Supporting Material

The specific selection rationale for the ranges of factors in $(NH_4)_2SO_4$-$NH_3\cdot H_2O$ electrolyte system:

Initial concentration of $[Pb(NH_3)_4]^{2+}$ controlled: In the $(NH_4)_2SO_4$-$NH_3\cdot H_2O$ electrolyte system, $PbSO_4$ was selectively dissolved to generate $[Pb(NH_3)_4]^{2+}$ as the sole lead source, avoiding the introduction of extraneous anions. Calculated amounts of $PbSO_4$ were dissolved in the $(NH_4)_2SO_4$-$NH_3\cdot H_2O$ electrolyte to theoretically achieve target $[Pb(NH_3)_4]^{2+}$ concentrations. During electrolyte preparation, it was observed that when the $[Pb(NH_3)_4]^{2+}$ concentration exceeded 9 g/L, its dissolution rate significantly decreased under identical conditions, leaving a portion of $PbSO_4$ undissolved in the system. A specific $[Pb(NH_3)_4]^{2+}$ concentration was essential because it provided initial lead ions, which mitigated cathode-side lead-ion depletion and enhanced mass transfer efficiency.

$NH_3\cdot H_2O$ concentration: In our system, the concentration of $NH_3\cdot H_2O$ during electrolyte preparation was strictly controlled in accordance with the Chinese National Standard GB/T 631-2007 (Standard Test Methods for Chemical Reagent—Ammonia Solution) to ensure reagent quality. Preliminary experiments demonstrated that an $NH_3\cdot H_2O$ concentration exceeding 11 mol/L resulted in excessive reagent consumption, causing the electrolyte volume to surpass the 200 mL threshold. Conversely, when the $NH_3\cdot H_2O$ concentration fell below 7 mol/L, a significant reduction in $PbSO_4$ dissolution efficiency was observed, leaving >5 wt.% of the initial $PbSO_4$ undissolved in the electrolyte system. Therefore, the $NH_3 \cdot H_2O$ concentration in our experiments was optimized within the range of 7–11 mol/L.

Current density: At a current density of 1 mA/cm^2, the limited ion migration rate resulted in a suboptimal lead recovery rate (39.25%). Increasing the current density enhanced the driving force for $[Pb(NH_3)_4]^{2+}$ ions in the electrolyte, accelerating ion diffusion and increasing the likelihood of collisions between $[Pb(NH_3)_4]^{2+}$ and electrons, while also reducing polarization during electrodeposition. However, when the current density exceeded 9 mA/cm^2, potential polarization at both the anode and cathode led to higher overpotential, further elevating specific energy consumption (up to 3523.27 kWh/t, see Table S1). Therefore, the current density in our experiments was optimized within the range of 1–9 mA/cm^2. The specific energy consumption at varying current densities is presented in Table S1.

Table S1 Specific energy consumption at varying current densities.

Current density (mA/cm^2)	Specific energy consumption (kWh/t)
1	337.5
3	381.46
5	951.8
7	1620.86
9	3523.27

Life cycle assessment: The 18 indicators considered in this assessment include: global warming (GW, kg CO_2 eq.), stratospheric ozone depletion (SOD, kg CFC-11 eq.), ionizing radiation (IR, kBq Co-60 eq.), ozone formation (human health) (OF-HH, kg NO_X eq.), fine particulate matter formation (PMF, kg $PM_{2.5}$ eq.), ozone formation (terrestrial ecosystems) (OF-TE, kg NO_X eq.), terrestrial acidification (TA, kg SO_2 eq.), freshwater eutrophication (FE, kg P eq.), marine eutrophication (ME, kg N eq.), terrestrial ecotoxicity (TE, kg 1,4-DCB), freshwater ecotoxicity (FE, kg 1,4-DCB), marine ecotoxicity (MET, kg 1,4-DCB), human carcinogenic toxicity (HCT, kg 1,4-DCB), human non-carcinogenic toxicity (HNCT, kg 1,4-DCB), land use (LU, m^2·a crop eq.), mineral resource scarcity (MRS, kg Cu eq.), fossil resource scarcity (FRS, kg oil eq.), and water consumption (WC, m^3).

Life cycle boundaries of the $(NH_4)_2SO_4$-$NH_3 \cdot H_2O$ processes:

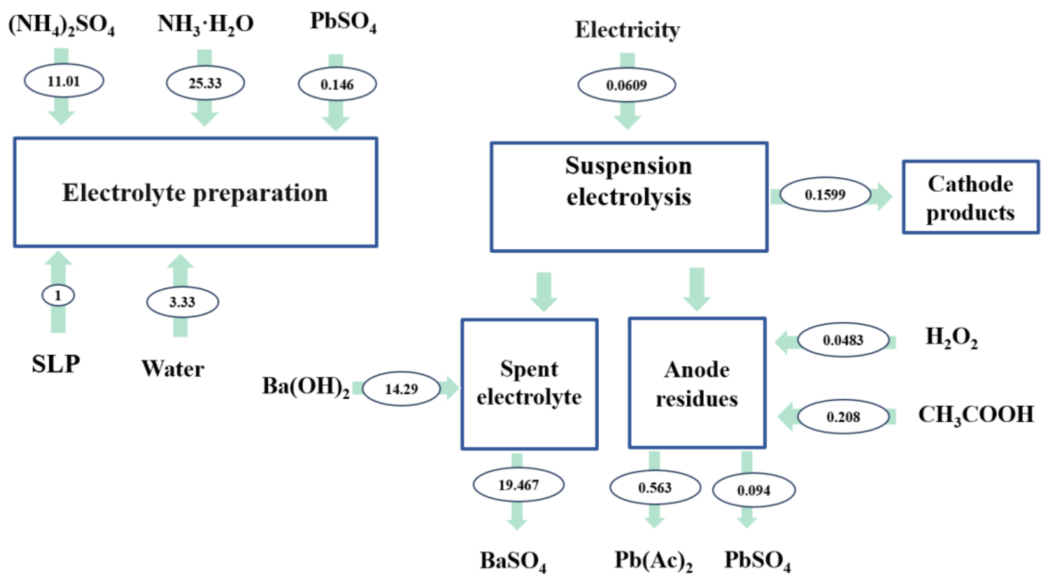

Fig. S1 Life cycle boundaries of the $(NH_4)_2SO_4$-$NH_3 \cdot H_2O$ processes.

Table S2 Life cycle inventory for recycling 1.0 kg SLP.

Items	Items	Amount	Unit	Corresponding LCI	Database	Comment
Input	Ammonium sulfate	11.01	kg	Ammonium sulfate {RoW}| market for ammonium sulfate | APOS, U	Ecoinvent 3	Avoided product/ Co-product
	Ultrapure water	3.333	kg	Water, ultrapure {RoW}| market for water, ultrapure | APOS, U	Ecoinvent 3	—
	Barium hydroxide	14.29	kg	Barium hydroxide {GLO}| market for barium hydroxide | APOS, U	Ecoinvent 3	Avoided product/ Co-product
	Acetic acid	0.208	kg	Acetic acid, without water, in 98% solution state {GLO}| market for | APOS, U	Ecoinvent 3	—
	Hydrogen peroxide	0.04837	kg	Hydrogen peroxide, without water, in 50% solution state {RoW}| market for hydrogen peroxide, without water, in 50% solution state | APOS, U	Ecoinvent 3	—
	Ammonium hydroxide	25.33	kW·h	Ammonia, steam reforming, liquid, at plant/RNA	Ecoinvent 3	—
	Spent lead paste	1	kg	Self-modeling		Avoided product/ Co-product
	Electricity	0.0609	kW·h	Electricity, low voltage {CN}| market group for | APOS, U	Ecoinvent 3	—
Output	Barium sulfate	19.4677	kg	Barium sulfide {GLO}| market for barium sulfide | APOS, U	Ecoinvent 3	Avoided product/ Co-product
	Lead	0.1599	kg	Lead {GLO}| market for | APOS, U	Ecoinvent 3	Avoided product/ Co-product
	lead sulfate	0.09403	kg	Self-modeling	Ecoinvent 3	Avoided product/ Co-product
	lead acetate	0.5636	kg	Self-modeling	Ecoinvent 3	Avoided product/ Co-product

Table S3 LCA data source for recycling 1.0 kg SLP.

Impact category	GW	SOD	IR	OF-HH	PMF	OF-TE	TA	FE	ME	TE	FET	MET	HCT	HNCT	LU	MRE	FRS	WC
Unit	kg CO_2 eq	kg CFC-11 eq	kBq Co-60 eq	kg NOx eq	kg PM2.5 eq	kg NOx eq	kg SO_2 eq	kg P eq	kg N eq	kg 1,4-DCB	kg 1,4-DCB	kg 1,4-DCB	kg 1,4-DCB	kg 1,4-DCB	$m^2 \cdot a$ crop eq	kg Cu eq	kg oil eq	m^3
Total	-119.7582206	-3.94834E-05	-6.134712378	-0.385959523	-0.226854371	-0.391763462	-0.444404058	-0.048164765	-0.008978674	-1136.317117	-27.1325114	-37.09611312	-10.46389527	-1259.380965	-2.918196132	-1.381858073	-24.88863349	-2.320571869
$(NH_4)_2SO_4$	-14.06200023	-5.14539E-06	-1.041157156	-0.048804301	-0.044751621	-0.049859251	-0.137825732	-0.011333278	-0.0071418	-694.4436745	-6.353451137	-8.501198741	-2.856562789	-171.7373156	-0.599498184	-0.686116843	-3.641285101	-0.292087192
H_2O	0.02038607	1.57119E-08	0.0017444	4.48954E-05	4.39087E-05	4.54077E-05	6.62964E-05	0.000159159	2.15421E-05	0.03201715	0.000585427	0.000787604	0.003318065	0.01613479	0.00038507	5.72354E-05	0.004972955	0.003826994
$Ba(OH)_2$	-42.97551184	-1.52389E-05	-2.837020992	-0.131065928	-0.094695046	-0.134838475	-0.191963406	-0.017465736	-0.001072886	-202.0459149	-14.08419389	-19.62978316	-2.688245048	-826.7458762	-1.032964278	-0.223898535	-10.44987385	-1.018358203
HAC	0.345228902	1.74189E-07	0.025268854	0.000865458	0.000597818	0.000923077	0.001262448	0.00016316	7.66762E-06	1.290178305	0.014559061	0.019306714	0.017929256	0.307403366	0.010508762	0.001052191	0.207538029	0.009181433
H_2O_2	0.074374769	2.16024E-08	0.002771708	0.000149640	0.000113170	0.000156037	0.000222364	5.01188E-05	4.84111E-06	0.320073626	0.004184134	0.005594077	0.009400982	0.094429209	0.001252131	0.000206012	0.022506168	0.003635561
$NH_3 \cdot H_2O$	19.3196584	1.56393E-07	0	0.016027449	0.039968159	0.016302675	0.166196426	0.000322531	0.000422185	0.284092582	0.039126229	0.050352326	0.004326596	1.747213952	0	0	4.742948924	0.038640915
SLP	-38.2069	-8.27803	-0.68049	-0.10619	-0.06163	-0.10651	-0.13935	-0.00845	-0.00052	-85.3987	-2.11774	-2.74964	-2.30831	-55.4259	-0.51176	-0.18726	-7.43927	-0.11144

	1314	E-06	1364	6778	5995	4391	8872	6439	3621	4486	5801	8672	5804	5767	3885	413	3414	9545
$BaSO_4$	- 23.3826 2778	- 6.57491 E-06	- 1.20935 0983	- 0.05890 7914	- 0.03219 8503	- 0.05969 2936	- 0.06526 5223	- 0.00670 1734	- 0.00039 4461	- 100.430 8973	- 3.31303 9466	- 4.56182 2206	- 1.15368 3045	- 169.413 5235	- 0.49350 4846	- 0.14611 4056	- 4.21356 4704	- 0.88687 9532
Pb	- 0.34242 983	- 1.31201 E-07	- 0.02365 6926	- 0.00098 0063	- 0.00108 1486	- 0.00100 6693	- 0.00264 9183	- 0.00030 5221	- 1.80865 E-05	- 8.95815 9558	- 0.16652 0979	- 0.22719 7094	- 0.22439 8217	- 7.54857 0194	- 0.01471 1668	- 0.03488 7375	- 0.08583 6936	- 0.00479 8981
$PbSO_4$	- 3.10190 782	- 6.69329 E-07	- 0.05483 5766	- 0.00862 6892	- 0.00501 5481	- 0.00865 0988	- 0.01135 0595	- 0.00068 788	- 4.26578 E-05	- 7.10537 4124	- 0.17522 802	- 0.22770 7431	- 0.19142 7493	- 4.64695 1434	- 0.04152 648	- 0.01587 7663	- 0.59883 6184	- 0.00881 4045
$Pb(AC)_2$	- 17.5134 1028	- 3.82761 E-06	- 0.31904 3223	- 0.04865 0782	- 0.02830 271	- 0.04881 4018	- 0.06397 05	- 0.00392 2366	- 0.00024 2202	- 39.9398 0101	- 0.98322 2468	- 1.27786 5024	- 1.07853 4152	- 26.0643 7686	- 0.23718 7575	- 0.08905 7671	- 3.45092 5236	- 0.05363 9086
Electricity	0.06693 2192	1.41024 E-08	0.00105 907	0.00018 5692	0.00010 3415	0.00018 6094	0.00023 1919	1.29205 E-05	8.05142 E-07	0.07908 7564	0.00243 5503	0.00306 8489	0.00229 6374	0.03642 506	0.00081 4822	4.27604 E-05	0.01299 5865	0.00016 9814

The analytical methods used to quantitatively analyze the chemical composition of raw SLP were based on standardized EDTA complexometric titration:

(1) PbO content is determined by EDTA titration in acetic acid (pH 5–6) using xylenol orange as an indicator, exploiting its selective solubility over other lead compounds.

(2) $PbSO_4$ does not react with acetic acid but is soluble in concentrated NaCl solution, where it dissociates into Pb^{2+} ions. The released Pb^{2+} is quantified via EDTA complexometric titration to determine the $PbSO_4$ content.

(3) PbO_2 is quantitatively reduced by H_2O_2 in nitric acid medium, with the excess H_2O_2 subsequently titrated against standardized $KMnO_4$ solution. Then the PbO_2 content is calculated from the $KMnO_4$ consumption based on stoichiometric relationships.

The metallic Pb content is determined by mass balance calculation, subtracting the quantified Pb species (PbO, PbO_2, and $PbSO_4$) from the total lead content obtained through inductively coupled plasma optical emission spectrometry (ICP-OES) analysis.