

## Supplementary Materials

# Building Polymeric Framework Layer for Stable Solid Electrolyte Interphase on Natural Graphite Anode

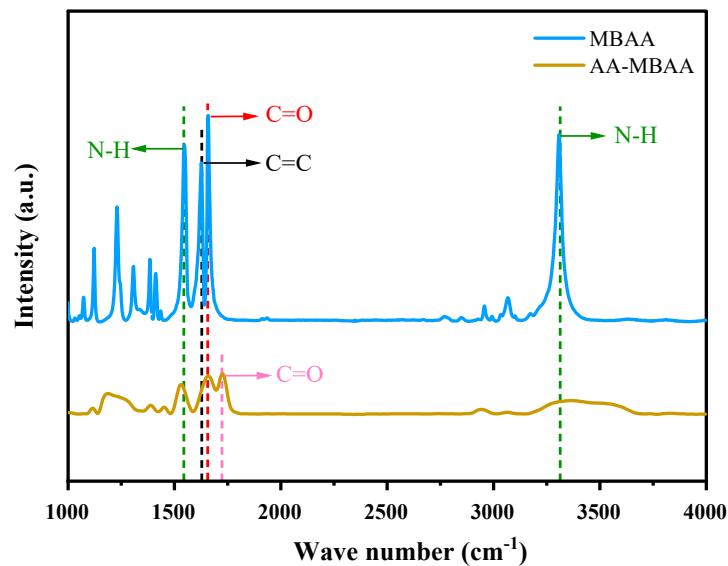
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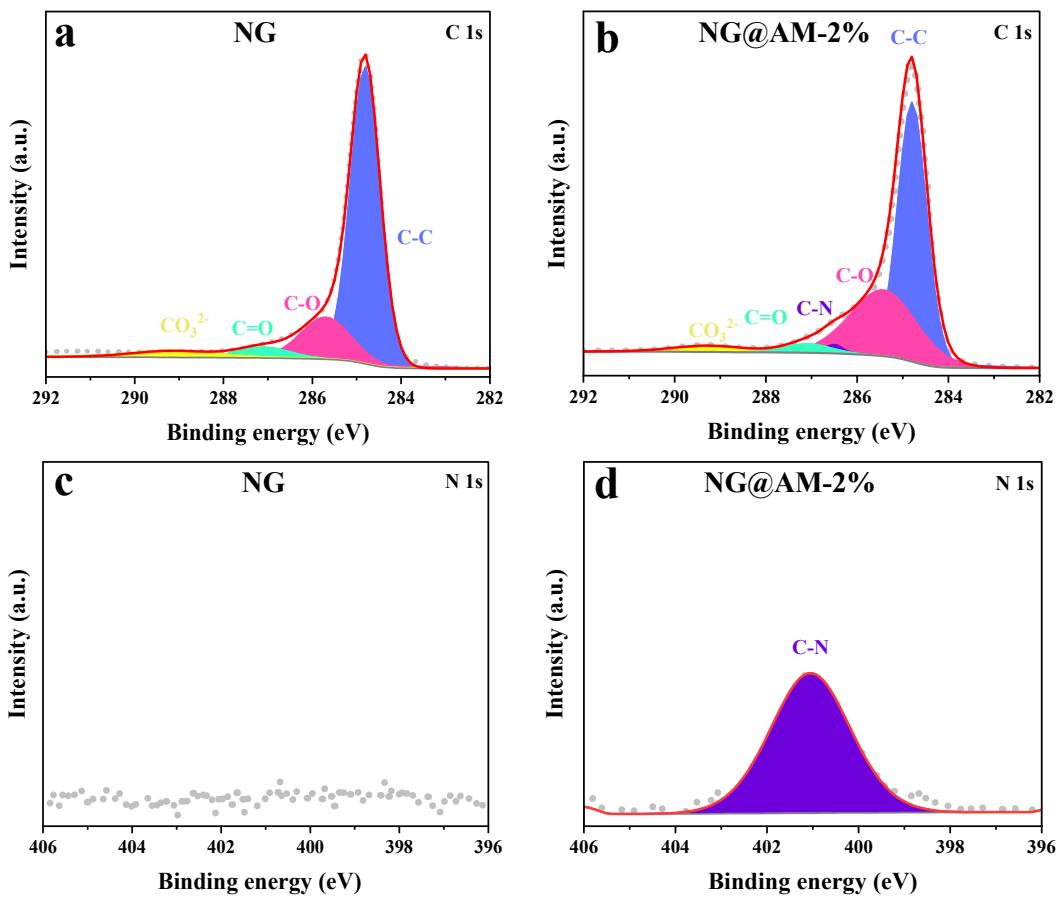
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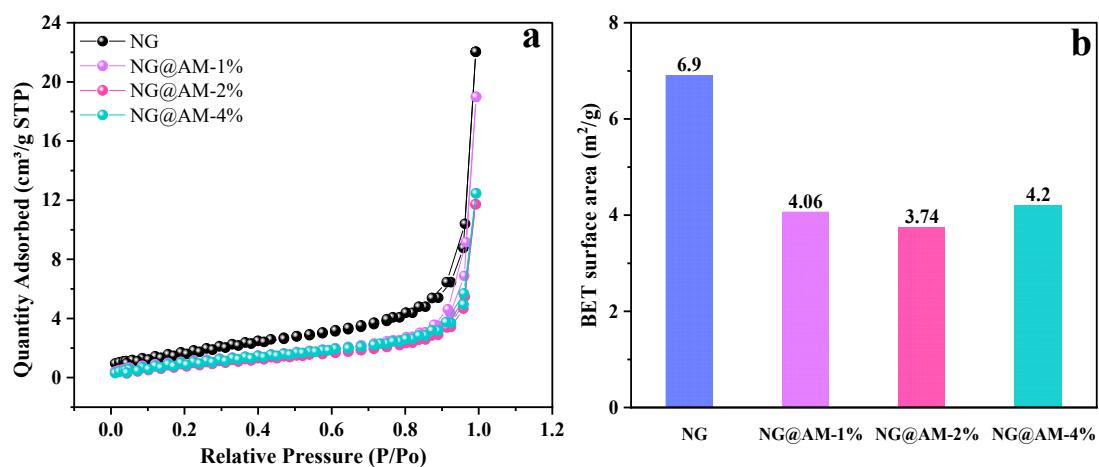
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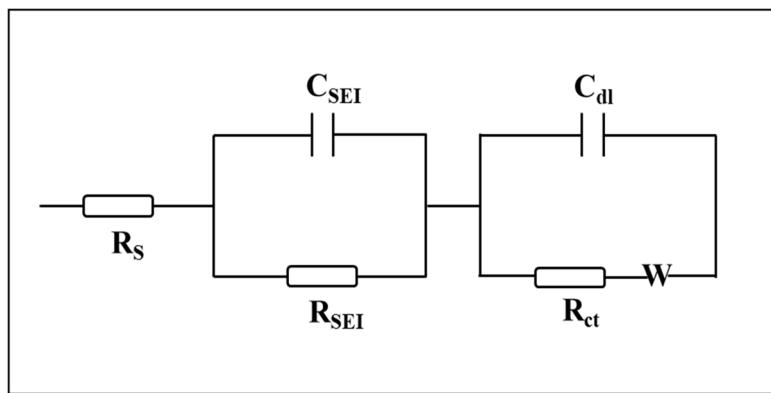
**Figure S1.** The FTIR spectra of the pure MBAA particles and AA-MBAA polymer.



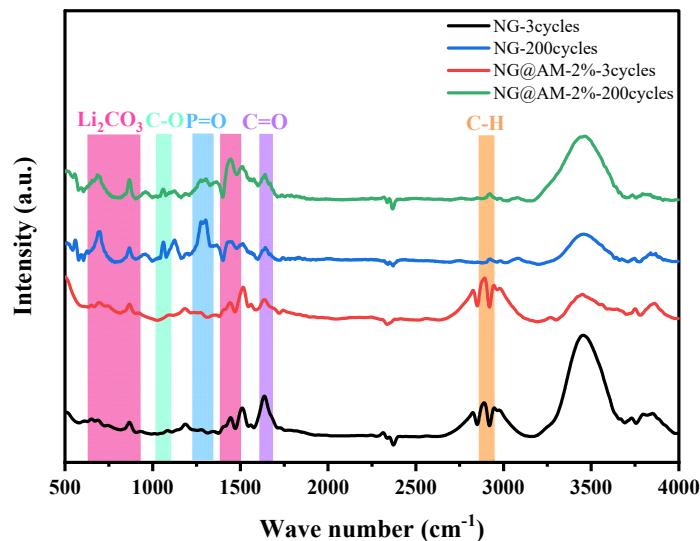
**Figure S2.** XPS spectra with fitted results for the pristine NG and NG@AM-2% electrodes: (a, b) C 1s and (c, d) N 1s.



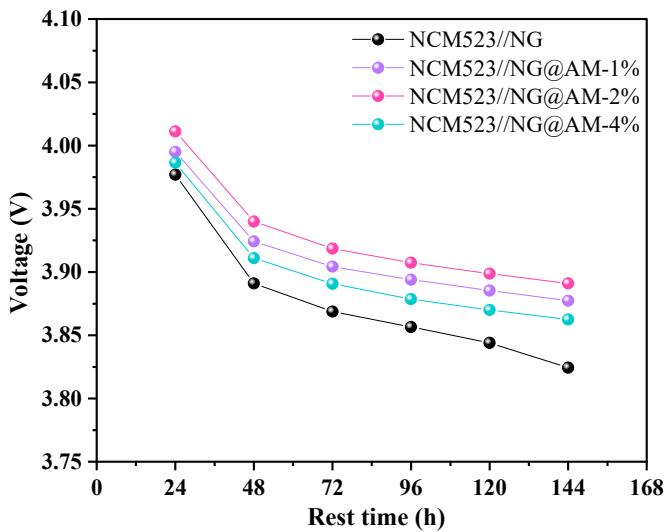
**Figure S3.** (a) The nitrogen adsorption-desorption isotherms and (b) the BET surface area of the pristine NG, NG@AM-1%, NG@AM-2% and NG@AM-4% electrodes.



**Figure S4.** The equivalent circuit for fitting the EIS.



**Figure S5.** The FTIR spectra of the pristine NG and NG@AM-2% anodes at different electrochemical stages.



**Figure S6.** Plot of voltage varies with rest time for full cells with different NG anodes stored at 25°C.

**Table S1.** The charge/discharge specific capacity (CSC/DSC) and initial coulombic efficiency of different NG anodes in the first cycle.

Samples	CSC (mAh g <sup>-1</sup> )	DSC (mAh g <sup>-1</sup> )	ICE (%)
NG	378.52	323.82	85.55
NG@AM-1%	385.07	339.45	88.15
NG@AM-2%	398.92	356.28	89.31
NG@AM-4%	399.04	348.88	87.43

**Table S2.** The corresponding resistance values of the pristine NG and NG grafted with different amounts of the AM polymer anodes after the 3 formation cycles.

R (Ω cm <sup>2</sup> )	R <sub>s</sub>	R <sub>SEI</sub>	R <sub>CT</sub>
NG	1.24	4.13	39.48
NG@AM-1%	1.74	6.04	45.42
NG@AM-2%	1.48	9.73	42.64
NG@AM-4%	2.32	18.34	39.95

**Table S3.** The corresponding resistance values of the pristine NG and NG grafted with different amounts of the AM polymer anodes after the rate test.

R ( $\Omega \text{ cm}^2$ )	R <sub>S</sub>	R <sub>SEI</sub>	R <sub>CT</sub>
NG	2.07	4.21	8.04
NG@AM-1%	1.59	2.16	4.30
NG@AM-2%	1.58	2.01	3.44
NG@AM-4%	1.61	2.87	4.87

**Table S4.** The corresponding resistance values of the pristine NG and NG grafted with different amounts of the AM polymer anodes after the 200 cycles.

R ( $\Omega \text{ cm}^2$ )	R <sub>S</sub>	R <sub>SEI</sub>	R <sub>CT</sub>
NG	11.68	23.02	45.06
NG@AM-1%	2.79	8.95	12.18
NG@AM-2%	2.17	4.21	8.04
NG@AM-4%	5.41	6.41	15.48

**Table S5.** The first charge/discharge specific capacity, initial coulombic efficiency and capacity retention (CR) after 500 cycles of full cells with different NG anodes.

Samples	CSC (mAh g <sup>-1</sup> )	DSC (mAh g <sup>-1</sup> )	ICE (%)	CR (500th)
NG	201.5	160.4	79.60	73.50%
NG@AM-1%	212.9	174.6	82.01	79.23%
NG@AM-2%	216.4	178.2	82.35	82.75%
NG@AM-4%	211.9	173.2	81.74	80.73%

**Table S6.** The K-values and self-discharge rates ( $\eta$ ) of full cells with different NG anodes at 25/50°C.

	NCM523//NG	NCM523//NG @AM-1%	NCM523//NG @AM-2%	NCM523//NG @AM-4%
K-25°C (mV/h)	1.27	0.98	1.01	1.03
$\eta$ -25°C (%)	8.57	7.24	6.61	3.64
$\eta$ -50°C (%)	12.27	11.66	5.62	3.52