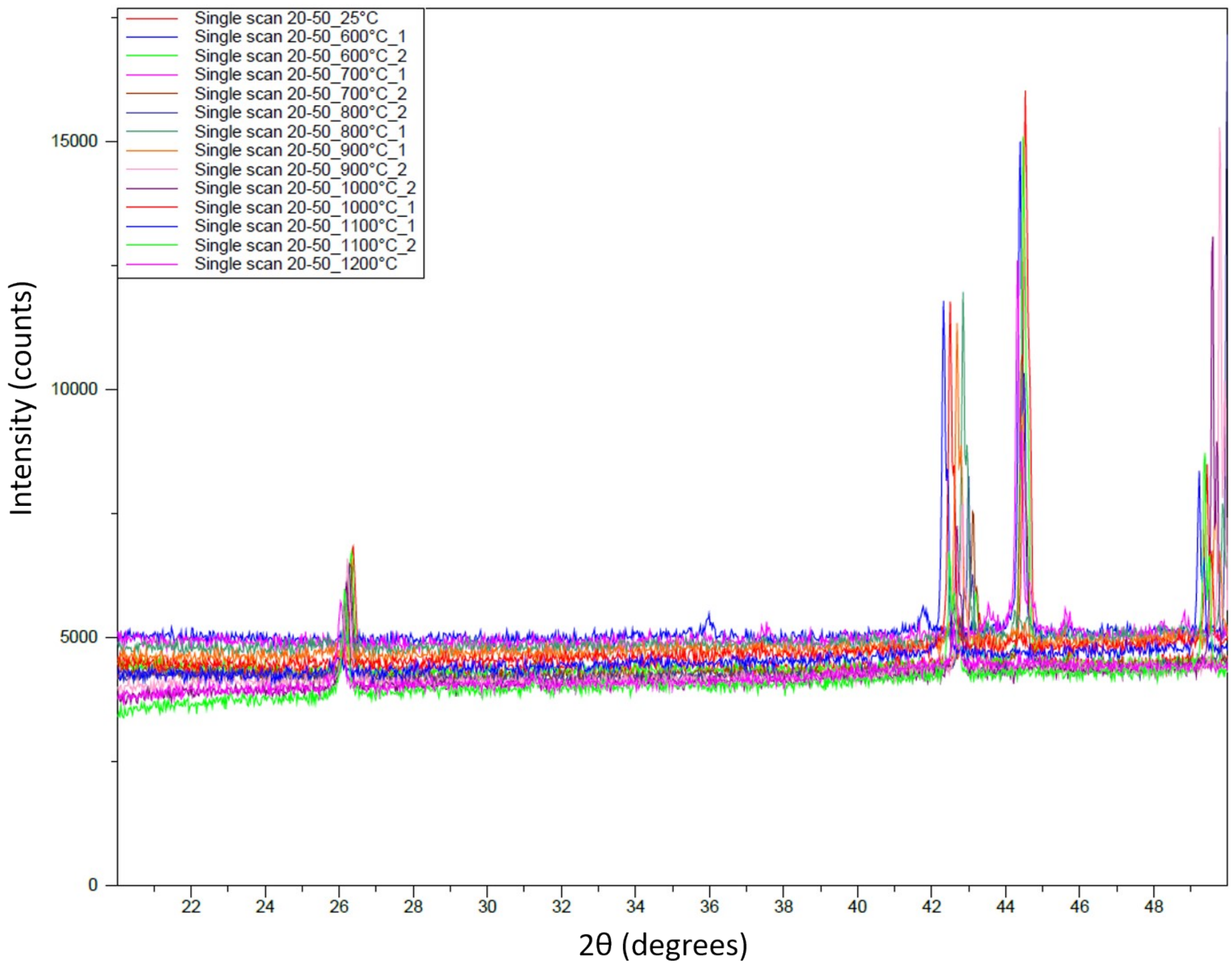

Figure S5. The overlay *in-situ* XRD patterns recorded during both heating and cooling.

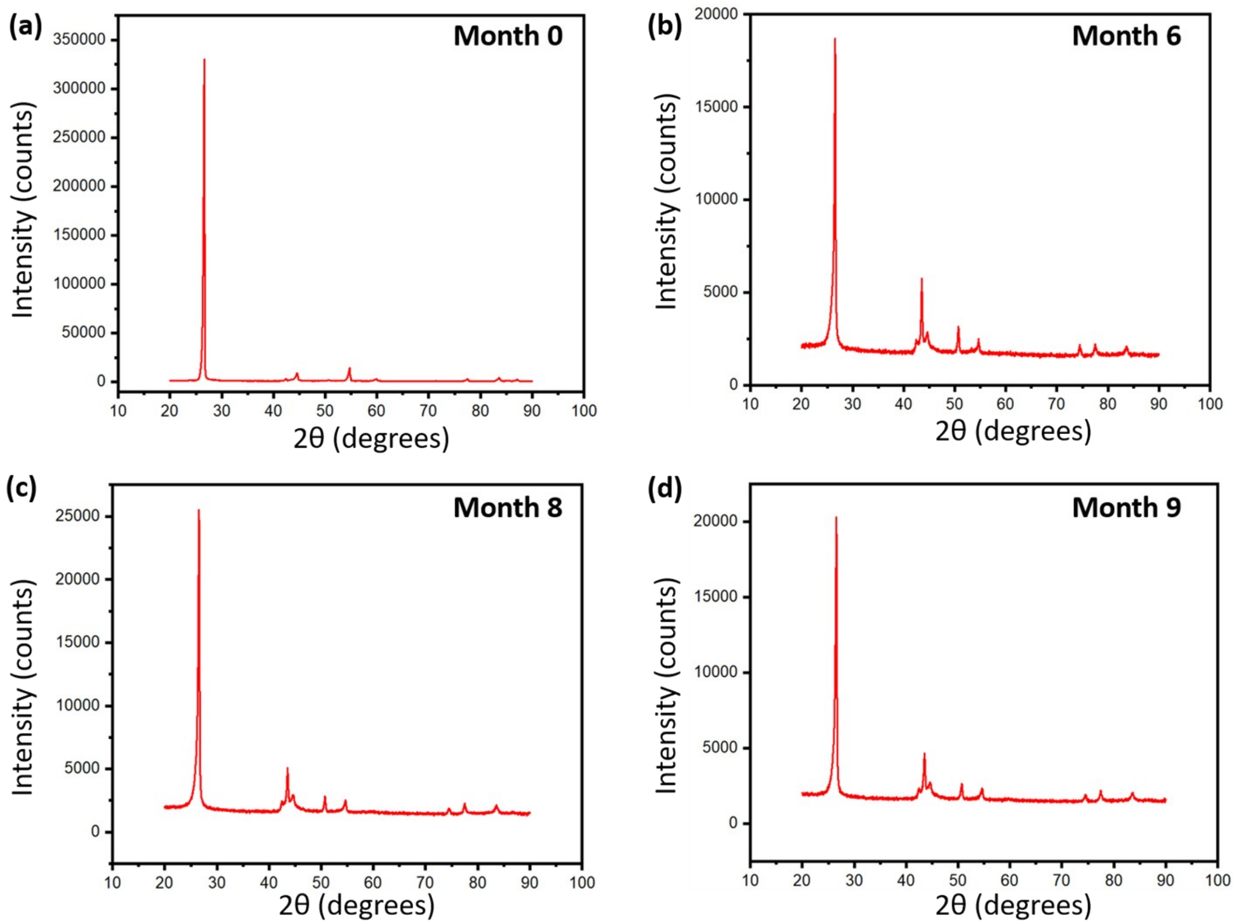

Figure S6. Effect of aging time on the XRD patterns of biographite (2.50×FC iron loading) at (a) month 0, (b) month 6, (c) month 8, and (d) month 9.