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Supporting Information

Efficient Polysulfides Barrier of Graphene Aerogel-Carbon Nanofibers-Ni Network for High-Energy-Density Lithium-Sulfur Batteries with Ultrahigh Sulfur Content

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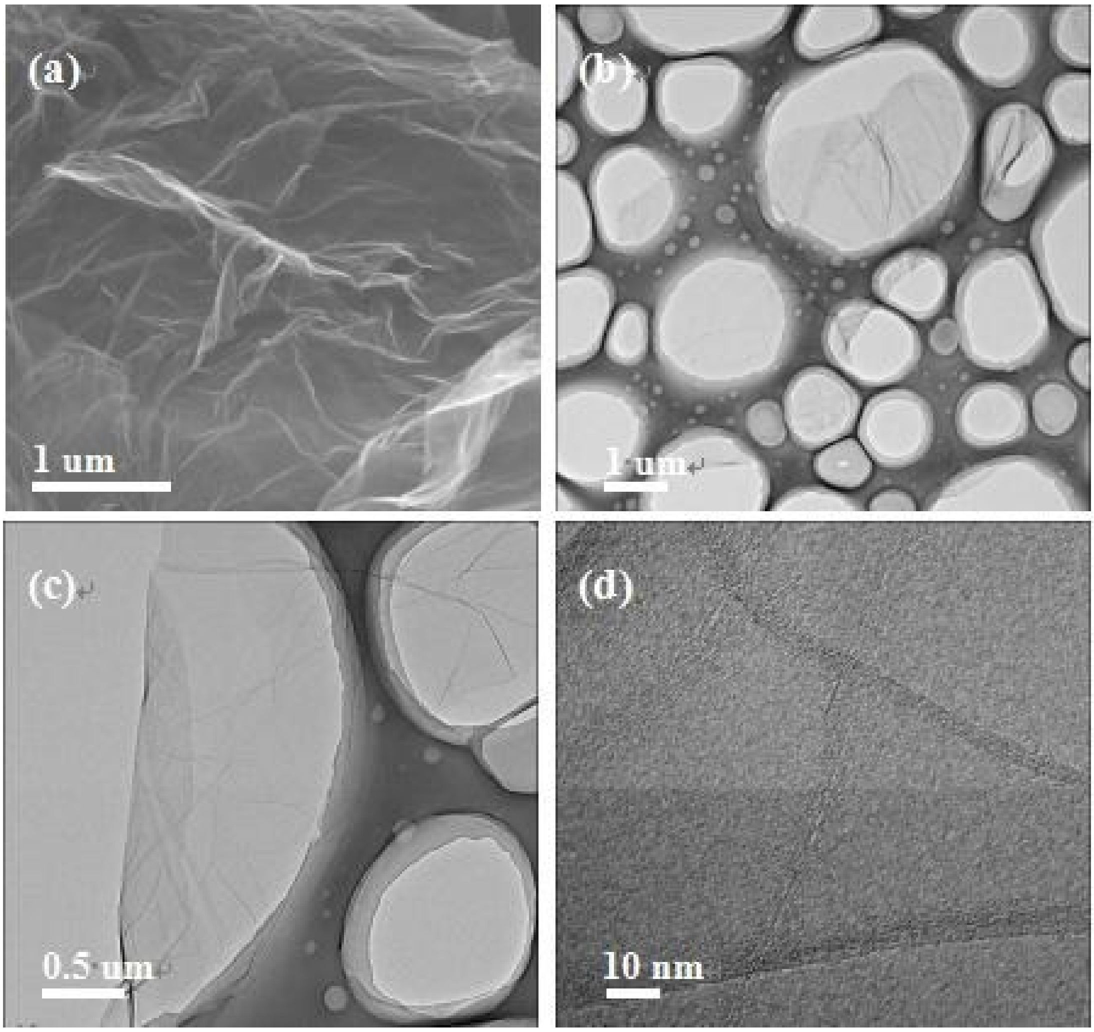

Figure S1. (a) SEM, (b, c) TEM and d) HRTEM images of exfoliated graphene oxide

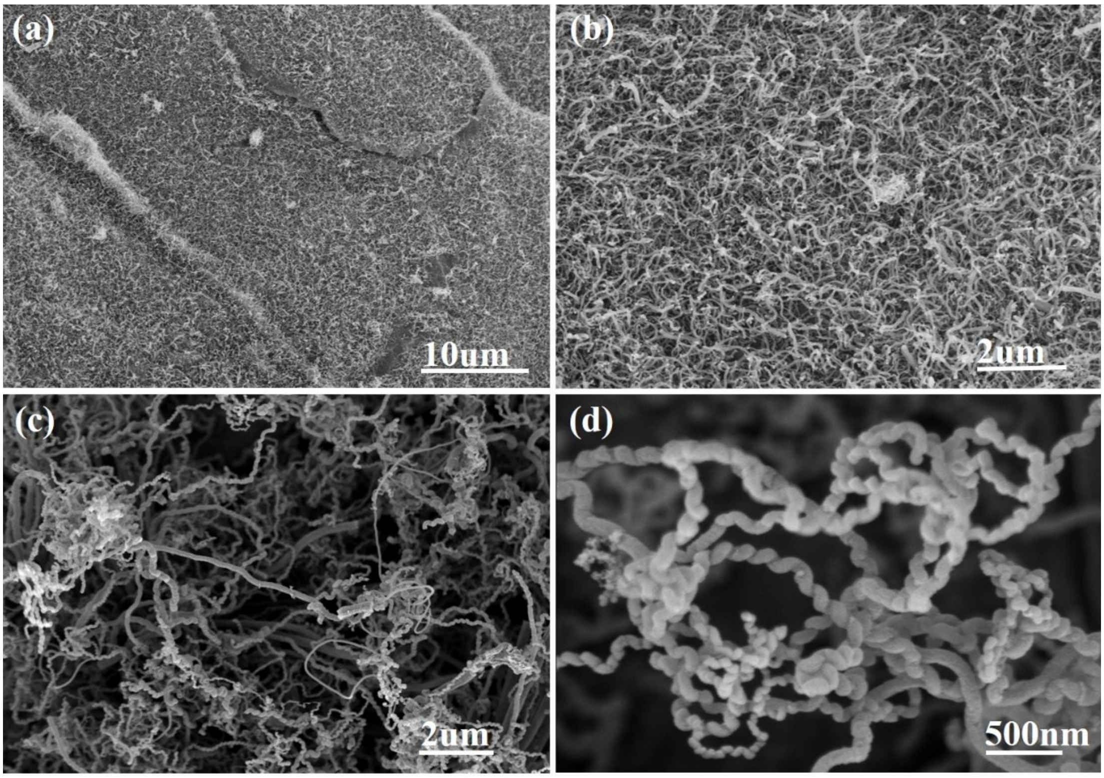

Figure S2. SEM images of GA-CNF-Ni hybrids synthesized with growth time of (a, b) 30 min and (c, d) 2 h.

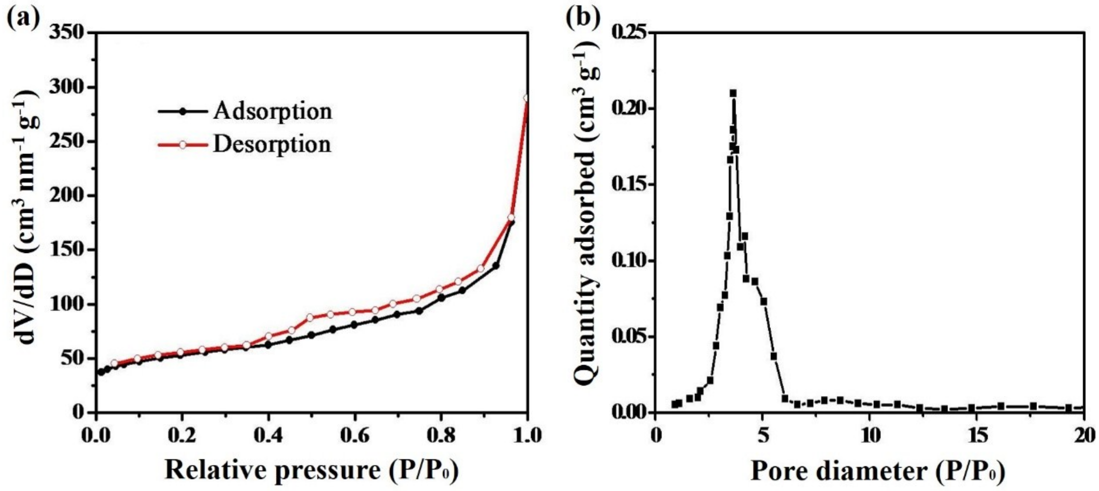

Figure S3. (a) N_2 adsorption-desorption isotherms of GA and (b) corresponding pore size distribution.

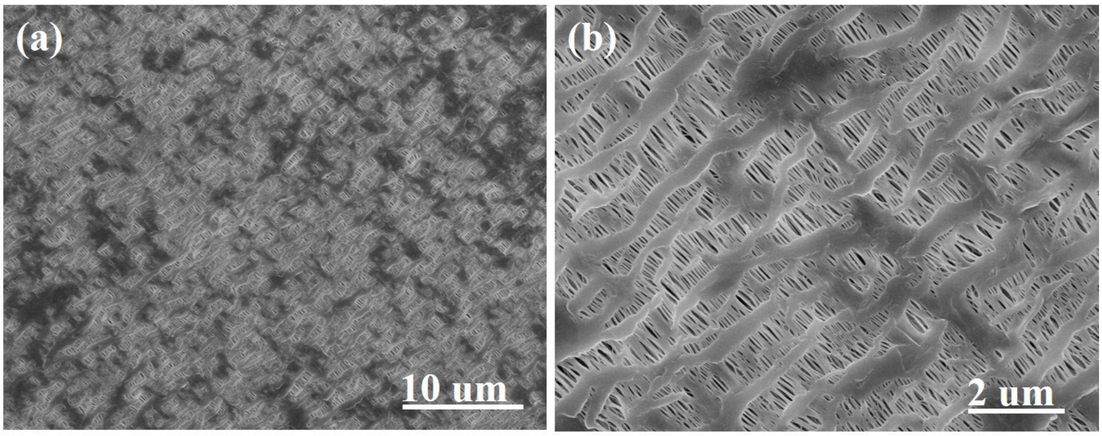

Figure S4. SEM images of pristine separator.

Table S1. Comparisons of electrical properties between GA-CNFs-Ni and GA/CNT coating layers

Coating layer	Resistivity ($\Omega \cdot cm$)	Conductivity (S cm^{-1})
GA-CNFs-Ni	6.8×10^{-3}	147
GA/CNT	1.2×10^{-2}	83

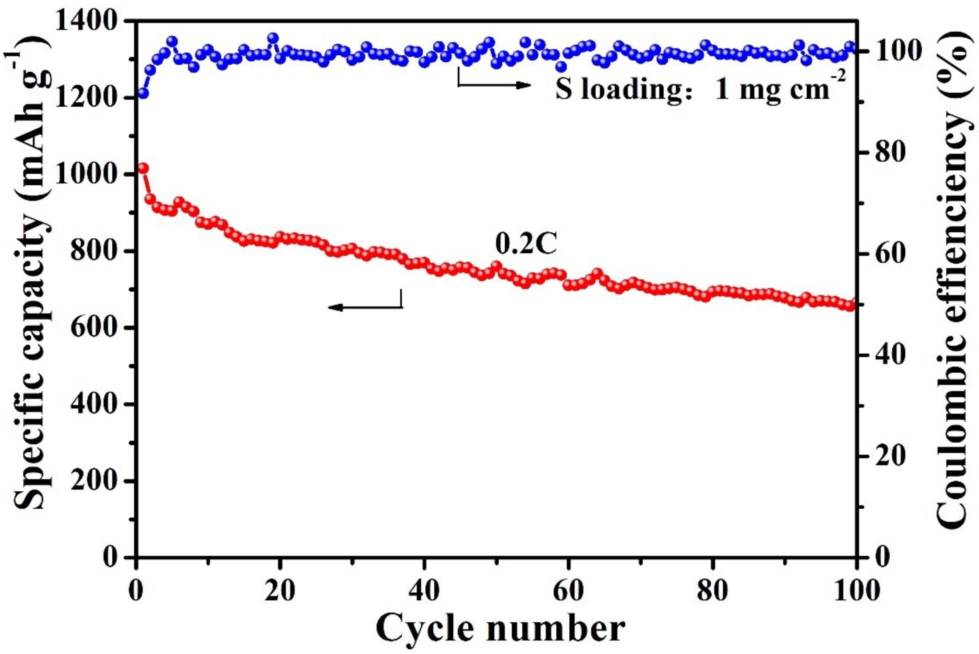

Figure S5. The cycle performance of GA-CNFs-Ni/S cathode with pristine separator at 0.2 C

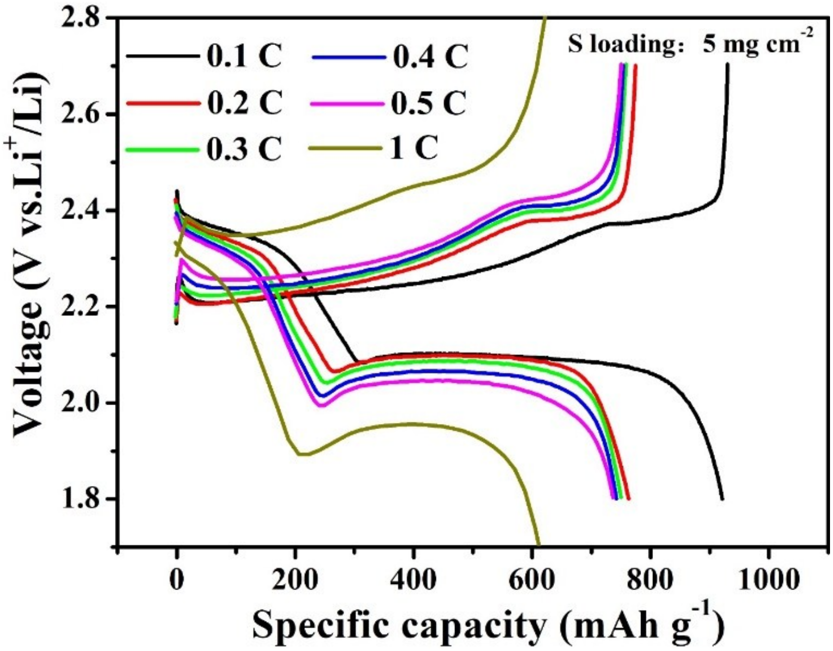

Figure S6. Charge-discharge voltage profiles of the cells fabricated with GA-CNFs modified separator at 0.2 C

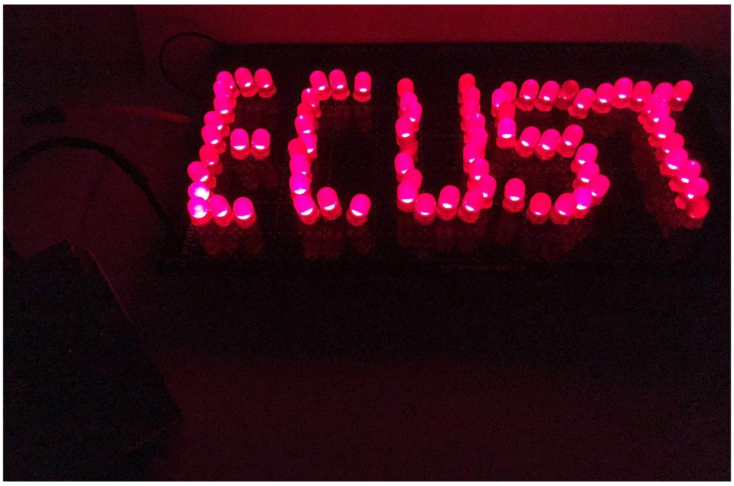

Figure S7. Optical image of LED lamps with the pattern of "ECUST" lighted by the cell based on GA-CNFs-Ni coated separator and ultra-high loading sulfur cathode.

Table S2. Comparison of electrochemical properties functional separators in Li–S cells

Coating or interlayer	Sulfur loading	Sulfur content	Cathode	Discharge Capacity	Areal capacity (mAh cm^{-2})	E/S ratio	Ref
MoO_3@CNT	1.0	60%	Super P/S	~755 mAh g^{-1}, 200th, 0.3 C	<2.0	N/A	[S1]
SPEEK	6.0	45%	CNT/S	650 mAh g^{-1}, 500th, 0.2 C	~3.9	N/A	[S2]
SSNS/CNT	1.0	65%	KB/S	680 mAh g^{-1}, 100th, 0.2 C	<2.0	N/A	[S3]
Ultrathin hollow carbon sphere	3.0	57%	CB/S	730 mAh g^{-1}, 100th, 0.2 C	~2.2	~13	[S4]
EUC-CNF	1.5	70%	Super P/S	~1038 mAh g^{-1}, 50th, 0.2 C	<2.0	N/A	[S5]
GO@MoS2	3.64	70%	CB/S	~600 mAh g^{-1}, 95th, 0.2 C	~2.2	~14	[S6]
Fe-N-carbon nanofiber	1.0	77%	S/KB	~601.9 mAh g^{-1}, 500th, 0.5 C	<2.0	N/A	[S7]
MoO_3	0.9-1.0	49%	CNT/S	~782 mAh g^{-1}, 100th, 0.5 C	<2.0	40	[S8]
Co_9S_8	2.0-5.6	70%	Super P/S	~985 mAh g^{-1}, 200th, 0.1 C	5.5	10	[S9]
Nano TiO_2	2.0	63%	AB/S	~883 mAh g^{-1}, 180th, 0.1 C	<2.0	N/A	[S10]
CNF@VS_2/CNT@GN	5.6	60%	Super P/S	~750 mAh g^{-1}, 43th, 0.3 C	4.1	N/A	[S11]
OCNT	2.5	65%	CB/S	~820 mAh g^{-1}, 60th, 0.2 C	~2.0	N/A	[S12]
TiO_2/C	1.1	54%	AB/S	~1000 mAh g^{-1}, 60th, 0.1 C	<2.0	36	[S13]
Gr-CNT-Ni	7.68	64%	polysulfide catholyte	~696 mAh g^{-1}, 100th, 0.2 C	~5.3	10	[S14]
CNT@ZIF	1.2	56%	AB/S	~1100 mAh g^{-1}, 60th, 0.2 C	<2.0	36	[S15]
CNF@ZrO_2	2.7	70%	CB/S	~800 mAh g^{-1}, 60th, 0.2 C	~2.1	40	[S16]
g-C_3N_4	4.0	45%	GO/S	~600 mAh g^{-1}, 60th, 0.2 C	~2.4	20	[S17]
aCNT	6.0	67%	aCNT/S	617 mA h g^{-1}, 60, 0.1 C	3.7	N/A	[S18]
CoP	3.24	56%	rGO/S	~800 mAh g^{-1}, 100th, 0.2 C	~2.7	N/A	[S19]
MnO_2	1.5-2.5	66%	KB/S	~603 mAh g^{-1}, 500th, 0.5 C	<2.0	7.0-8.8	[S20]

rGO@SL	3.8	75%	CB/S	700 mAh g^{-1} 50^{th}, 0.05 C	2.7	8	[21]
MoP/rGO	3.6-3.9	85.6%	CNTs-AB/S	760 mAh g^{-1} 300^{th}, 0.5 C	3.2	15	[22]
TiN	1.3	70%	Super P/S	744 mAh g^{-1} 200^{th}, 0.5 C	＜2.0	46	[23]
Sb_2S_3	1.0	65%	KB/S	680 mAh g^{-1} 100^{th}, 0.2 C	＜2.0	50	[24]
GA-CNF-Ni	5.0	90%	CB/S	890 mAh g^{-1}, 90^{th}, 0.2 C	4.5	12	This work
	10.0			550 mAh g^{-1}, 60^{th}, 0.1 C	5.5	6	

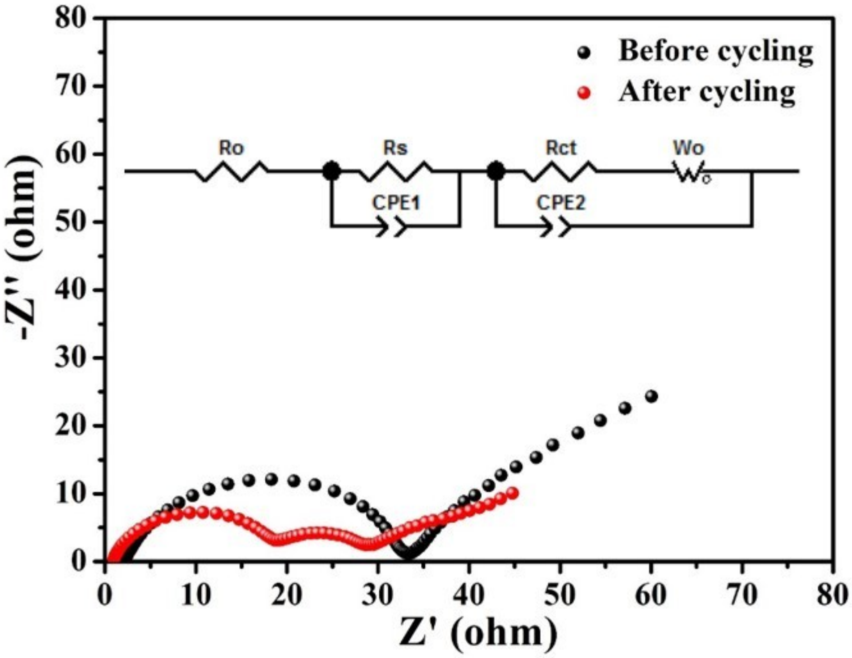

Figure S8. Nyquist plots and corresponding equivalent circuit (insert) of the cells based on "double high" sulfur cathode and GA-CNFs-Ni modified separator before and after cycling.

Table. S3. The parameters of Nyquist plot of the cells based on "double high" sulfur cathode and GA-CNFs-Ni modified separator before and after cycling

Samples	**R_o (Ω)**	**R_s (Ω)**	**R_{ct} (Ω)**
Before cycling	2.2	30.4	-
After cycling	0.9	18.4	8.2

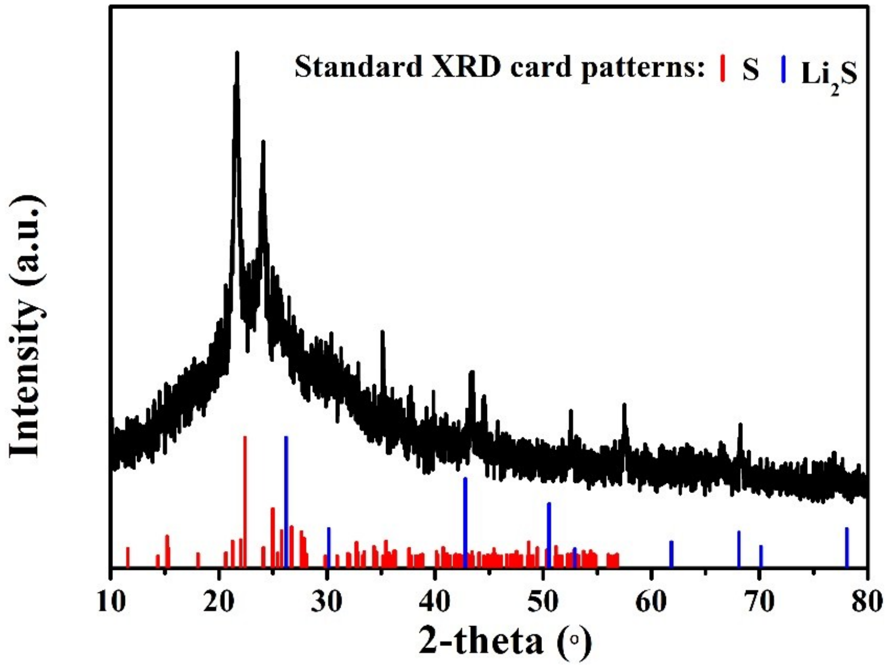

Figure S9. XRD patterns of GA-CNFs-Ni coated separator after cycling and standard XRD patterns sulfur and Li_2S powder.

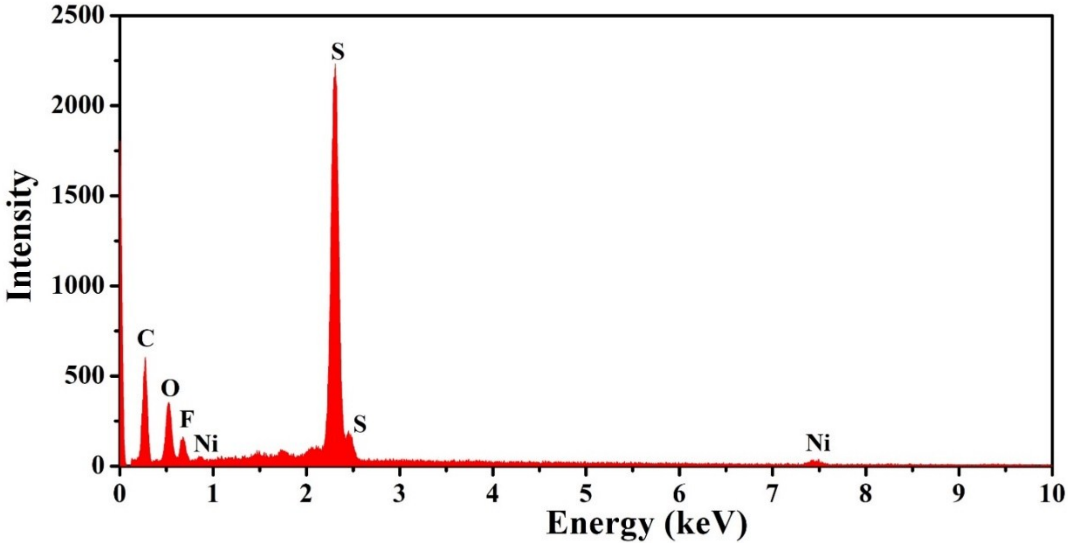

Figure S10. EDX spectra of GA-CNFs-Ni coated separator after 90 cycles at 0.2 C

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