Supporting Information

Machine learning models accelerate deep eutectic solvents discovery for the recycling of Lithium-ion battery cathodes

Fengyi Zhou,[‡,a] Dingyi Shi,[‡,b] Wenbo Mu,[c] Shao Wang,[a] Zeyu Wang,[a] Chenyang Wei,[a] Ruiqi Li,[*,b] Tiancheng Mu[*,a]

[a] Department of Chemistry, Renmin University of China, Beijing 100872, China.

[b] College of Information Science and Technology, Beijing University of Chemical Technology, Beijing 100029, China.

[c] Department of Computer Science and Engineering, University of California San Diego, La Jolla, CA, 92093-0404, USA.

Department of Chemistry, Renmin University of China, Beijing 100872, China. Email: zhoufengyi@ruc.edu.cn

College of Information Science and Technology, Beijing University of Chemical Technology, Beijing 100029, China. Email: sdy@buct.edu.cn

Department of Computer Science and Engineering, University of California San Diego, La Jolla, CA, 92093-0404, USA. Email: wmu@ucsd.edu (WM)

Department of Chemistry, Renmin University of China, Beijing 100872, China. Email: wangzeyu@ruc.edu.cn

Department of Chemistry, Renmin University of China, Beijing 100872, China. Email: *wangshao@ruc.edu.cn*

College of Information Science and Technology, Beijing University of Chemical Technology, Beijing 100029, China. Email: lir@buct.edu.cn (RL)

Department of Chemistry, Renmin University of China, Beijing 100872, China. Email: tcmu@ruc.edu.cn (TM)

‡These authors contributed equally.

Table S1 Electrochemical data of DESs in LCO dataset.

DES	ratio	E_m	E_{re}	E
ChCl : Glycol	1:1	0.41	0.365	0.045
ChCl : Glycol	1:1.5	0.39	0.32	0.070
ChCl : Glycol	1:2	0.36	0.355	0.005
ChCl : Glycol	1:4	0.28	0.35	-0.070
ChCl : Ethylene glycol	1:2	0.29	0.34	-0.050
ChCl : Ethylene glycol	1:3	0.27	0.34	-0.070
ChCl : Ethylene glycol	1:4	0.2	0.335	-0.135
ChCl : Urea	1:2	0.15	0.325	-0.175
ChCl : Oxalic acid	1:1	0.95	0.355	0.595
PTSA·H_2O:ChCl	1:1	1.02	0.37	0.650
PTSA·H_2O:ChCl:H_2O	1:1:1	0.99	0.35	0.640
PTSA·H_2O:ChCl:H_2O	1:1:2	0.98	0.33	0.650
ChCl : L-ascorbic acid : H_2O	2:1:0	0.18	0.325	-0.145
ChCl : : L-ascorbic acid : H_2O	2:1:2	-0.07	0.285	-0.355
ChCl : : L-ascorbic acid : H_2O	2:1:4	-0.076	0.25	-0.326
ChCl : : L-ascorbic acid : H_2O	2:1:6	-0.11	0.215	-0.325
ChCl : PTSA (ρ-toluenesulfonic acid)	1:1	1.02	0.31	0.710
ChCl : Malonic aicd	1:1	0.87	0.36	0.510
ChCl : Formic acid	1:2	0.78	0.275	0.505
ChCl : acetic acid	1:2	0.64	0.345	0.295
ChCl : propionic acid	1:2	0.63	0.365	0.265
ChCl : n-butyric acid	1:2	0.48	0.375	0.105
ChCl : Formic acid : H_2O	1:2:1.4	0.88	0.285	0.595
ChCl : Formic acid : H_2O	1:2:3.2	0.87	0.255	0.615
ChCl : Formic acid : H_2O	1:2:5.5	0.81	0.235	0.575
ChCl : Formic acid : H_2O	1:2:8.6	0.685	0.215	0.470
ChCl : L-(+)-tartaric acid:H_2O	1:1:1.1	0.87	0.27	0.600
ChCl : L-(+)-tartaric acid:H_2O	1:1:0	0.88	0.33	0.550
ChCl : L-(+)-tartaric acid:H_2O	1:1:2.2	0.86	0.25	0.610
ChCl : L-(+)-tartaric acid:H_2O	1:1:3.3	0.86	0.24	0.620
ChCl : succinic acid : EG	1:1:1	0.76	0.315	0.445
Guanidine hydrochloride : lactic acid	1:2	0.87	0.265	0.605
Ethylene glycol : sulfosalicylic acid dihydrate	8:1	0.85	0.32	0.530
Ethylene glycol : sulfosalicylic acid dihydrate	12:1	0.83	0.335	0.495
Ethylene glycol : sulfosalicylic acid dihydrate	10:1	0.83	0.33	0.500
Ethylene glycol : monohydrate citric acid	1:1	0.645	0.245	0.400
Ethylene glycol : monohydrate citric acid	1.5:1	0.64	0.23	0.410
Ethylene glycol : monohydrate citric acid	2:1	0.57	0.235	0.335
Ethylene glycol : monohydrate citric acid	2.5:1	0.57	0.22	0.350
Ethylene glycol : monohydrate citric acid	3:1	0.51	0.245	0.265
Ethylene glycol : monohydrate citric acid	3.5:1	0.49	0.245	0.245
PEG200 : PTSA·H_2O	1:1	1.031	0.28	0.751

$E=E_m-E_{re}$, E_m is the measured potential in DES, E_{re} is the potential of internal reference.

Table S2 Electrochemical data of DESs in DES dataset.

DES	ratio	E_m	E_{re}	E
ChCl : Glutaric acid	1:1	0.78	0.315	0.465
ChCl : 1,2-Propanediol	1:2	0.38	0.395	-0.015
ChCl : 1,2-Propanediol	1:3	0.18	0.400	-0.220
ChCl : 1,2-Propanediol	1:4	0.20	0.400	-0.200
ChCl : 1,2-Propanediol	1:5	0.23	0.400	-0.170
ChCl : 1,2-Propanediol	1:6	0.32	0.395	-0.075
ChCl :1,4-Butanediol	1:3	0.44	0.395	0.045
ChCl :1,4-Butanediol	1:4	0.42	0.395	0.025
ChCl :1,4-Butanediol	1:5	0.40	0.395	0.005
ChCl :1,4-Butanediol	1:6	0.33	0.390	-0.060
ChCl : Phenol	1:2	0.23	0.273	-0.043
ChCl : Phenol	1:3	0.28	0.260	0.020
ChCl : Phenol	1:4	0.48	0.260	0.220
ChCl : Phenol	1:5	0.47	0.280	0.190
ChCl : Phenol	1:6	0.49	0.270	0.220
ChCl : o-Cresol	1:2	0.41	0.293	0.118
ChCl : o-Cresol	1:3	0.49	0.295	0.195
ChCl : o-Cresol	1:4	0.49	0.270	0.220
ChCl : o-Cresol	1:5	0.49	0.258	0.233
ChCl : o-Cresol	1:6	0.49	0.275	0.215
Betaine : Lactic acid : H_2O	1:2:2	0.566	0.275	0.291
Histidine : Lactic acid : H_2O	1:9:7	0.41	0.285	0.125
Betaine : DL-Malic Acid : H_2O	2:1:3	0.616	0.229	0.388
ChCl : Fructose:H_2O	1:1:1	0.46	0.320	0.140
ChCl : Xylitol	1:1	0.58	0.350	0.230
ChCl : Triethylene glycol	1:3	0.313	0.350	-0.037
ChCl : Triethylene glycol	1:4	0.21	0.380	-0.170
ChCl : Triethylene glycol	1:5	0.474	0.390	0.084
ChCl : Triethylene glycol	1:6	0.49	0.400	0.090
N,N-diethylethanolammonium chloride : Glycerol	1:2	0.54	0.335	0.205
N,N-diethylethanolammonium chloride: Ethylene glycol	1:2	0.28	0.395	-0.115
ChCl : Glycolic acid	1:1	0.86	0.355	0.505
ChCl : Levulinic acid	1:2	0.85	0.390	0.460
ChCl : Levulinic acid	1:3	0.90	0.400	0.500
Tetraethylammonium chloride : Levulinic acid	1:3	0.76	0.435	0.325
Trimethylamine hydrochloride : Levulinic acid	1:3	0.81	0.405	0.405

$E=E_m-E_{re}$, E_m is the measured potential in DES, E_{re} is the potential of internal reference.

Table S3 Data of LIB dataset collected from previous work.

DESs	ratio	L:S /g·g^{-1}	T range /℃	Time range /h	No. of data points	Ref.
ChCl : Glycol	1:1, 1:1.5, 1:2, 1:4	50	120-200	1-20	54	[1]
ChCl : EG	1:2, 1:3, 1:4	89.4 , 50	25-190	1-72	68	[2, 3]
ChCl : Urea	1:2	50	150-180	1-24	70	[4]
ChCl : Oxalic acid	1:1	50 , 150	30-110	0.083-8	87	[5, 6]
PTSA·H_2O : ChCl:H2O	1:1:0, 1:1:2, 1:1:4, 1:1:6	37.85, 19.23, 9.61, 3.83, 2.39	50-120	0.083-2.5	25	[7]
ChCl : L-ascorbic acid:H2O	2:1:0, 2:1:2, 2:1:4, 2:1:6	25	30-70	0.25-1.5	25	[8]
ChCl : PTSA	1:1	60.54	25-150	6-72	13	[9]
ChCl : Malonic aicd	1:1	60.54	25-150	6-72	13	[9]
ChCl : Formic acid	1:2	50	30-90	12	7	[10]
ChCl : acetic acid	1:2	50	30-90	12	7	[10]
ChCl : propionic acid	1:2	50	30-90	12	7	[10]
ChCl : n-butyric acid	1:2	50	30-90	12	7	[10]
ChCl : Formic acid : H_2O	1:2:1.4, 1:2:3.2, 1:2:5.5, 1:2:8.6	50	60-100	0.083-0.5	18	[11]
ChCl : L-(+)-tartaric acid:H_2O	1:1:0, 1:1:1.1, 1:1:2.2, 1:1:3.3	27.78. 38.46, 40, 50, 66.67	70	0.25-21	28	[12]
ChCl : succinic acid : EG	1:1:1	300	90-150	0.5-16	36	[13]
EG : sulfosalicylic acid dihydrate	8:1, 12:1, 10:1	10, 15, 17.14, 20, 24, 30	70-110	2-12	34	[14]
EG : monohydrate citric acid	1:1, 1.5:1, 2:1, 2.5:1, 3:1, 3.5:1	84	70-100	1-12	58	[15]
PEG200 : PTSA·H_2O	1:1	50	25-120	0.17-96	10	[16]
Guanidine hydrochloride : lactic acid	1:2	50	25-80	0.5-24	17	[17]

Table S4 Data of MO dataset collected from previous work.

DESs	ratio	L:S	T range /℃	Time /h	MO	No. Of data points	Ref.
ChCl: Sulphosalicylic acid	7:3, 3:7, 1:1, 4:1	25	60-100	0.083-5	CuO, ZnO, CoO, Fe_2O_3	92	[18]
ChCl:EG	1:2	30,50	50	1-72	FeO, Fe_3O_4, CoO, Cu_2O, CuO, MnO	26	[19]
ChCl:oxaclic $2H_2O$	1:1	30	50	1-72	MnO, Fe_2O_3, Fe_3O_4, CoO, CuO, Cu_2O, ZnO, PbO	20	[19]
ChCl:Levinulic acid	1:2	30	50	1-72	CoO, MnO	12	[19]
ChCl:PTSA	1:2, 2:1, 1:1	20	50	24	MnO, Fe_2O_3, Fe_3O_4, CuO Cu_2O, ZnO	18	[20]
ChCl:malonic acid	1:1	50	50	48	MnO, FeO, Fe_3O_4, CoO, CuO, Cu_2O, ZnO,	7	[21]
TEAB:CAA	1:2	20	50	24	Fe_3O_4, CuO, ZnO, PbO	4	[22]
TPAB:CAA	1:2	20	50	24	Fe_3O_4, CuO, ZnO, PbO	4	[22]
TBAB:CAA	1:1, 1:2	20	50	24	Fe_3O_4, CuO, ZnO, PbO	8	[22]
TBAHS:CAA	1:1, 1:2	20	50	24	Fe_3O_4, CuO, ZnO, PbO	8	[22]

Table S5 The ranges of the grid-search for each algorithm in training the models

Algorithm	Hyperparameter	Range
Support Vector Regression	C	0.1,0.2,0.3,0.5, 1,2,3,10,20
	gamma	1, 0.1, 0.01,0.001
Random Forest	n_estimators	5,10,20,50,70,80,100
	max_depth	2,3,5,7,9,10,20
	max_features	0.6,0.7,1
Extreme Gradient boosting	n_estimators	5, 10, 20, 50, 70, 100, 200
	max_depth	5, 6, 7, 8, 9, 10
	max_delta_step	1, 3, 5, 7
	subsample	0.4, 0.6, 0.8, 1
	colsample_bytree	0.5, 0.7, 0.9

Table S6 Prediction results on the training set for the four model inputs with different feature groups.

Feature group	model	R^2	MSE
0	SVR	0.6759	0.0057
	RForest	0.9935	0.0001
	XGBoost	0.9938	0.0001
	NN	0.8184	0.0032
1	SVR	0.6918	0.0054
	RForest	0.9948	0.0001
	XGBoost	0.9939	0.0001
	NN	0.9869	0.0002
2	SVR	0.6954	0.0053
	RForest	0.9596	0.0007
	XGBoost	0.9609	0.0007
	NN	0.9004	0.0017
3	SVR	0.7020	0.0025
	RForest	0.9598	0.0007
	XGBoost	0.9611	0.0007
	NN	0.9333	0.0012
4	SVR	0.7318	0.0047
	RForest	0.8910	0.0019
	XGBoost	0.8939	0.0019
	NN	0.8842	0.0020

Table S7 The performance of different feature sets on the XGBoost model.

Feature set		R^2	MSE
The optimal feature set	train	0.9779	0.0005
	test	0.9072	0.0038
Without pKa of HBD	train	0.9702	0.0008
	test	0.8607	0.0032
Without logβ	train	0.9842	0.0004
	test	0.8499	0.0035
Without energy	train	0.9692	0.0008
	test	0.8721	0.0030

The optimal feature set includes L:S, logβ, energy, temperature, time, pKa of HBD, water content, MW-HBD, MW-HBA, and number of O.

Table S8 Experimental values of Co solubility and std deviations of LCO leaching experiments in different DESs predicted by the CGAN model.

DES	Co-1 (mg/g)	Co-2 (mg/g)	Co-3 (mg/g)	Average values of Co solubility (mg/g)	Std deviation
1ChCl:1Glutaric acid	16.08	15.64	16.08	15.93	0.254
ChCl:1Glycolic acid	16.12	15.81	15.79	15.91	0.185
ChCl:3Levulinic acid	14.9	14.5	14.87	14.76	0.223
Trimethylamine hydrochloride:3Levulinic_acid	13.61	12.99	13.14	13.25	0.323
ChCl:6 O-cresol	0.32	0.24	0.23	0.26	0.048

Fig. S1 The photo of the working electrode used in electrochemical experiments.

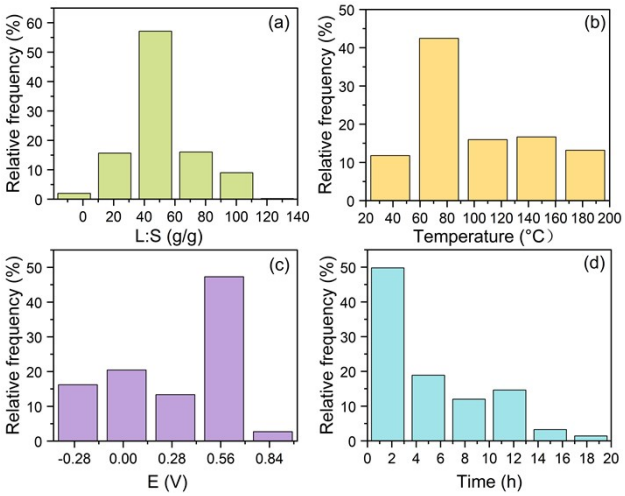

Fig. S2 The distribution of the inputs, (a)the ratio of liquid to solid, L:S, (b)leaching temperature, (c)E(reaction potential), (d)leaching time.

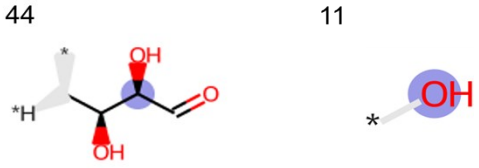

Fig. S3 Structure 44 and structure 11 are obtained by the Morgan fingerprint.

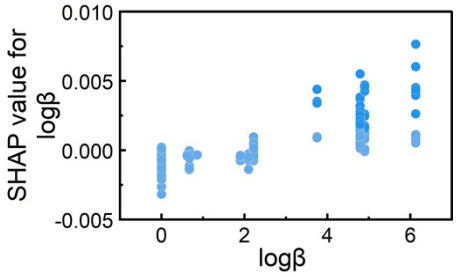

Fig. S4 The dependence plots of SHAP values with logβ.

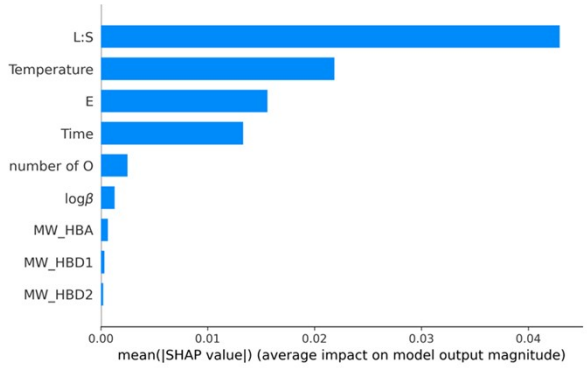

Fig.S5 The Bar chart of mean importance of different feature

Referenc

e

1. H. Yu, S. Wang, Y. Li, Q. Qiao, K. Wang and X. Li, *Green Process. Synth.*, 2022, **11**, 868-874.
2. N. Peeters, K. Janssens, D. de Vos, K. Binnemans and S. Riaño, *Green Chem.*, 2022, **24**, 6685-6695.
3. K. Wang, T. Hu, P. Shi, Y. Min, J. Wu and Q. Xu, *ACS Sustain. Chem. Eng.*, 2022, **10**, 1149-1159.
4. S. Wang, Z. Zhang, Z. Lu and Z. Xu, *Green Chem.*, 2020, **22**, 4473-4482.
5. Q. Lu, L. Chen, X. Li, Y. Chao, J. Sun, H. Ji and W. Zhu, *ACS Sustain. Chem. Eng.*, 2021, **9**, 13851-13861.
6. T. Li, Y. Xiong, X. Yan, T. Hu, S. Jing, Z. Wang and X. Ge, *J. Energy Chem.*, 2022, **72**, 532-538.
7. M. J. Roldan-Ruiz, M. L. Ferrer, M. C. Gutierrez and F. del Monte, *ACS Sustain. Chem. Eng.*, 2020, **8**, 5437-5445.
8. Y. Hua, Y. Sun, F. Yan, S. Wang, Z. Xu, B. Zhao and Z. Zhang, *Chem. Eng. J.*, 2022, **436**, 133200.
9. B. Lu, R. Du, G. Wang, Y. Wang, S. Dong, D. Zhou, S. Wang and C. Li, *Environ. Res.*, 2022, **212**, 113286.
10. L. L. Chen, Y. H. Chao, X. W. Li, G. L. Zhou, Q. Q. Lu, M. Q. Hua, H. P. Li, X. G. Ni, P. W. Wu and W. S. Zhu, *Green Chem.*, 2021, **23**, 2177-2184.
11. M. Liu, W. Ma, X. Zhang, Z. Liang and Q. Zhao, *Mater. Chem. Phys.*, 2022, **289**, 126466.
12. C. Ma, M. Svärd and K. Forsberg, *Resources, Conservation and Recycling*, 2022, **186**, 106579.
13. F. Huang, T. Li, X. Yan, Y. Xiong, X. Zhang, S. Lu, N. An, W. Huang, Q. Guo and X. Ge, *ACS Omega*, 2022, **7**, 11452-11459.
14. S. Tang, M. Zhang and M. Guo, *ACS Sustain. Chem. Eng.*, 2022, **10**, 975-985.
15. G. Zeng, J. Yao, C. Liu, X. Luo, H. Ji, X. Mi and C. Deng, *ACS Sustain. Chem. Eng.*, 2021, **9**, 16133-16142.
16. Y. Chen, Y. Wang, Y. Bai, Y. Duan, B. Zhang, C. Liu, X. Sun, M. Feng and T. Mu, *ACS Sustain. Chem. Eng.*, 2021, **9**, 12940-12948.
17. Y. Tian, W. Chen, B. Zhang, Y. Chen, R. Shi, S. Liu, Z. Zhang and T. Mu, *ChemSusChem*, 2022, **15**, e202200524.
18. I. V. Zinov'eva, A. Y. Fedorov, N. A. Milevskii, Y. A. Zakhodyaeva and A. A. Voshkin, *Theor. Found. Chem. Eng.*, 2021, **55**, 371-379.
19. I. M. Pateli, D. Thompson, S. S. M. Alabdullah, A. P. Abbott, G. R. T. Jenkin and J. M. Hartley, *Green Chem.*, 2020, **22**, 5476-5486.
20. N. R. Rodriguez, L. Machiels and K. Binnemans, *ACS Sustain. Chem. Eng.*, 2019, **7**, 3940-3948.
21. A. P. Abbott, G. Capper, D. L. Davies, K. J. McKenzie and S. U. Obi, *J. Chem. Eng. Data*, 2006, **51**, 1280-1282.
22. K. A. Omar and R. Sadeghi, *J. Mol. Liq.*, 2022, **347**.