Supplemental Information

Supplemental Table and Figure

Table S1. Components and content of spent lithium batteries.

Element	Li	Co	Ni	Mn	Al	Cu
Content	6.5%	20.4%	20.9%	18.2%	0.46%	0.05%

The cathode powder was procured from the cathode powder that had been subjected to pre-treatment by the merchant in large quantities of the LNCM111-type used lithium-ion battery (the used LNCM111 was discharged in a sodium chloride solution with a mass fraction of A 10% fraction was then obtained, and the discharged battery was subsequently dried and disassembled to obtain the positive and negative electrodes. These were roasted for two hours at 450°C, during which the electrolyte and binder were decomposed. Further leaching with water yielded a pure positive electrode powder that could be used in this experiment.

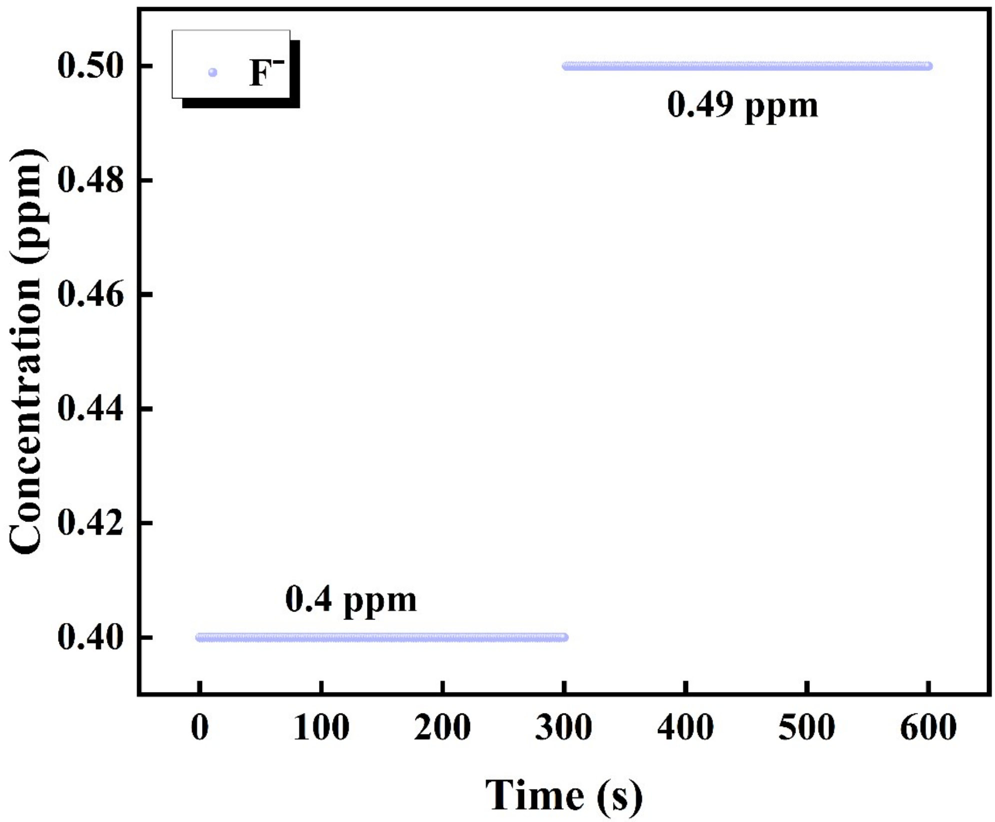

Figure.S1. LNCM111 Elemental Fluorine in Anode Powder Test Chart

Table S2. Uniform Design Experimentation of factor levels.

Factor	1	2	3	4
Temperature (℃)	**40**	**50**	**60**	**70**
Time (min)	**3**	**6**	**9**	**12**
S/L (g L^{-1})	**20**	**25**	**30**	**35**

The elemental fluorine content was determined using an ion chromatography (IC) instrument. The US Thermo Fisher Scientific ICS-5000+ was employed, utilising an Aptar 855 potentiometric titration robot, an ISE-fluoride ion-selective electrode, an LL ISE reference electrode, and a sample dilution-adjustment pH measurement method.

Table S3. The regression equations derived from the Mixing Design and Uniform experimental design are related to the correlation coefficient R^2 and p-value statistics.

	Mixing Design		Uniform Experimental Design	
Model	**p-value**	**R^2**	**p-value**	**R^2**
Ni	<0.002	0.96	<0.001	0.99
Co	<0.002	0.96	<0.001	0.99
Mn	<0.002	0.96	<0.001	0.99
Li	<0.002	0.96	<0.001	0.99

Table S4. Statistical data on fitted values and errors of Nickel leaching rates in Mixing Experiments.

No	Observed values	Fitted value	Error (%)
1	0.7660	0.7523	0.0137
2	0.7508	0.7528	-0.0020
3	0.7777	0.7670	0.0107
4	0.8914	0.8708	0.0206
5	0.7570	0.7610	-0.0040
6	0.7827	0.7888	-0.0061
7	0.7895	0.8026	-0.0131
8	0.7695	0.7520	0.0175

9	0.7878	0.8002	-0.0124
10	0.7788	0.7930	-0.0142
11	0.7887	0.7994	-0.0107

Table S5. Statistical data on fitted values and errors of Manganese leaching rates in Mixing Experiments.

No	Observed values	Fitted value	Error (%)
1	0.7709	0.7531	0.0178
2	0.7545	0.7573	-0.0028
3	0.7762	0.7642	0.0120
4	0.8914	0.8660	0.0254
5	0.7751	0.7801	-0.0050
6	0.7843	0.7897	-0.0054
7	0.7798	0.7958	-0.0160

8	0.7792	0.7580	0.0212
9	0.7803	0.7998	-0.0195
10	0.7776	0.7925	-0.0149
11	0.7864	0.7991	-0.0127

Table S6. Statistical data on fitted values and errors of Cobalt leaching rates in Mixing Experiments.

No	Observed values	Fitted value	Error
1	0.8007	0.7951	0.0056
2	0.8054	0.8066	-0.0012
3	0.8299	0.8225	0.0074
4	0.8910	0.8781	0.0129
5	0.7815	0.7847	-0.0032
6	0.8296	0.8337	-0.0041
7	0.8183	0.8258	-0.0075

Table S7. Statistical data on fitted values and errors of Lithium leaching rates in Mixing Experiments.

No	Observed values	Fitted value	Error
1	0.8232	0.8456	-0.0224
2	0.8694	0.8671	0.0023
3	0.8720	0.8829	-0.0109
4	0.7535	0.7769	-0.0234
5	0.7861	0.7830	0.0031
6	0.8537	0.8470	0.0067
7	0.8481	0.8314	0.0167
8	0.8146	0.8263	-0.0117
9	0.8456	0.8174	0.0282
10	0.8338	0.8232	0.0106
11	0.8189	0.8180	0.0009
8	0.8072	0.7927	0.0145
9	0.8303	0.8328	-0.0025
10	0.8172	0.8287	-0.0115
11	0.8220	0.8324	-0.0104

Table S8. Data statistics for Uniform Experimental Design Nickel fitted values and relative errors.

Samples	Observed values	Fitted value	Error	Relative error (%)
1	0.9873	0.9642	0.0231	2.3360
2	0.7809	0.7578	0.0231	2.9535
3	0.9618	0.9821	-0.0203	2.1149
4	0.7764	0.8423	-0.0659	8.4874
5	0.7343	0.7224	0.0119	1.6192
6	0.9922	0.9691	0.0231	2.3245
7	0.7677	0.7880	-0.0203	2.6496
8	0.9888	0.9941	-0.0053	0.5361
9	0.9497	0.9327	0.0170	1.7949
10	0.7412	0.7615	-0.0203	2.7444
11	0.7645	0.7475	0.0170	2.2298
12	0.7781	0.7611	0.0170	2.1908

Table S9. Data statistics for Uniform Experimental Design Cobalt fitted values and relative errors.

Samples	Observed values	Fitted value	Error	Relative error (%)
1	0.9896	0.9664	0.0232	2.3462
2	0.7844	0.7612	0.0232	2.9599
3	0.9655	0.9844	-0.0189	1.9592
4	0.7791	0.8454	-0.0663	8.5144
5	0.7388	0.7263	0.0125	1.6905
6	0.9943	0.9711	0.0232	2.3351
7	0.7706	0.7895	-0.0189	2.4548
8	0.9921	0.9980	-0.0059	0.5903
9	0.9449	0.9293	0.0156	1.6509
10	0.7361	0.7550	-0.0189	2.5698
11	0.7607	0.7451	0.0156	2.0507
12	0.7796	0.7640	0.0156	2.0010

Table S10. Data statistics for Uniform Experimental Design Manganese fitted values and relative errors.

Samples	Observed values	Fitted value	Error	Relative error (%)
1	0.9995	0.9811	0.0184	1.8374
2	0.8177	0.7993	0.0184	2.2459
3	0.9991	1.0199	-0.0208	2.0831
4	0.8188	0.8713	-0.0525	6.4082
5	0.7758	0.7679	0.0079	1.0221
6	0.9999	0.9815	0.0184	1.8366
7	0.8084	0.8292	-0.0208	2.5745
8	0.9998	1.0025	-0.0027	0.2683
9	0.9987	0.9805	0.0182	1.8213
10	0.7924	0.8132	-0.0208	2.6265
11	0.8182	0.8000	0.0182	2.2230
12	0.8218	0.8036	0.0182	2.2133

Table S11. Data statistics for Uniform Experimental Design Lithium fitted values and relative errors.

Samples	Observed values	Fitted value	Error	Relative error (%)
1	0.9995	0.9811	0.0184	1.8374
2	0.8177	0.7993	0.0184	2.2459
3	0.9991	1.0199	-0.0208	2.0831
4	0.8188	0.8713	-0.0525	6.4082
5	0.7758	0.7679	0.0079	1.0221
6	0.9999	0.9815	0.0184	1.8366
7	0.8084	0.8292	-0.0208	2.5745
8	0.9998	1.0025	-0.0027	0.2683
9	0.9987	0.9805	0.0182	1.8213
10	0.7924	0.8132	-0.0208	2.6265
11	0.8182	0.8000	0.0182	2.2230
12	0.8218	0.8036	0.0182	2.2133

Table S12. Raw data on Al and Cu concentration in leached solution

Samp	No 1. (ppm)	No 2. (ppm)	No 3. (ppm)	Average values（ppm）
Al	0.066	0.069	0.066	0.067
Cu	0.011	0.010	0.009	0.010

Table S13. Statistical and quantitative comparison of cost, dosage, and toxicity of DES and its components.

DES components	Unit cost of reagents ($/g)	Reagent usage (g)	Synthetic DES cost ($)	Toxicity and characterization	Ref

The concentrations of Al and Cu were tested under the optimal leaching conditions, and the average values were 0.067 ppm and 0.010 ppm for each group of tests repeated three times (Table S12). These results indicate that only a minimal amount of Al and Cu remained in the cathode powder during the pretreatment process.

ChCl-EG	0.043$/g	5g	0.21$	EG:Low toxicity for animals	1
ChCl-LA	0.057$/g	7.5g	0.4275$	LA:Non-toxicity	2
BeCl-EG	0.043$/g	20g	0.37$	EG:Low toxicity for animals	3
BeCl-OxA	0.26$/g	/	/	OxA:Highly toxic and strongly corrosive	4
BeCl-CA	0.091$/g	14g	1.27$	CA:Non-toxicity	5
EG-SAD	0.043$/g	30ml	1.28$	SAD:Toxic and causes water pollution	6
DMT/OxA/H_2O	0.28$/g	14g	3.85$	OxA:Highly toxic and strongly corrosive	7
DMSP-VC-H_2O	0.083$/g	2.5g	0.16$	VC:Non-toxicity	**(This work)**

References

1. M. K. Tran, M.-T. F. Rodrigues, K. Kato, G. Babu and P. M. J. N. E. Ajayan, 2019, **4**, 339-345.
2. Y. Tian, W. Chen, B. Zhang, Y. Chen, R. Shi, S. Liu, Z. Zhang and T. J. C. Mu, 2022, **15**, e202200524.
3. Y. Luo, C. Yin, L. Ou and C. J. G. C. Zhang, 2022, **24**, 6562-6570.
4. X. Chang, M. Fan, C. F. Gu, W. H. He, Q. Meng, L. J. Wan and Y. G. J. A. C. I. E. Guo, 2022, **61**, e202202558.
5. Y. Luo, L. Ou and C. J. W. M. Yin, 2023, **164**, 1-8.
6. S. Tang, M. Zhang, M. J. A. S. C. Guo and Engineering, 2022, **10**, 975-985.
7. Y. Luo, C. Yin and L. J. S. o. t. T. E. Ou, 2023, **902**, 166095.