Supplementary Information

RedCat, an Automated Discovery Workflow for Aqueous Organic Electrolytes

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S1 Thermodynamic principle

The thermodynamic mechanism of organic molecules in redox flow batteries can be described as follows:

$$\mathrm{M} \;+\; 2\mathrm{H}^{+} \;+\; 2\mathrm{e}^{-} \;\rightarrow\; \mathrm{MH_2} \tag{S1}$$

where M, H^+ and MH_2 denote the organic molecule, proton, and hydrogenated molecule, respectively. According to the Nernst equation, the redox potential (E^{o}) can be given by:

$$E^{o} \;=\; -\frac{\Delta G_{\mathrm{rxn}}}{nF} \tag{S2}$$

where ΔG_{rxn}, represents the Gibbs free energy difference between the reactant and product. n and F represent the number of transferred electrons and Faraday constant, respectively. ΔG_{rxn} can be expressed as:

$$\Delta G_{\mathrm{rxn}} \;=\; \Delta E_{\mathrm{rxn}} \;+\; p\Delta V_{\mathrm{rxn}} \;-\; T\Delta S_{\mathrm{rxn}} \;+\; \Delta(ZPE) \tag{S3}$$

where ΔE_{rxn} is the internal energy difference, namely reaction energy. ΔV_{rxn}, ΔS_{rxn}, and $\Delta(ZPE)$ denote the changes in volume, entropy, and zero-point energy, respectively. p and T indicate pressure and temperature, respectively. The terms $p\Delta V_{\mathrm{rxn}}$ and $T\Delta S_{\mathrm{rxn}}$ are much smaller than ΔE_{rxn} [S1, S2] and therefore ignored in the equation. Additionally, $\Delta(ZPE)$ is sufficiently small to be neglected especially in high-throughput computational screening studies [S3–S5]. Thus, the redox potential can be approximated as:

$$E^{o} \;=\; -\frac{\Delta E_{\mathrm{rxn}}}{nF} \tag{S4}$$

where ΔE_{rxn} can be shown as:

$$\Delta E_{\mathrm{rxn}} \;=\; E_{\mathrm{MH_2}} \;-\; E_{\mathrm{M}} \;-\; E_{\mathrm{H_2}} \tag{S5}$$

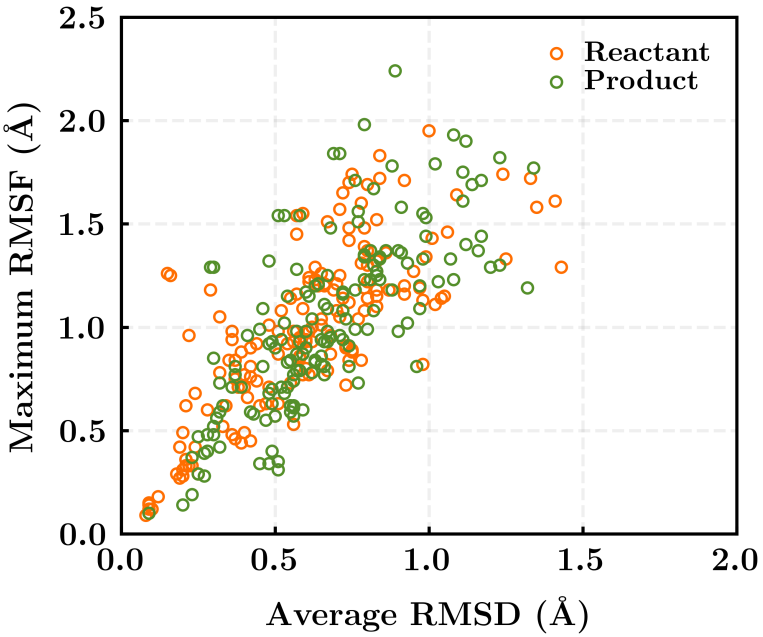

Fig. S1 A 2D scatter plot illustrating the distribution of 160 molecules based on their maximum RMSF values and average RMSD values. The orange circles represent reactant molecules, while the green circles denote product molecules.

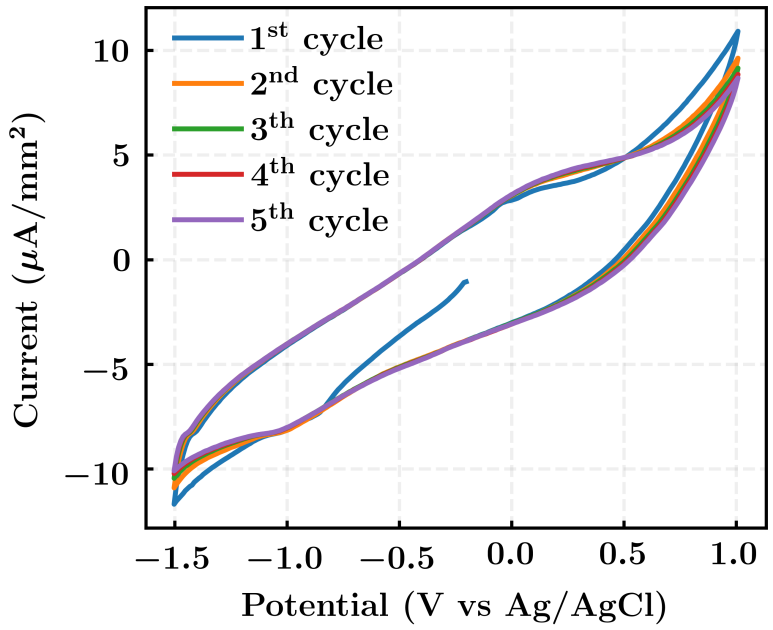

Fig. S2 Cyclic voltammograms of 1M KCl electrolyte background solution, scanned at a rate of 20 mV/s.

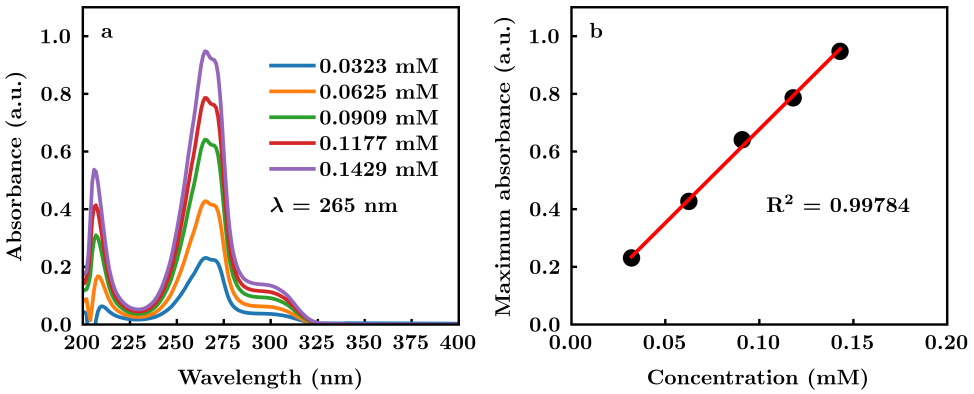

Fig. S3 (a) UV-Vis spectra of N-[(pyrazin-2-yl)methyl]formamide at various concentrations. (b) Standard calibration curve plotting absorbance at $\lambda = 265$ nm against concentration for N-[(pyrazin-2-yl)methyl]formamide.

Fig. S4 Computational screening process of 64 experimental molecules within the calibrated dataset. The screening criteria are depicted within the blue arrows, and the numbers below indicate the number of molecules that pass each screening step. Consequently, the initial 64 molecules were reduced to 3 after sequentially applying the five screening criteria outlined in the blue arrows.

Table S1: Detailed information on the computational screening of 64 experimental molecules . Tick marks indicate molecules that meet the screening criteria at each step, while crosses denote those that do not.

#	**Molecular SMILES**	**Available in PubChem database**	**Similarity based filtering**	**Reaction energy (ΔE_{rxn} ¡ 0 eV)**	**Solubility (S_{ML} ¿ 0.1 mol/L)**	**Vendor availability**
1	O=c1c(S(=O)(=O)O)cc(=O)c(c2=O)c1c(=O)c(c23)cccc3	x				
2	c1cc(F)cc(c12)nc3c(n2C)nc(=O)[nH]c3=O	x				
3	c1c(F)ccc(c12)nc3c(n2C)nc(=O)[nH]c3=O	x				
4	c1c(C)c(F)cc(c12)nc3c(n2C)nc(=O)[nH]c3=O	x				
5	c1c(Cl)c(F)cc(c12)nc3c(n2C)nc(=O)[nH]c3=O	x				
6	COc(c1)ccc(c12)n(C)c3c(n2)c(=O)[nH]c(n3)=O	x				
7	c1cccc(c12)C(=O)C(=C(C2=O)O)CC=C(C)C	✓	x			
8	c1cccc(c12)C(=O)C(O)=CC2=O	✓	x			
9	O=C1C(=O)C=CC=C1	✓	x			
10	O=C1C(=O)C=Cc(c12)cccc2	✓	x			
11	O=C1C=CC(=O)C=C1	✓	x			
12	OC1=CC(=O)C(O)=CC1=O	✓	x			
13	O=C1C(O)=C(Cl)C(=O)C(=C1Cl)	✓	x			
14	O=C1C(Cl)=C(Cl)C(=O)C(Cl)=C1C	✓	x			
15	O=C1C=CC(=O)C(O)=C1	✓	x			
16	CC1=CC(=O)C=CC1=O	✓	x			
17	O=C1C=CC(=O)C(Cl)=C	✓	x			
18	c1cccc(c12)c(=O)c3c(c2=O)c(=O)c(SCCS(=O)(=O)O)c(c3=O)SCCS(=O)(=O)O	✓	x			
19	c1cc(C)cc(c12)nc3c(n2C)nc(=O)[nH]c3=O	✓	x			
20	c1c(C)ccc(c12)nc3c(n2C)nc(=O)[nH]c3=O	✓	x			
21	c1c(C)c(C)cc(c12)nc3c(n2C)nc(=O)[nH]c3=O	✓	x			
22	c1cc(Cl)cc(c12)nc3c(n2C)nc(=O)[nH]c3=O	✓	x			
23	c1c(Cl)ccc(c12)nc3c(n2C)nc(=O)[nH]c3=O	✓	x			
24	c1c(Cl)c(Cl)cc(c12)nc3c(n2C)nc(=O)[nH]c3=O	✓	x			
25	c1c(Cl)c(C)cc(c12)nc3c(n2C)nc(=O)[nH]c3=O	✓	x			
26	c1c(C)c(Cl)cc(c12)nc3c(n2C)nc(=O)[nH]c3=O	✓	x			
27	c1cc(N(C)C)cc(c12)n(C)c3c(n2)c(=O)[nH]c(n3)=O	✓	x			

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#	Molecular SMILES	Available in PubChem database	Similarity based filtering	Reaction energy (ΔE_{rxn} ¡ 0 eV)	Solubility (S_{ML} ¿ 0.1 mol/L)	Vendor availability
28	c1cc(C#N)cc(c12)n(C)c3c(n2)c(=O)[nH]c(n3)=O	✓	x			
29	N#Cc(c1)ccc(c12)n(C)c3c(n2)c(=O)[nH]c(n3)=O	✓	x			
30	O=C1C=CC(=O)c(c12)cccc2	✓	✓	x		
31	c1cccc(c12)C(=O)c3c(C2=O)cccc3	✓	✓	✓	x	
32	c1cccc(c12)c3c(C(=O)C2=O)cccc3	✓	✓	✓	x	
33	c1cc(S(=O)(=O)O)cc(c12)C(=O)c3c(C2=O)ccc(c3)S(=O)(=O)O	✓	✓	✓	x	
34	c1cccc(C2=O)c1C(=O)c(c23)cc(cc3)S(=O)(=O)O	✓	✓	✓	x	
35	c1cccc(C2=O)c1C(=O)c(c23)cc(S(=O)(=O)O)c(O)c3O	✓	✓	✓	x	
36	Oc1c(S(=O)(=O)O)cc(O)c(c12)C(=O)c3c(C2=O)cccc3	✓	✓	✓	x	
37	c1c(O)ccc(c12)C(=O)c3c(C2=O)ccc(c3)O	✓	✓	✓	x	
38	c1ccc(O)c(c12)C(=O)c3c(C2=O)cccc3O	✓	✓	✓	x	
39	Oc1cccc(c12)C(=O)c3c(C2=O)cccc3O	✓	✓	✓	x	
40	Oc1c(O)ccc(c12)C(=O)c3c(C2=O)cccc3	✓	✓	✓	x	
41	Oc1ccc(O)c(c12)C(=O)c3c(C2=O)cccc3	✓	✓	✓	x	
42	c1ccc(O)c(c12)C(=O)c3c(C2=O)cc(C(=O)O)cc3O	✓	✓	✓	x	
43	c1ccc(O)c(c12)C(=O)c3c(C2=O)cc(CO)cc3O	✓	✓	✓	x	
44	c1c(C)cc(O)c(c12)C(=O)c3c(C2=O)cc(O)cc3O	✓	✓	✓	x	
45	Oc1ccc(O)c(c12)C(=O)c3c(C2=O)ccc(O)c3O	✓	✓	✓	x	
46	O=C1C(F)=C(F)C(=O)C(F)=C1F	✓	✓	✓	x	
47	O=C1C(O)=C(O)C(=O)C(O)=C1O	✓	✓	✓	x	
48	O=S(=O)(O)CCSc(c1O)c(SCCS(=O)(=O)O)c(O)c(c12)C(=O)c3c(C2=O)cccc3	✓	✓	✓	x	
49	c1ccc(S(=O)(=O)O)c(c12)C(=O)c3c(C2=O)cccc3	✓	✓	✓	x	
50	O=S(=O)(O)c(cc1)cc(c12)C(=O)c3c(C2=O)cc(S(=O)(=O)O)cc3	✓	✓	✓	x	
51	c1ccc(S(=O)(=O)O)c(c12)C(=O)C=CC2=O	✓	✓	✓	x	
52	O=S(=O)(O)c(cc1)cc(c12)C(=O)C=CC2=O	✓	✓	✓	x	
53	O=c1c(=O)c(S(=O)(=O)O)cc(c2=O)c1c(=O)c(c23)cccc3	✓	✓	✓	x	
54	c1cccc(c12)nc3c(n2C)nc(=O)[nH]c3=O	✓	✓	✓	x	

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Table S1 – Continued from previous page

#	Molecular SMILES	Available in PubChem database	Similarity based filtering	Reaction energy (ΔE_{rxn} ¡ 0 eV)	Solubility (S_{ML} ¿ 0.1 mol/L)	Vendor availability
55	c1c(C)c(C)cc(c12)nc3c(n2)c(=O)[nH]c(=O)[nH]3	✓	✓	✓	x	
56	O=C(O)c(c1)ccc(c12)nc3c(n2)c(=O)[nH]c(=O)[nH]3	✓	✓	✓	x	
57	COc(c1)c(OC)cc(c12)nc3c(n2)c(=O)[nH]c(=O)[nH]3	✓	✓	✓	x	
58	c1cc(O)cc(c12)nc3c(n2)c(=O)[nH]c(=O)[nH]3	✓	✓	✓	x	
59	c1cccc(C2=O)c1C(=O)c(c23)c(O)c(CS(=O)(=O)O)c(c3O)CS(=O)(=O)O	✓	✓	✓	✓	x
60	c1cc(S(=O)(=O)O)c(O)c(c12)C(=O)c3c(C2=O)ccc(c3O)S(=O)(=O)O	✓	✓	✓	✓	x
61	O=S(=O)(O)c(cc1)cc(c12)C(=O)c3c(C2=O)c(O)c(O)c(c3)S(=O)(=O)O	✓	✓	✓	✓	x
62	O=S(=O)(O)c(c1O)ccc(c12)C(=O)c3c(C2=O)ccc(c3O)S(=O)(=O)O	✓	✓	✓	✓	✓
63	c1ccc(S(=O)(=O)O)c(c12)C(=O)c3c(C2=O)cccc3S(=O)(=O)O	✓	✓	✓	✓	✓
64	c1ccc(S(=O)(=O)O)c(c12)C(=O)c3c(C2=O)c(S(=O)(=O)O)ccc3	✓	✓	✓	✓	✓

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