Electronic Supplementary Material (ESI) for Green Chemistry.

Efficient separation of electrode active materials and current collector metal foils from spent lithium-ion batteries by green deep eutectic solvent

Yunhui Hua[a,b], Zhenghe Xu[c], Baojun Zhao[b,d] and Zuotai Zhang[a*]

a. School of Environmental Science and Engineering, Southern University of Science and Technology (SUSTech), Shenzhen 518055, China.

b. School of Chemical Engineering, The University of Queensland, St Lucia 4072, Brisbane, Australia.

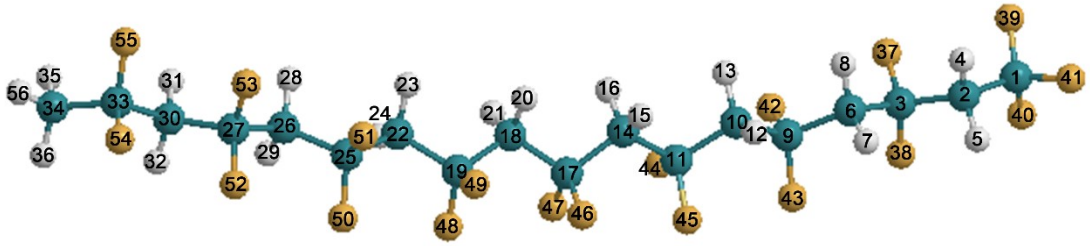
c. Department of Materials Science and Engineering, Southern University of Science and Technology (SUSTech), Shenzhen 518055, China.

d. Faculty of Materials, Metallurgy and Chemistry, Jiangxi University of Science and Technology, Ganzhou 341000, China.

*Corresponding Author:

Prof. Zuotai Zhang, E-mail address: zhangzt@sustech.edu.cn

Table S1 Calculated chemical bond energies in the simulated PVDF molecular chain.



PVDF/C-H	Bond energy (kcal/mol)	PVDF/C-F	Bond energy (kcal/mol)	PVDF/C-C	Bond energy (kcal/mol)
2-4	117.42	3-37	117.97	2-3	101.60
2-5	117.12	3-38	117.71	3-6	101.92
6-7	115.93	9-42	117.85	6-9	100.94
6-8	115.73	9-43	117.74	9-10	101.26
10-12	115.73	11-44	117.87	10-11	101.07
10-13	115.66	11-45	117.82	11-14	101.26
14-15	115.66	17-46	117.88	14-17	101.14
14-16	115.65	17-47	117.85	17-18	101.29
18-20	115.63	19-48	117.89	18-19	101.14
18-21	115.65	19-49	117.88	19-22	101.30
22-23	115.60	25-50	117.89	22-25	101.05
22-24	115.65	25-51	117.92	25-26	101.32
26-28	115.53	27-52	117.92	26-27	100.93
26-29	115.67	27-53	118.06	27-30	101.49
30-31	115.46	33-54	119.43	30-33	102.56
30-32	115.71	33-55	119.99		
Average	115.86	Average	118.10	Average	101.35

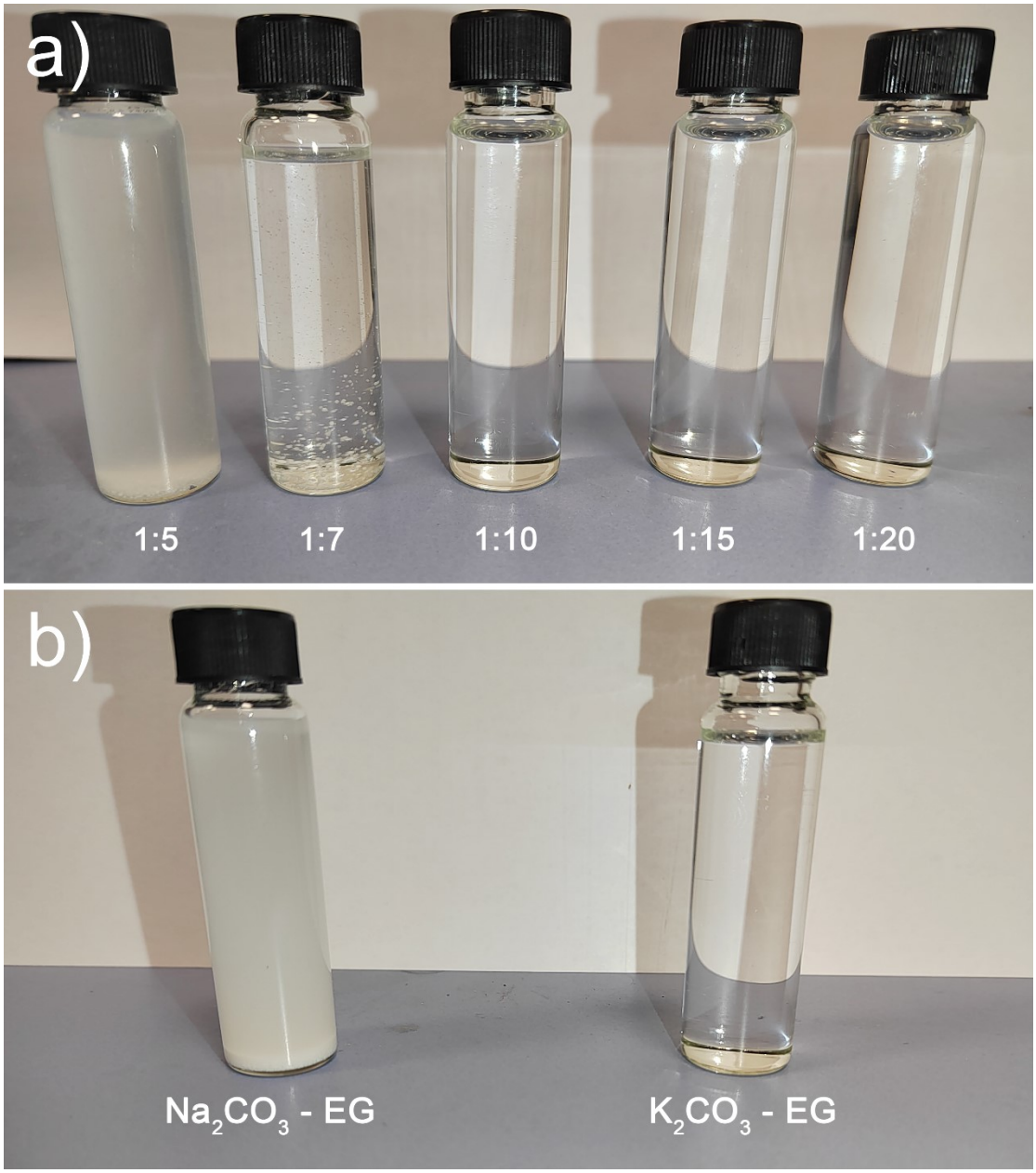


Fig. S1 a) DES synthesis with different K_2CO_3: EG molar ratio. The 1:10 is suitable to form homogeneous eutectic solvent with enough K_2CO_3. b) DES synthesis using different sorts of carbonate salt. The K_2CO_3 can form the homogeneous DES with EG (right) while the Na_2CO_3 (left) does not have the same ability.

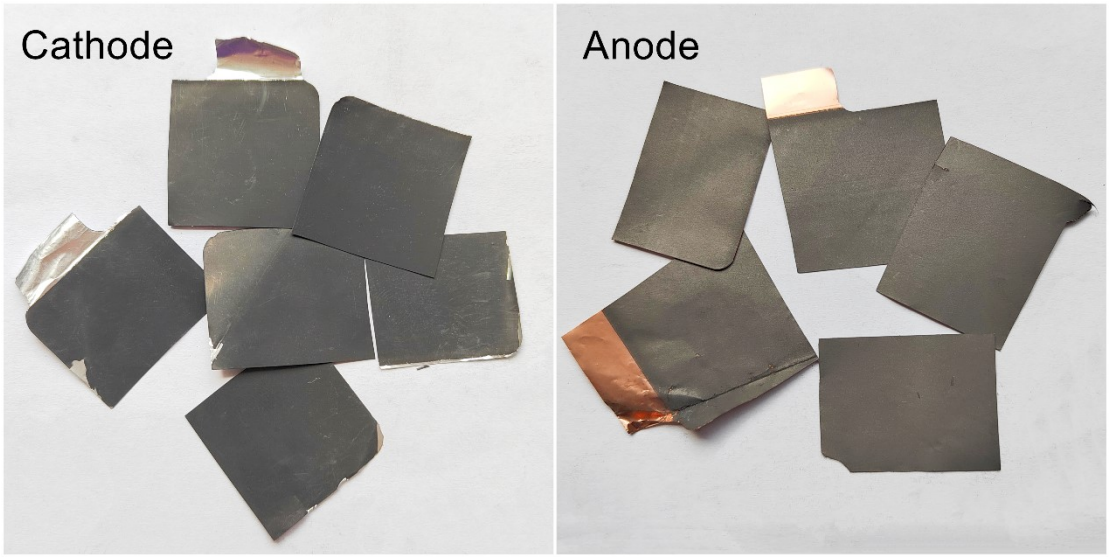


Fig. S2 Cathode and anode pieces before separation in this study

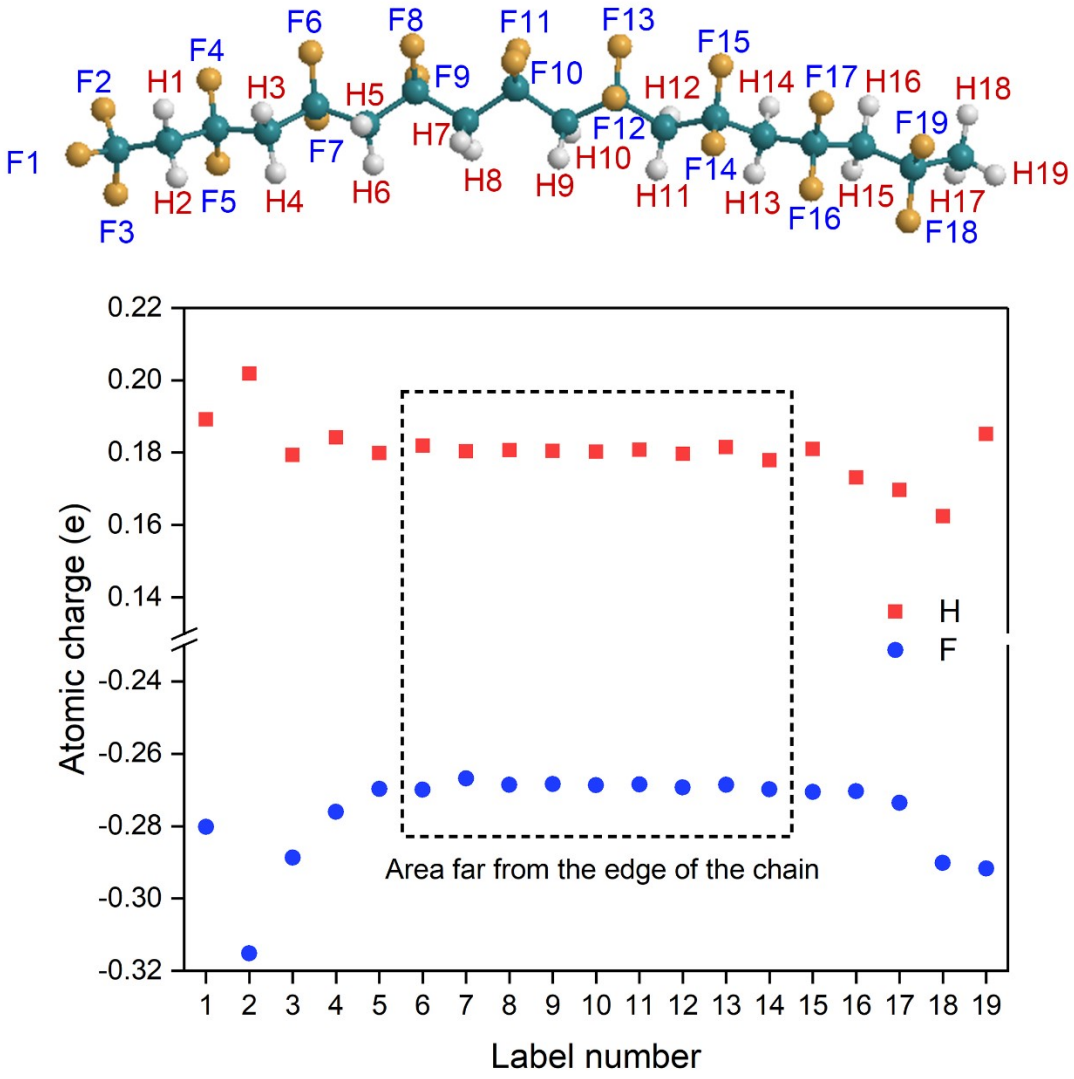


Fig. S3 Atomic charges of H and F on the PVDF molecular chain which was simulated by using 18 carbon atoms. The area far from the edge of the chain has almost stable atomic charge distributions, which can be regarded as similar to the condition on the actual polymer.

Fig. S4 The reaction phenomenon of cathode electrode in 20% K_2CO_3 water solution. Gas is clearly seen generating from the solution, and the Al foil is being corroded quickly (left). After reaction, Al foil has been severely corroded and mixed with the active material, making them impossible to be separated (right).

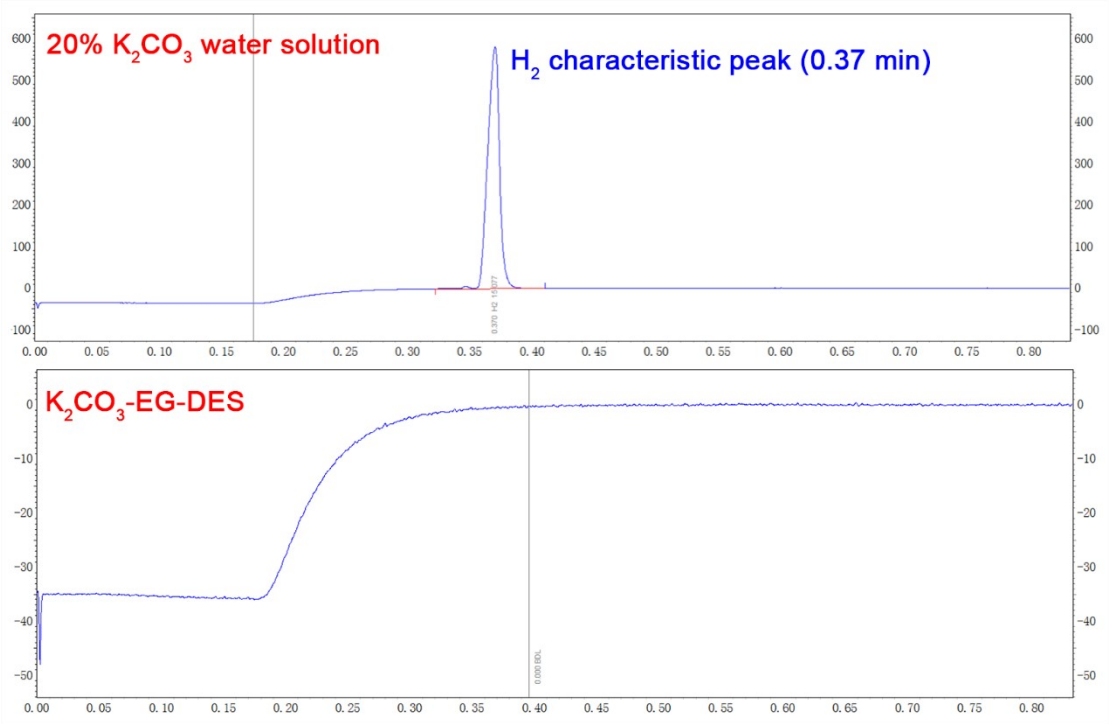


Fig. S5 Comparison of generated H_2 gas between 20% K_2CO_3 water solution and K_2CO_3-EG DES systems by GC-MS. Reactions in water solution and DES are conducted at 80 ℃ and 100 ℃ respectively. H_2 is clearly detected from water solution group, but not detected from DES group (Characteristic peak at 0.37 min).

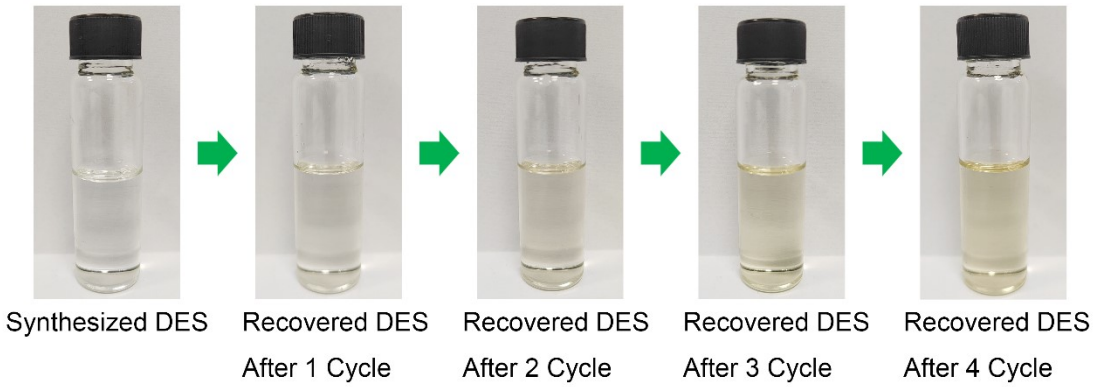


Fig. S6 Optical photographs of synthesized DES and recovered DES after different cycles.

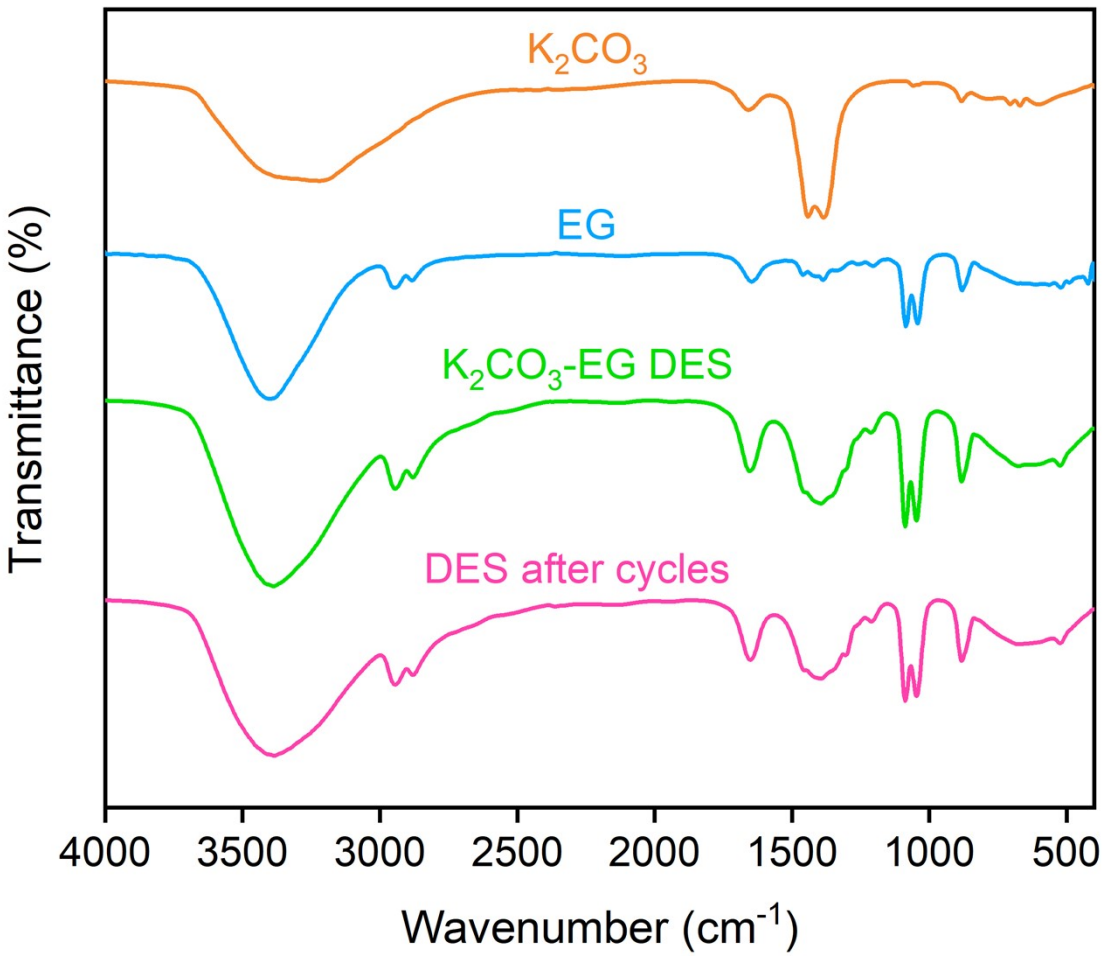


Fig. S7 FT-IR spectra of K_2CO_3, EG, synthesized DES and the recovered DES after cycles. There are no obvious changes in the spectra of the recovered DES, indicating its stability under the reaction condition and the feasibility for multiple cycles.

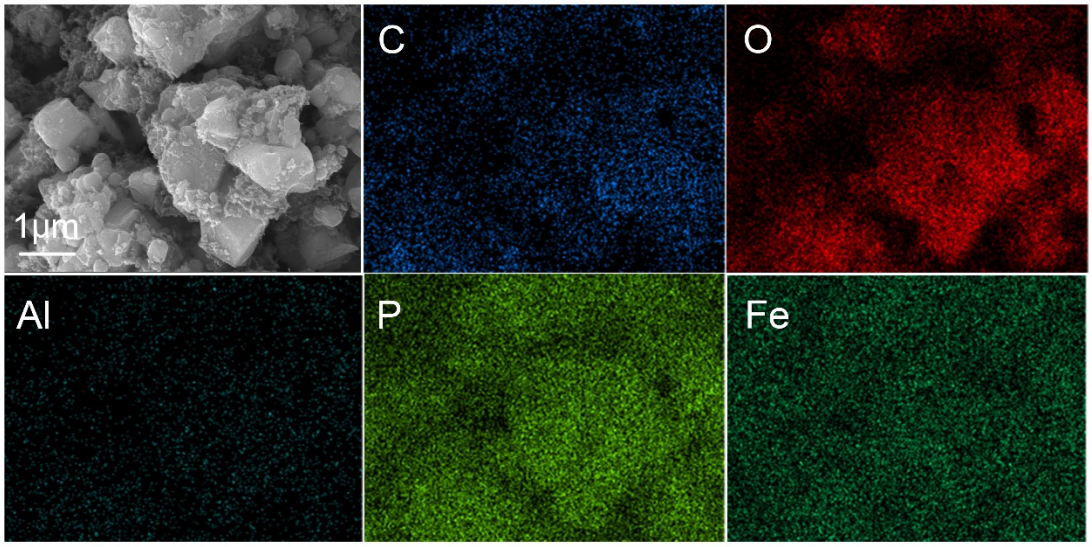


Fig. S8 SEM and EDX mapping images of separated cathode active material from spent LFP battery cathode electrode

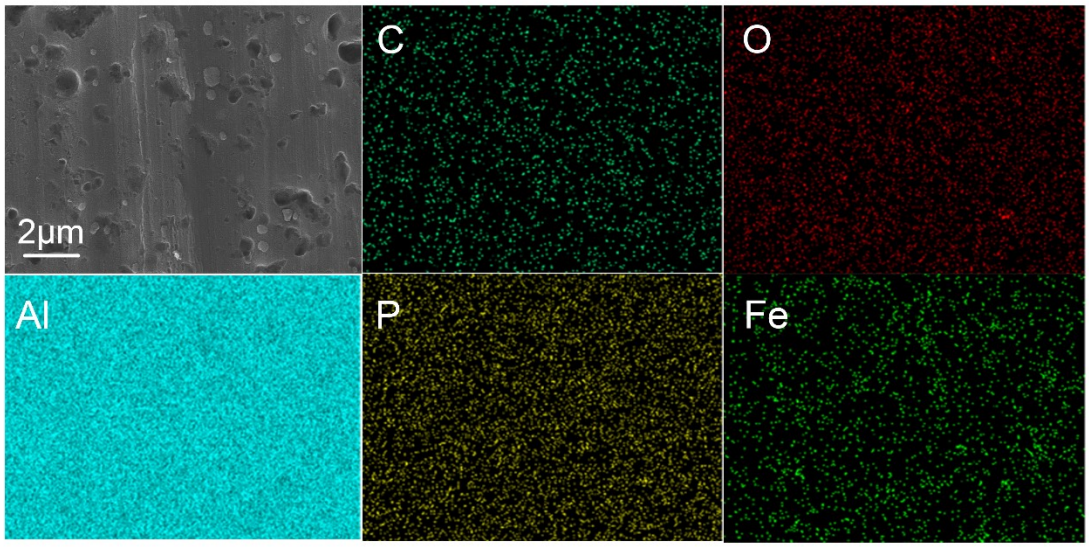


Fig. S9 SEM and EDX mapping images of separated Al metal foil from spent LFP battery cathode electrode.

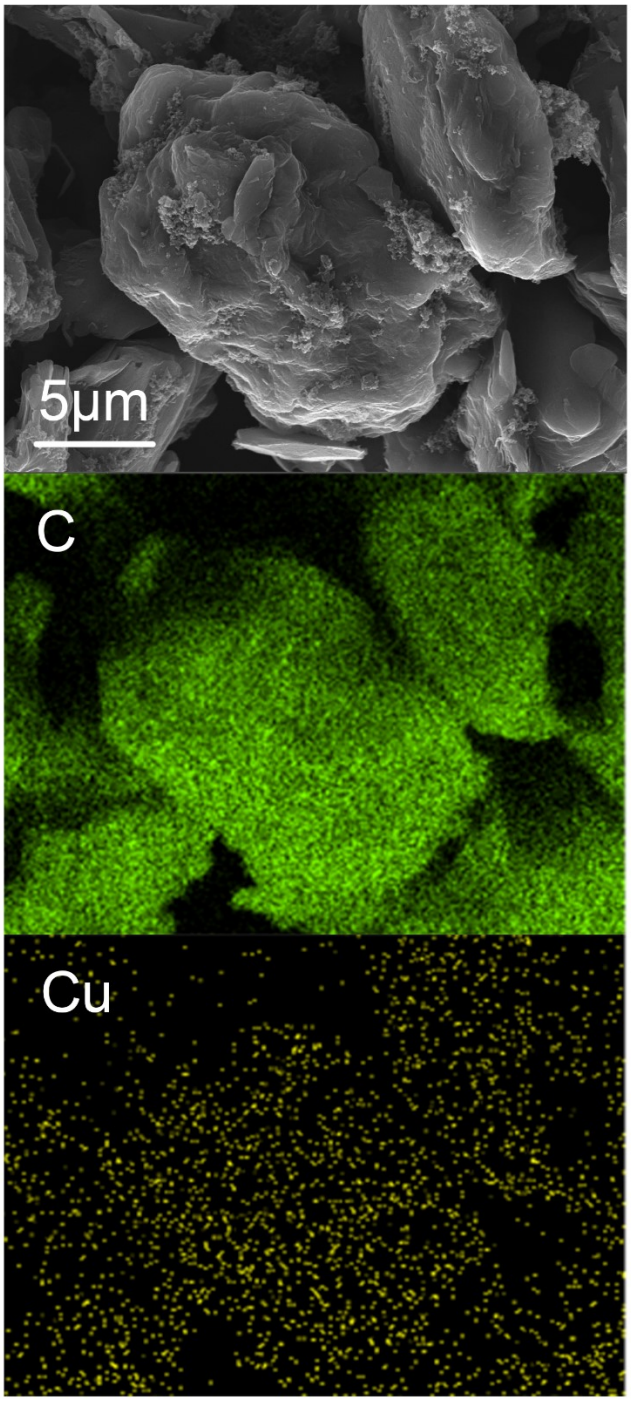


Fig. S10 SEM and EDX mapping images of separated anode graphite from spent LFP battery anode electrode

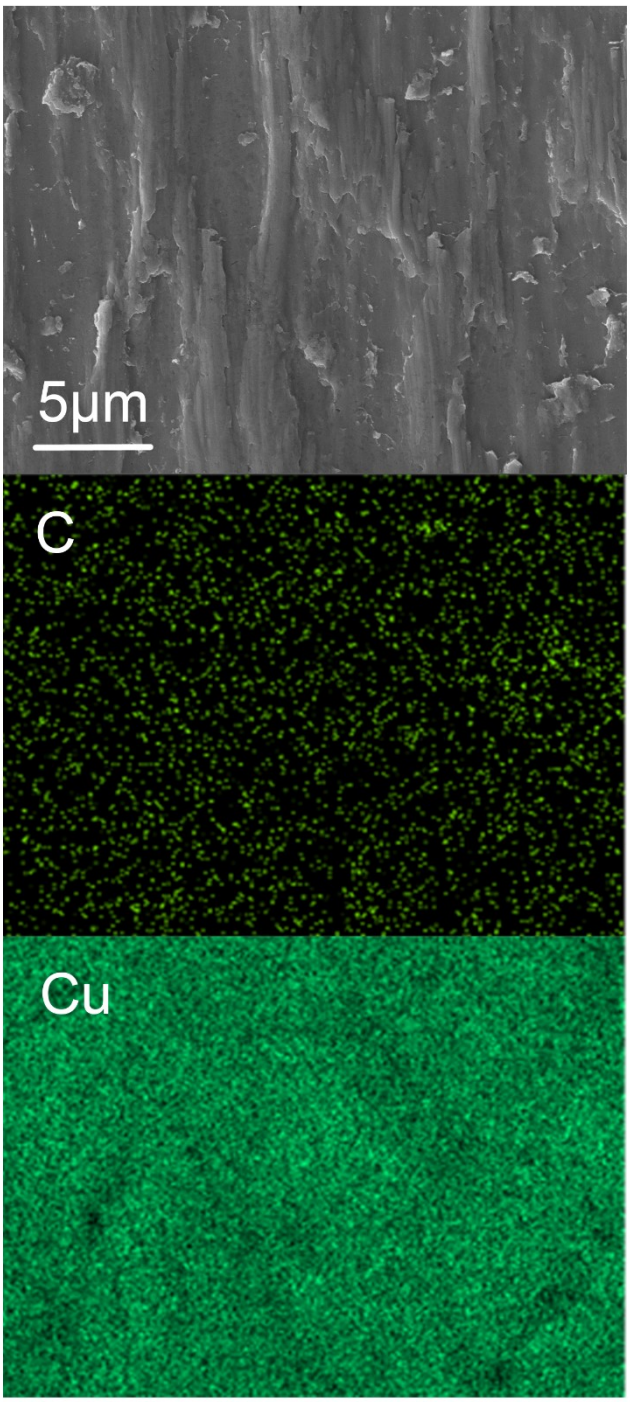


Fig. S11 SEM and EDX mapping images of separated Cu metal foil from spent LFP battery anode electrode.

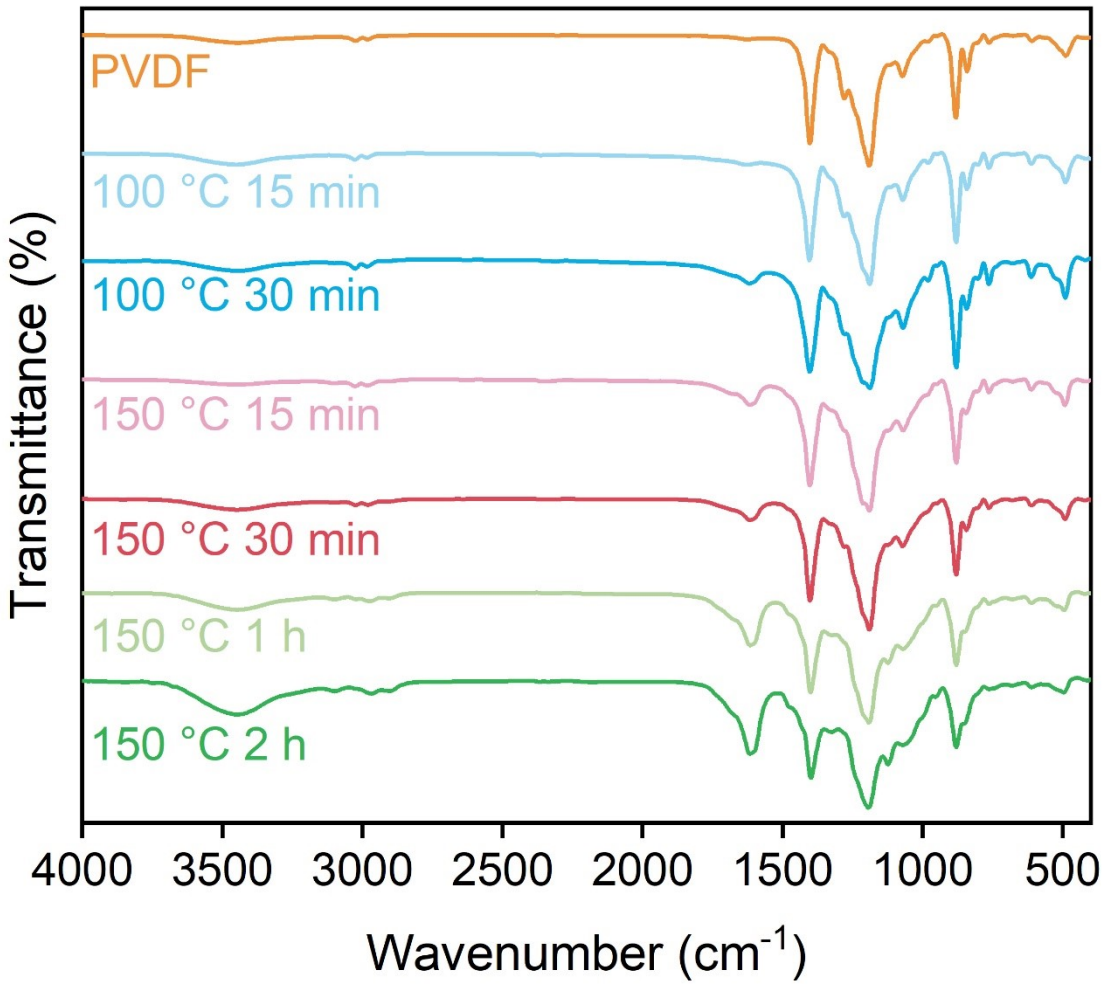


Fig. S12 FT-IR spectra of PVDF powder after reaction with K_2CO_3-EG DES under different conditions. Higher temperatures and longer time lead to deeper degradation.

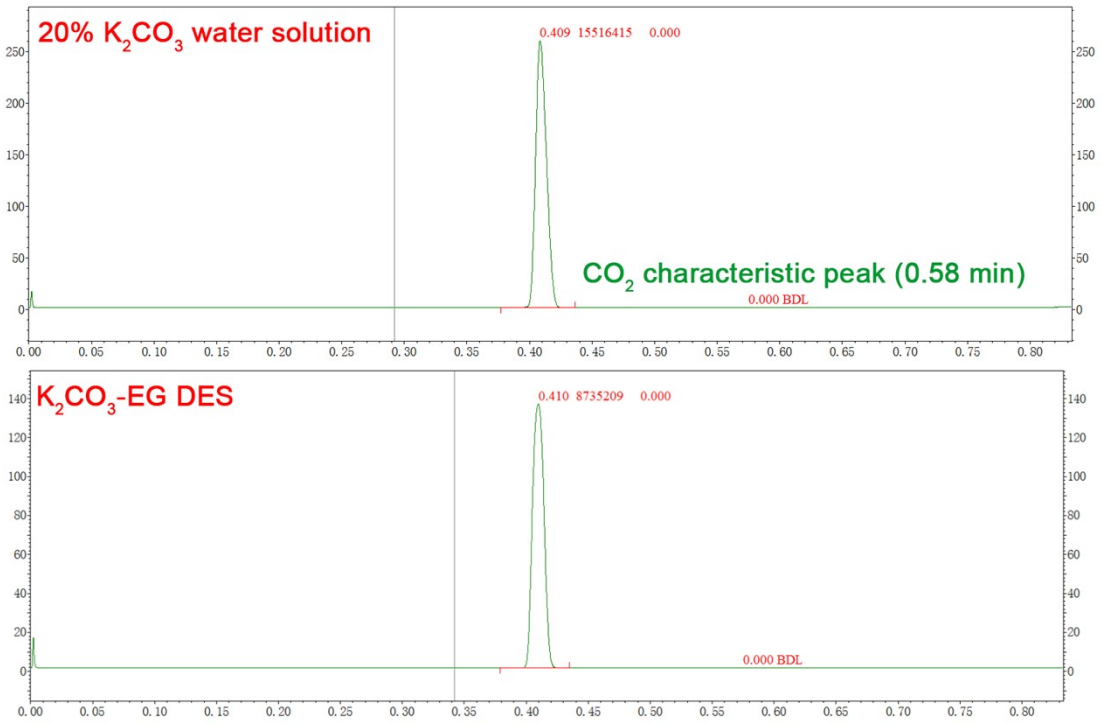


Fig. S13 Comparison of generated CO_2 gas between 20% K_2CO_3 water solution and K_2CO_3-EG DES systems by GC-MS. Reactions in water solution and DES are conducted at 80 ℃ and 100 ℃ respectively. Both systems have no CO_2 generated during the process (Characteristic peak of CO_2 should appear at 0.58 min)