Electronic Supplementary Information for:

Polymer Reinforced Carbon Fiber Interfaces for High Energy Density Structural Lithium-Ion Batteries

*Kathleen Moyer[a], Nora Ait Boucherbil[b], Murtaza Zohair[a], Janna Eaves-Rathert[b], Cary L. Pint[c,]**

[a]Interdisciplinary Materials Science Program, Vanderbilt University, Nashville, TN 37235

[b]Department of Mechanical Engineering, Vanderbilt University, Nashville, TN 37235

[c]Department of Mechanical Engineering, Iowa State University, Ames, IA 50011

KEYWORDS: structural battery, multifunctional energy storage composites, carbon fiber, lithium-ion battery, interface engineering, battery material delamination

*Corresponding Author: carypint@iastate.edu

Figure S1

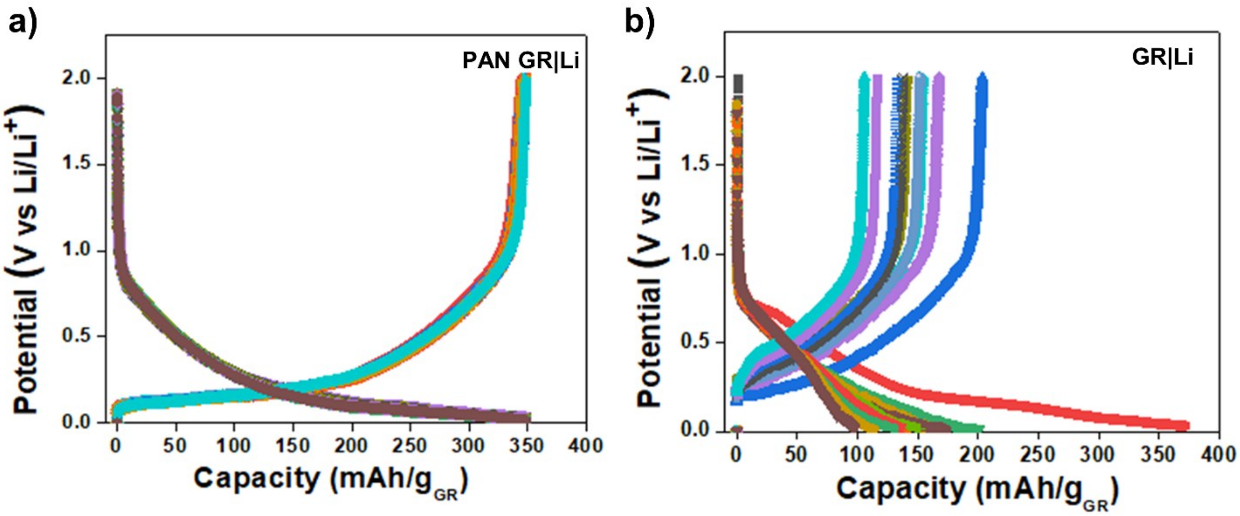


Fig. S1: Galvanostatic half-cell cycling at 0.1 C for 100 cycles. Every 10th cycle is shown for a) PAN GR|Li and b) GR|Li electrodes. Capacity is with respect to GR active material.

Figure S2

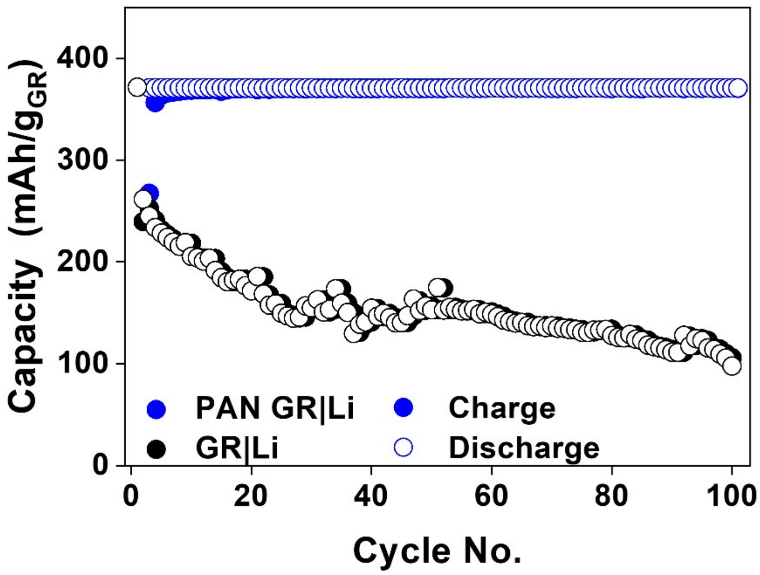


Fig. S2: Half-cell capacity vs cycle number for 100 cycles at 0.1 C for PAN GR|Li and GR|Li electrodes. Capacity is with respect to GR active material.

Figure S3

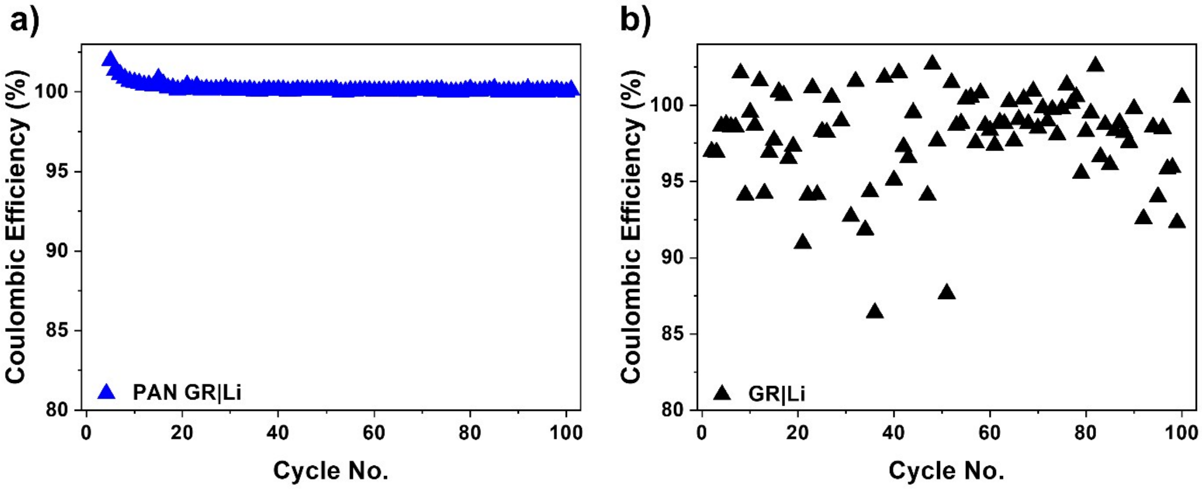


Fig. S3: Coulombic efficiency vs cycle number for 100 cycles at 0.1 C of a) PAN GR|Li and b) GR|Li half-cells.

Figure S4

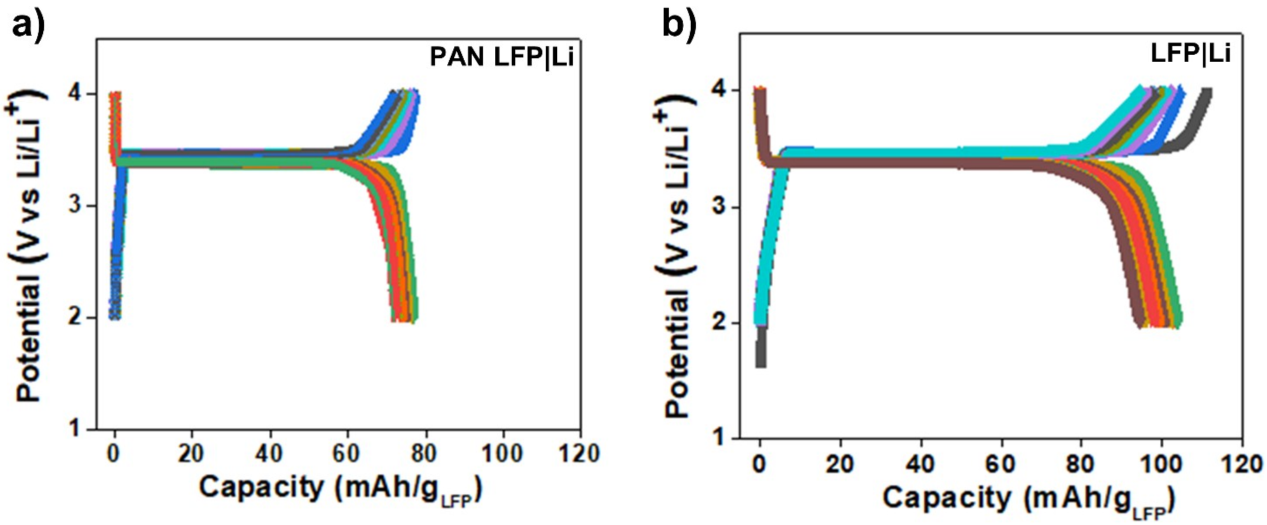


Fig. S4: Galvanostatic charge-discharge curves of every 10th cycle for half-cells cycled at 0.1 C for 100 cycles for a) PAN LFP|LI and b) LFP|Li electrodes. Capacity is with respect to active material of LFP.

Figure S5

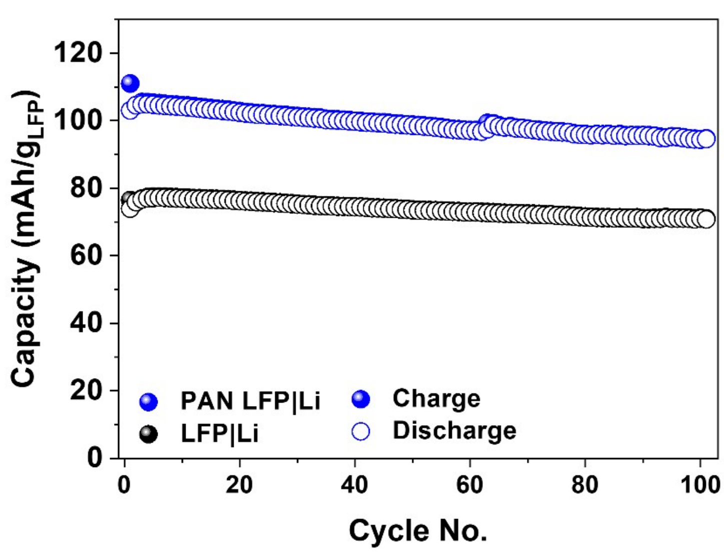


Fig. S5: Capacity with respect to LFP active material vs. cycle number for 100 cycles at 0.1C for PAN LFP|Li and LFP|Li half-cells.

Figure S6

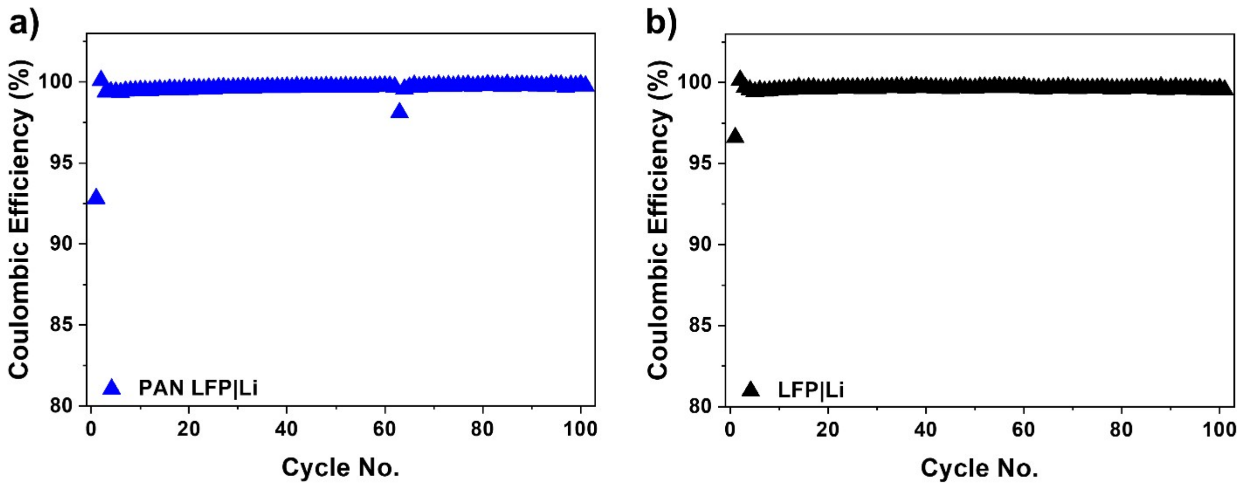


Fig. S6: Coulombic efficiency for half-cells tested at 0.1 C for 100 cycles for a) PAN LFP|Li and b) LFP|Li electrodes.

Figure S7

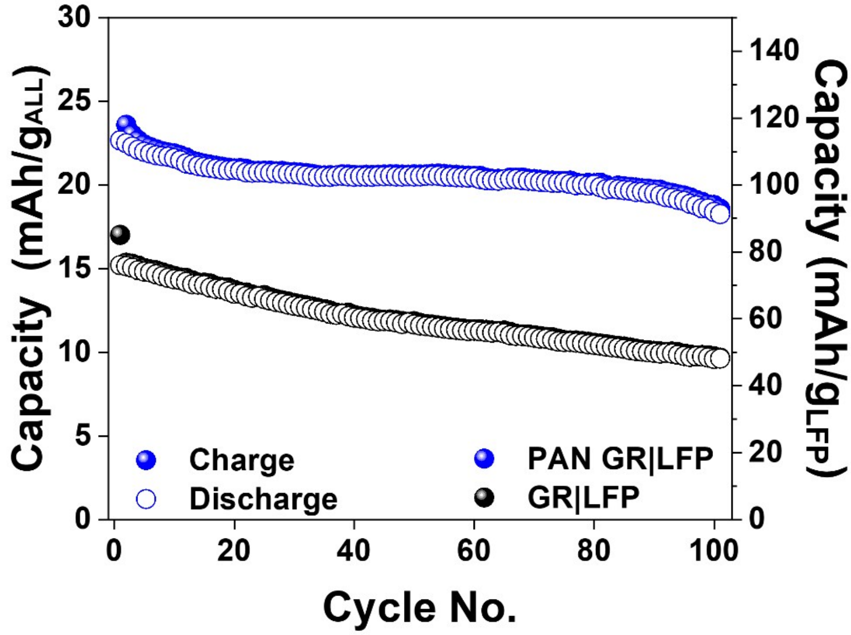


Fig. S7: Coulombic efficiency for half-cells tested at 0.1 C for 100 cycles for a) PAN GR|LFP and b) GR|LFP electrodes.

Figure S8

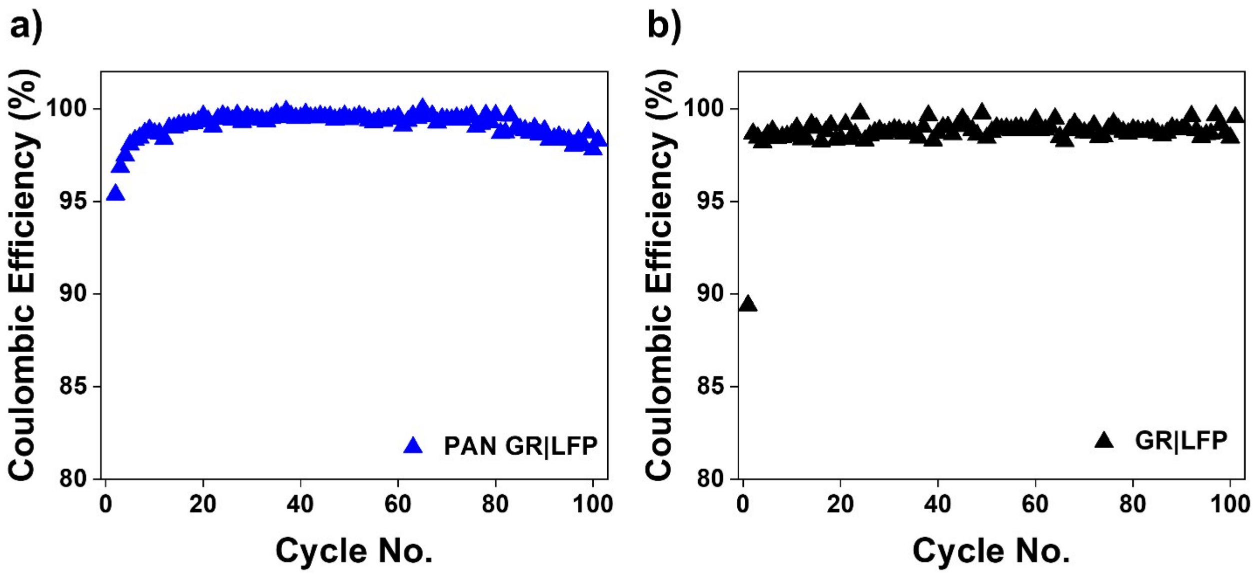


Fig. S8: Coulombic efficiency vs cycle number for galvanostatic cycling of a) PAN GR|LFP and b) GR|LFP electrodes at 0.1 C for 100 cycles.

Figure S9

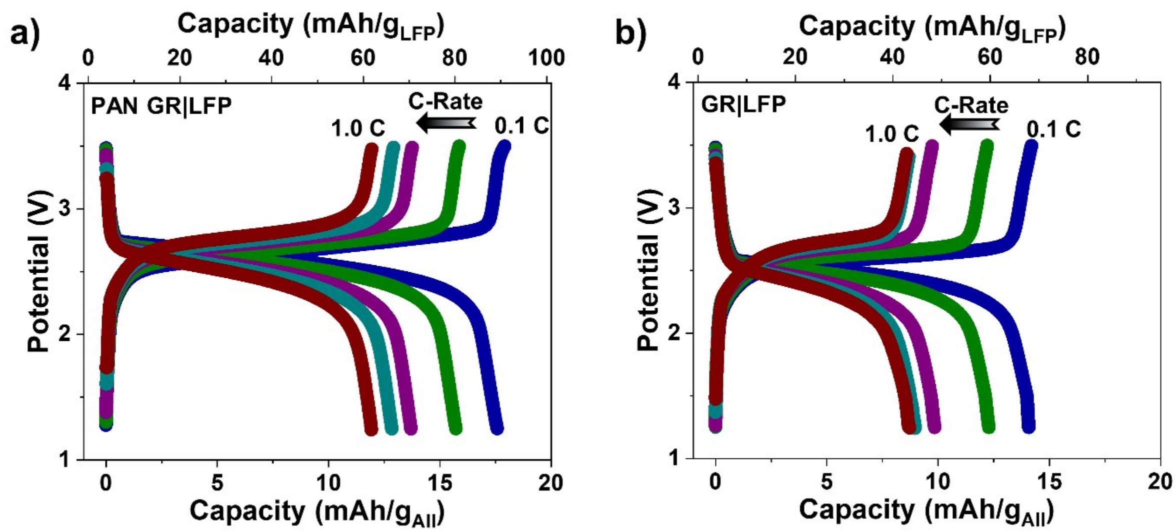


Fig. S9: Galvanostatic charge-discharge curves for second cycle of each rate of rate study for a) PAN GR|LFP and b) GR|LFP electrodes.

Figure S10

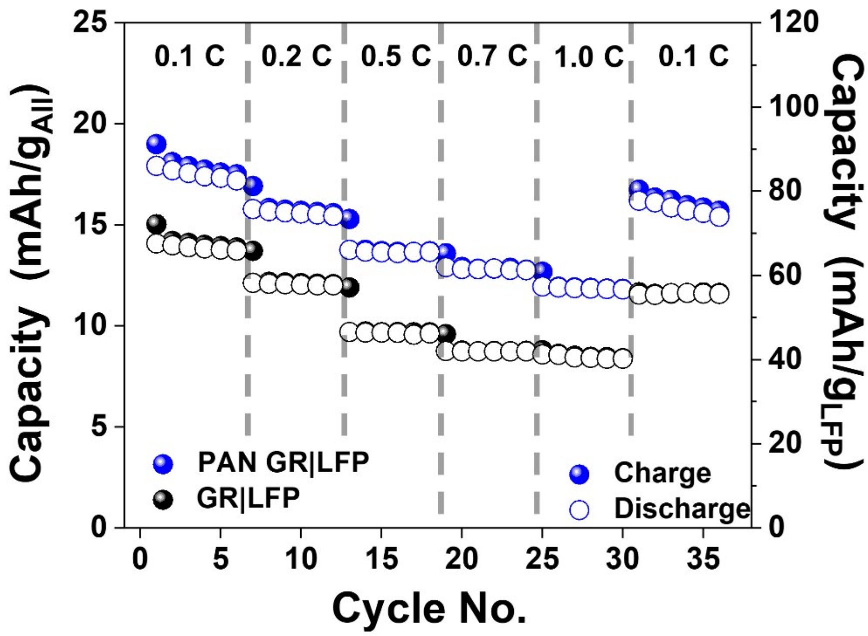


Fig. S10: Rate-study for PAN GR|LFP and GR|LFP full-cell electrodes at 0.1 C, 0.2 C, 0.5 C, 0.7 C, and 1.0 C. Capacity with respect to all mass, inactive and active material, (left y-axis) and with respect to LFP active material (right y-axis).

Figure S11

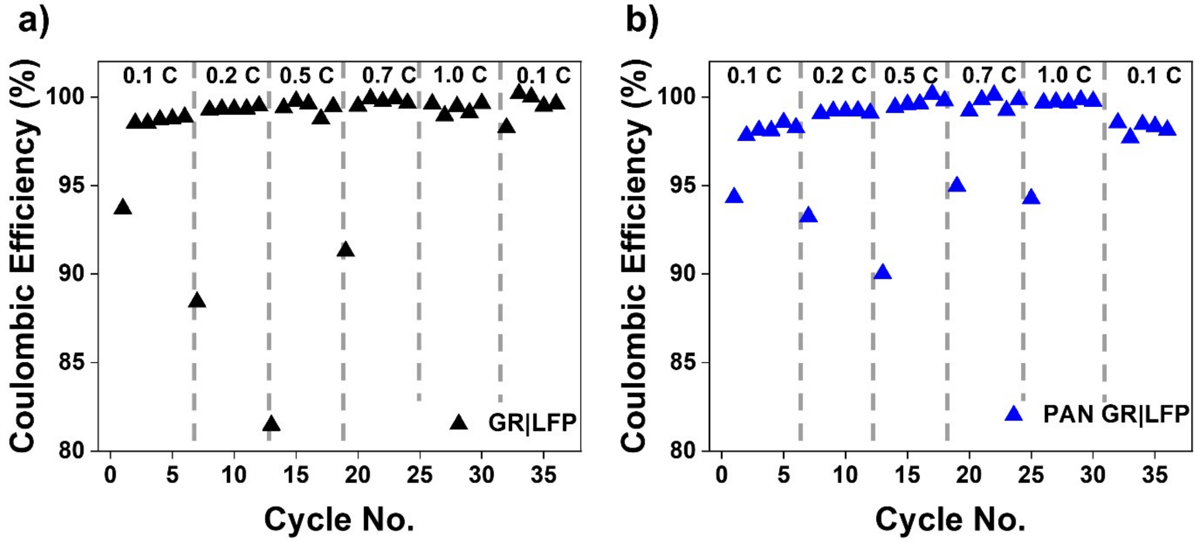


Fig. S11: Coulombic efficiency vs cycle number for rate study of a) PAN GR|LFP and b) GR|LFP full-cell electrodes.

Figure S12

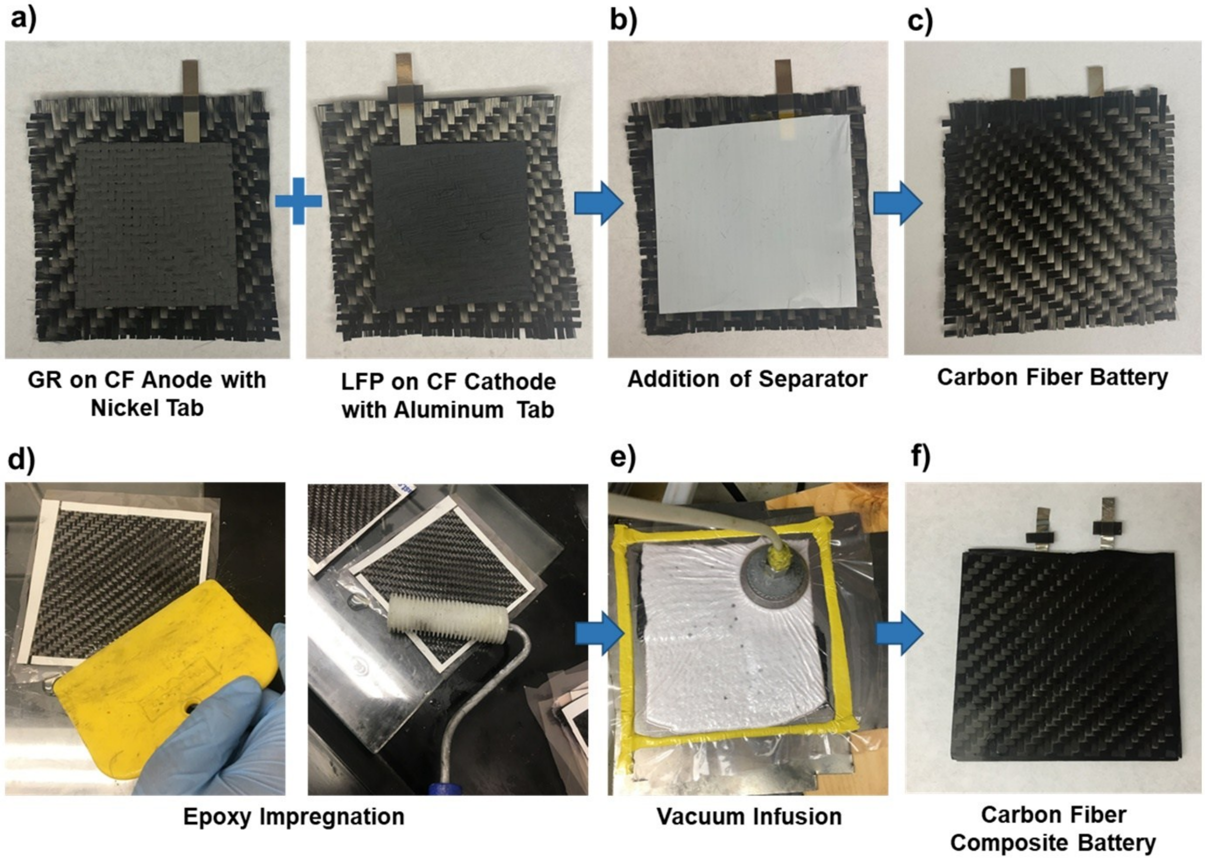


Fig. S12: Composite layup process: a) GR anode on CF current collector with nickel tab and LFP cathode on CF current collector with aluminum tab, b) addition of separator, c) carbon fiber battery, d) epoxy impregnation, e) vacuum infusion process, f) carbon fiber composite structural battery.

Figure S13

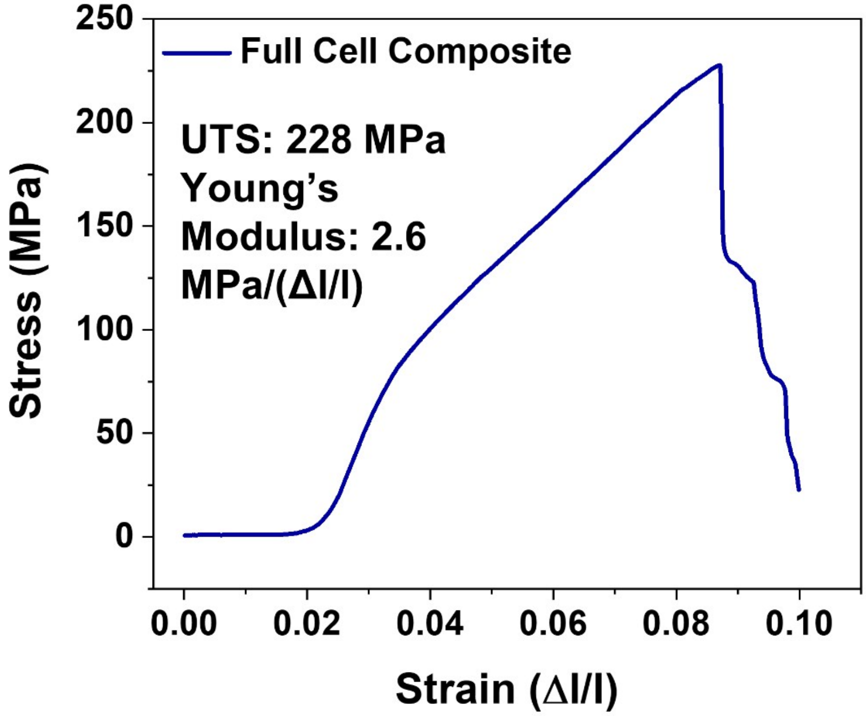


Fig. S13: Tensile testing of composite structural battery.

Figure S14

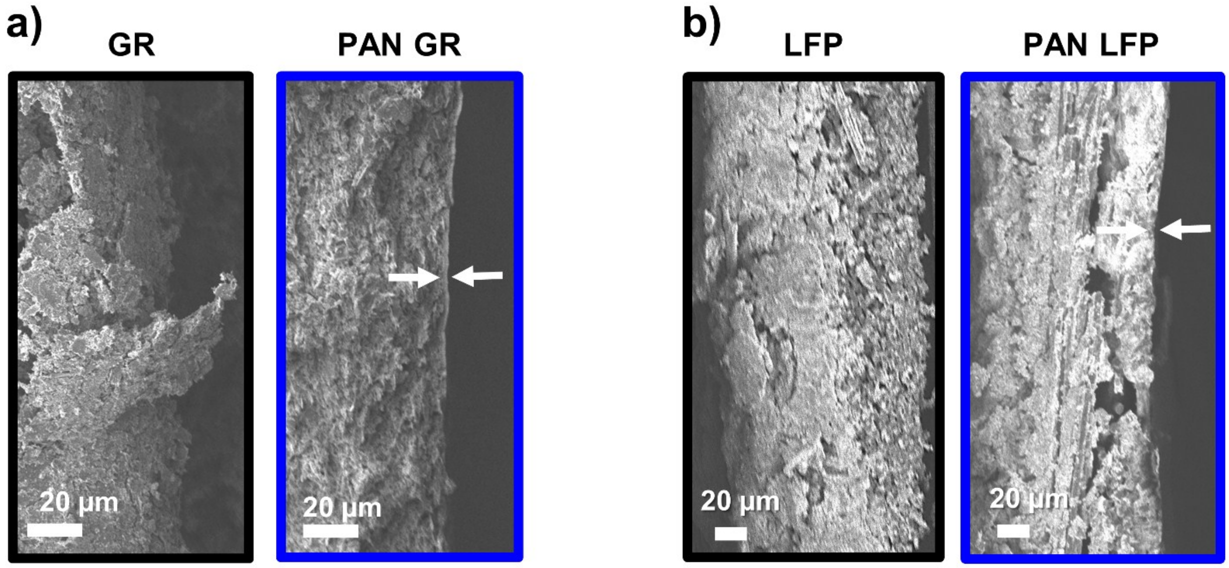
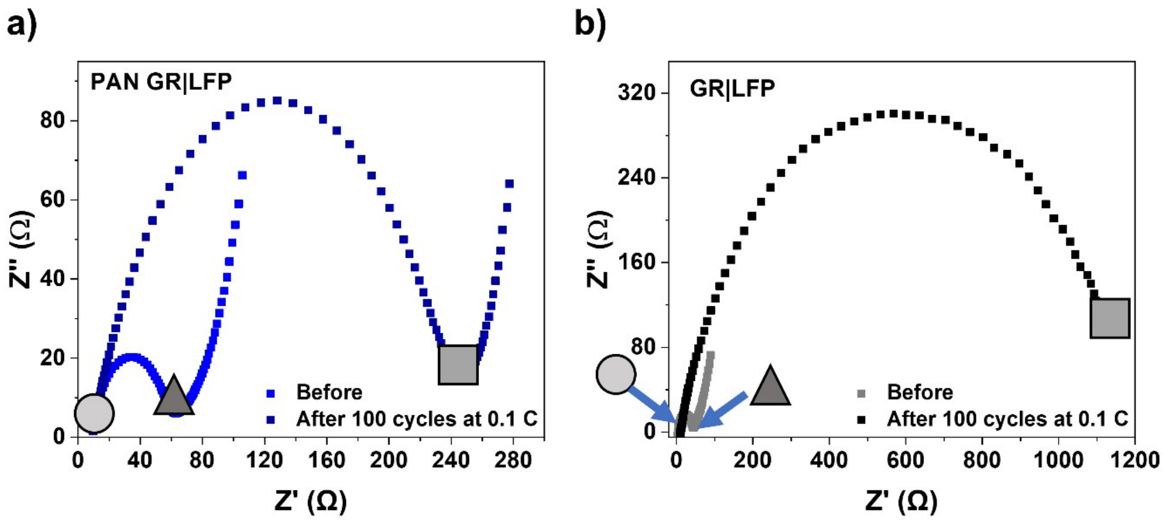


Fig. S14: Cross-section SEMs of a) graphite and b) lithium iron phosphate electrode materials after 100 cycles at 0.1 C with and without PAN.

Figure S15



	PAN GR|LFP	GR|LFP
R_S ●	9.9	10.8
$R_S + R_{CT\ Before}$ ▲	63.1	45.3
$R_{CT\ Before}$ ▲ - ●	53.2	14.4
$R_S + R_{CT\ After}$ ■	247.4	1151.6
$R_{CT\ After}$ ■ - ●	237.5	1140.8

Fig. S15: Analysis of electrochemical impedance spectroscopy data from Fig.4d, e in the manuscript a) PAN GR|LFP and b) GR|LFP full-cells before and after 100 cycles at 0.1 C, and f) capacity retention over 100 cycles at 0.1 C.[1-3]

Modified Multifunctional Efficiency Calculations

Modified Multifunctional Efficiency ($\hat{\sigma}^{mf}$):[4]

$$\hat{\sigma}^{mf} = 2\sqrt{\sigma^{e} \times \sigma^{s}}$$

Where σ^{e} is the ratio of energy density:

$$\sigma^{e} = \frac{\Gamma}{\Gamma_{typ}}$$

And σ^{s} is the ratio of specific strength:

$$\sigma^{s} = \frac{S}{S_{typ}}$$

Here, the typically energy density of a graphite|lithium iron phosphate was assumed to be 90 Wh/kg at 1C.

$$\sigma^{e}{}_{PAN\ coated} = \frac{\Gamma}{\Gamma_{typ}} = \frac{39.96\ Wh/kg}{90\ Wh/kg} = 0.33$$

$$\sigma^{e}{}_{Uncoated} = \frac{\Gamma}{\Gamma_{typ}} = \frac{15.41\ Wh/kg}{90\ Wh/kg} = 0.17$$

The theoretical specific strength of the Fibre Glast System 1000 Laminating Epoxy Resin Standard Part Kit 1000/1025 as reported by the manufacturer is 257 MPa. Upon testing the strength of the epoxy in a tensile test with the full-cell carbon fiber composite, the experimental specific strength was 228 MPa.

$$\sigma^{s} = \frac{S}{S_{typ}} = \frac{228}{257} = 0.887$$

And the modified multifunctional advantage for each type of electrode is:

$$\hat{\sigma}^{mf}{}_{PAN\ Coated} = 2\sqrt{\sigma^{e} \times \sigma^{s}} = 2\sqrt{0.33 \times 0.887} = 1.08\ (>1)$$

$$\hat{\sigma}^{mf}{}_{Uncoated} = 2\sqrt{\sigma^{e} \times \sigma^{s}} = 2\sqrt{0.17 \times 0.887} = 0.78\ (<1)$$

REFERENCES

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