



Screening Deep Eutectic Solvents for Selective Lithium Recovery From Spent Battery Leachate

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Abstract

Hydrometallurgical recycling of lithium-ion batteries recovers cobalt, nickel and manganese efficiently, but lithium is commonly left to the end of the flowsheet, where it is precipitated from a dilute, sodium-rich solution with appreciable loss. Deep eutectic solvents, and hydrophobic ones in particular, have been proposed as extractants that might separate lithium from the transition metals more selectively and with less hazardous diluents. The number of possible mixtures is very large, and published tests use different feeds, phase ratios and measures of success, so that candidates cannot easily be ranked. This article proposes a staged screening procedure intended to make such ranking possible. It defines the leachate and the separation target first, assembles candidates on stated chemical grounds, and then passes them through successive tiers: physical properties and phase stability, single-metal shake tests, a synthetic mixed leachate, stripping and reuse, and finally a real leachate. For each tier the quantities to be recorded and the reasons for rejecting a candidate are specified. The procedure is a proposal built from solvent extraction practice and the published literature. It has not been applied or validated here, and the article states what would need to be shown before it could be relied on.

Keywords: Deep eutectic solvents, Lithium recovery, Battery recycling, Solvent extraction, Hydrometallurgy, Screening protocol

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1. Introduction

Spent lithium-ion batteries are both a waste problem and a metal resource. Recycling routes and their limitations have been reviewed by Harper et al. (2019) and, with emphasis on the chemistry, by Lv et al. (2018). In the hydrometallurgical route the cathode material is leached, typically in mineral or organic acid with a reducing agent, and the resulting solution is treated by precipitation and solvent extraction to separate cobalt, nickel and manganese (Chagnes and Pospiech, 2013). Lithium follows these steps in the aqueous phase and is usually recovered last, as the carbonate, from a liquor that by then contains much sodium. Recovery at that point is incomplete (Swain, 2017).

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Regulation has raised the stakes. The European Union's batteries regulation sets a target for lithium recovery from waste batteries of 50 per cent by the end of 2027 and 80 per cent by the end of 2031 (European Union, 2023). Meeting figures of that order calls for separation steps aimed at lithium itself and not only at the more valuable transition metals.

Deep eutectic solvents are one of the options under study. This article does not report tests of any solvent. It proposes a procedure by which candidate solvents can be screened in a consistent order and against stated criteria, so that results from different laboratories can be compared.

2. Background

Two bodies of knowledge underlie the procedure: the nature of deep eutectic solvents and the established practice of solvent extraction.

2.1. Deep eutectic solvents

The term was introduced by Abbott *et al.* (2003) for mixtures of choline chloride and urea, which at a 1:2 molar ratio are liquid at room temperature although both components are high-melting solids. Mixtures of choline chloride with carboxylic acids behave similarly. The general picture is of a hydrogen bond acceptor and a hydrogen bond donor whose interaction lowers the melting point, and reviews describe the range of compositions and uses (Smith *et al.*, 2014). The definition has been tightened by Martins *et al.* (2019), who argue that the name should be kept for mixtures whose melting point lies well below that predicted for an ideal mixture, a test that many so-called deep eutectic solvents have never been put to.

In battery recycling these liquids have been used in two distinct ways. Hydrophilic mixtures such as choline chloride with ethylene glycol can dissolve cathode oxides directly, as Tran *et al.* (2019) showed. That is leaching. The present article concerns the second use, liquid-liquid extraction from an aqueous leachate, which requires a solvent immiscible with water. Hydrophobic deep eutectic solvents of this kind were introduced by van Osch *et al.* (2015), who combined decanoic acid with quaternary ammonium salts, and were soon applied to the removal of alkali and transition metal ions from water (van Osch *et al.*, 2016). Mixtures designed for lithium, in which a β -diketone is paired with a neutral organophosphorus donor, have also been described (Hanada and Goto, 2021).

2.2. Solvent extraction principles

Extraction performance is expressed through a few standard quantities (Rydberg *et al.*, 2004). The distribution ratio D of a metal is its total concentration in the organic phase divided by that in the aqueous phase at equilibrium. The separation factor between two metals is the ratio of their distribution ratios. The percentage extracted depends on D and on the phase volume ratio, and should never be quoted without the latter. For lithium there are two possible strategies: extract the transition metals and leave lithium in the raffinate, or extract lithium away from everything else. The lithium ion is small, strongly hydrated and a hard acid in the sense of Pearson (1963), so that it is extracted only by hard oxygen donors and usually from neutral or alkaline solution. Any screening scheme has to state which strategy a candidate is meant to serve.

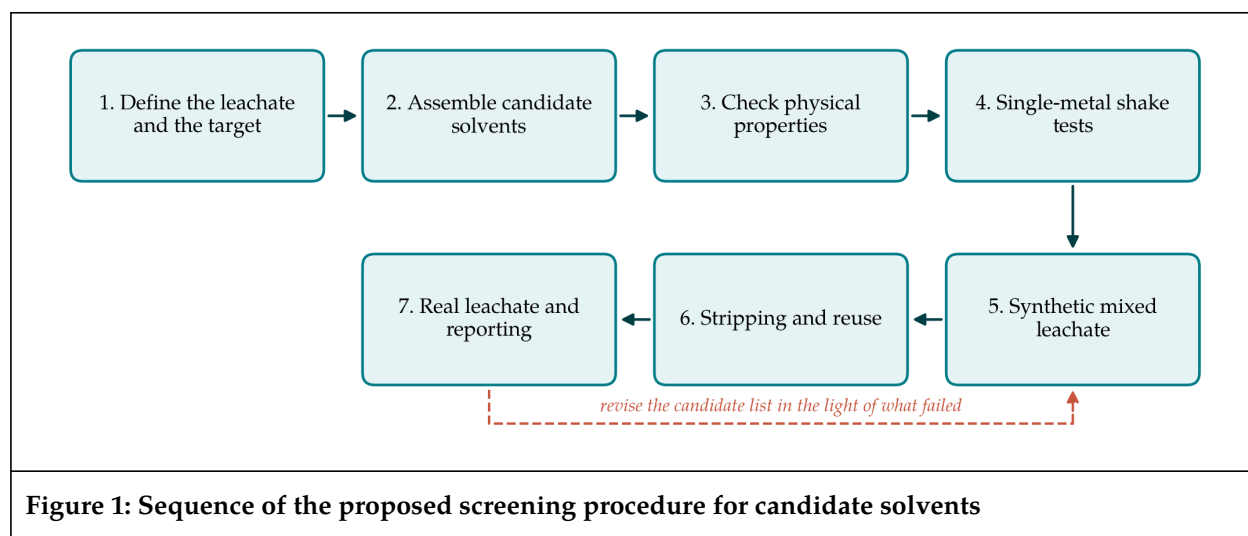
3. The Proposed Screening Procedure

The procedure has seven steps, arranged so that inexpensive tests remove unsuitable candidates before costly ones are run. Figure 1 shows the sequence and Table 1 gives, for each experimental tier, the quantity recorded and the grounds for rejection. Thresholds are left to the user, since they depend on the flowsheet in view; what the procedure fixes is that they be set before testing begins.

3.1. Step 1: Define the leachate and the target

The composition of the feed determines everything that follows. The user records the cathode chemistry or mixture assumed, the leaching acid and its residual concentration, the pH, and the expected concentrations of lithium, cobalt, nickel and manganese together with the impurities aluminium, copper and iron and any sodium introduced by neutralisation. The point in the flowsheet at which the solvent

would act is stated, as is the strategy, lithium-selective or lithium-rejecting.



3.2. Step 2: Assemble candidates on stated grounds

Candidates are chosen for reasons that can be written down: an acidic component able to exchange protons for metal ions, a neutral oxygen donor to complete the coordination of lithium, a long alkyl chain for hydrophobicity. Where resources allow, a computational prescreen with a model such as COSMO-RS (Klamt, 1995) can rank mixtures by predicted mutual solubility with water. Each candidate is prepared at a defined molar ratio, and its melting behaviour is checked so that it is known to be a liquid mixture and, if the name is to be used, a deep eutectic in the stricter sense.

3.3. Step 3: Check physical properties and phase stability

A solvent that cannot be handled is rejected before any metal is added. Viscosity and density are measured at the working temperature, since a viscous phase of density close to that of water will not disengage. The candidate is then contacted with metal-free aqueous solution at the pH of the feed, and both phases are analysed for the solvent components. This test is essential. The components of a hydrophobic eutectic mixture partition into water to different extents, so that the organic phase after contact may no longer have the composition that was prepared (Florindo et al., 2019). Loss of a component also contaminates the raffinate, and a mixture that sheds one of its constituents at this stage is set aside whatever its promise as an extractant.

Table 1: Experimental tiers of the proposed procedure, the quantity recorded at each and the grounds for rejecting a candidate

Tier	Test	Quantity recorded	Reject if
Physical	Contact with metal-free aqueous phase	Viscosity, density, component loss to water	Slow disengagement or marked leaching
Single metal	Shake test with each metal alone	Distribution ratio against pH	No pH window separates lithium
Mixed feed	Synthetic leachate; repeated loading	Separation factors; capacity; mass balance	Selectivity lost or phases unstable
Stripping	Contact loaded solvent with strip liquor	Fraction stripped; solvent composition	Metal cannot be recovered cleanly
Reuse	Several extraction and stripping cycles	Change in distribution ratio per cycle	Performance declines steadily
Real feed	Leachate from actual battery material	All of the above	Behaviour departs from synthetic feed

3.4. Step 4: Single-metal shake tests

Each metal of interest is extracted separately from a solution matching the ionic background of the feed. Equal volumes are shaken at controlled temperature until equilibrium, the time to equilibrium having been established in a preliminary run. Both phases are analysed, for example by inductively coupled plasma optical emission spectrometry after digestion or stripping of the organic phase, so that the mass balance can be closed and not inferred by difference. The equilibrium pH is measured, not the initial one. Plotting the distribution ratio against equilibrium pH for each metal shows at once whether a window exists in which lithium is separated from the rest.

3.5. Step 5: Synthetic mixed leachate

Candidates that pass are tested on a synthetic solution containing all the metals at the concentrations defined in Step 1. Competition for the extractant, changes in ionic strength and co-extraction often erode the selectivity seen with single metals. The variables that matter most, usually pH, phase ratio, temperature and the molar ratio of the solvent components, are best explored with a factorial design instead of one factor at a time, so that interactions are detected with a manageable number of runs (Montgomery, 2017). Loading capacity is determined by repeated contact of the same solvent with fresh feed, with attention to the appearance of a third phase or a precipitate.

3.6. Step 6: Stripping and reuse

An extractant that will not release the metal is of no use. The loaded solvent is contacted with a strip solution, commonly dilute acid, and the fraction of each metal returned to the aqueous phase is measured. The solvent is then analysed for its own components and returned to extraction for several cycles. A steady fall in distribution ratio, a drift in component ratio or a rise in viscosity is a ground for rejection.

3.7. Step 7: Real leachate and reporting

The few remaining candidates are tested on leachate prepared from actual spent cathode material, which contains fluoride and phosphate from the electrolyte salt, organic residues from the binder and trace metals absent from synthetic solutions. Results are reported in a fixed form: feed composition, solvent composition and water content, phase ratio, temperature, equilibrium pH, distribution ratios with their replicate scatter, separation factors and mass balance closure.

4. Fitting the Procedure to Practice

The procedure is written for a laboratory with ordinary equipment: a thermostatted shaker, a viscometer, a pH meter and access to elemental analysis. The early tiers are quick, and most candidates are expected to fail there. Two additional considerations belong alongside the technical tiers. One is hazard. A mixture is not benign because it is called a deep eutectic solvent, and the toxicity, biodegradability and flammability of each component should be looked up and recorded, in the spirit of the questions Jessop (2011) posed for green solvents generally. The other is the benchmark. Every campaign should include a conventional extractant in a hydrocarbon diluent, tested under identical conditions, because a new solvent has to be judged against what industry already uses.

Record keeping deserves the same attention as the chemistry. Each candidate should carry a single identifier through all tiers, with the supplier and stated purity of every component, the water content of the mixture as prepared, and the date of preparation, since hygroscopic mixtures change on standing. Candidates rejected at an early tier are reported together with the reason. Negative results of this kind are rarely published, yet they are what would let another laboratory avoid repeating the same unproductive tests, and a screening campaign that lists only its successes gives a misleading impression of how selective the screen was.

5. Limitations and What Remains to Be Shown

The procedure is proposed and has not been validated here. Its ordering of tiers rests on the judgement that phase stability and component leaching eliminate more candidates, more cheaply, than selectivity

tests do; that judgement should be checked by running a set of candidates through the tiers in more than one order. Whether rankings obtained with a synthetic leachate hold for real ones is likewise an empirical matter. Shake tests give equilibrium data and say nothing about behaviour in mixer-settlers or columns, where viscosity and interfacial properties govern throughput. Nor does the procedure address cost or life-cycle impact, which need separate analysis once a candidate has survived. An interlaboratory comparison on a shared synthetic feed would be the most direct test of whether the scheme delivers the comparability it aims for.

A further limitation concerns the feed itself. Cathode chemistries are changing, with nickel-rich layered oxides and lithium iron phosphate taking a growing share, and a leachate defined today may not represent the material arriving at recyclers some years from now. Iron phosphate cathodes in particular give leachates rich in iron and phosphate and poor in cobalt and nickel, for which the balance between the lithium-selective and lithium-rejecting strategies may be quite different. The procedure accommodates this through Step 1, but rankings obtained for one feed should not be carried over to another without repeating the mixed-feed tier.

6. Conclusion

Selective recovery of lithium is the weak point of hydrometallurgical battery recycling, and hydrophobic deep eutectic solvents are plausible candidates for improving it. The staged procedure described here defines the feed and target first, rejects unstable or unworkable mixtures early, measures distribution ratios against equilibrium pH with closed mass balances, and insists on stripping, reuse, a real feed and a conventional benchmark. It is offered as a framework for consistent testing, and its usefulness remains to be demonstrated in the laboratory.

Conflict of interest

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References

- Abbott, A.P., Capper, G., Davies, D.L., Rasheed, R.K. and Tambyrajah, V. (2003). [Novel solvent properties of choline chloride/urea mixtures](#). *Chemical Communications*, (1): 70-71.
<https://doi.org/10.1039/B210714G>
- Chagnes, A. and Pospiech, B. (2013). [A brief review on hydrometallurgical technologies for recycling spent lithium-ion batteries](#). *Journal of Chemical Technology & Biotechnology*, 88(7): 1191-1199.
<https://doi.org/10.1002/jctb.4053>
- European Union (2023). [Regulation \(EU\) 2023/1542 of the European Parliament and of the Council of 12 July 2023 concerning batteries and waste batteries](#). Official Journal of the European Union, L 191.
- Florindo, C., Branco, L.C. and Marrucho, I.M. (2019). [Quest for green-solvent design: From hydrophilic to hydrophobic \(deep\) eutectic solvents](#). *ChemSusChem*, 12(8): 1549-1559.
- Hanada, T. and Goto, M. (2021). [Synergistic deep eutectic solvents for lithium extraction](#). *ACS Sustainable Chemistry & Engineering*, 9(5): 2152-2160.
- Harper, G., Sommerville, R., Kendrick, E., Driscoll, L., Slater, P., Stolkin, R., Walton, A., Christensen, P., Heidrich, O., Lambert, S., Abbott, A., Ryder, K., Gaines, L. and Anderson, P. (2019). [Recycling lithium-ion batteries from electric vehicles](#). *Nature*, 575: 75-86.
<https://doi.org/10.1038/s41586-019-1682-5>
- Jessop, P.G. (2011). [Searching for green solvents](#). *Green Chemistry*, 13: 1391-1398.
<https://doi.org/10.1039/C0GC00797H>
- Klamt, A. (1995). [Conductor-like screening model for real solvents: A new approach to the quantitative](#)

- calculation of solvation phenomena. *The Journal of Physical Chemistry*, 99(7): 2224-2235. <https://doi.org/10.1021/j100007a062>
- Lv, W., Wang, Z., Cao, H., Sun, Y., Zhang, Y. and Sun, Z. (2018). A critical review and analysis on the recycling of spent lithium-ion batteries. *ACS Sustainable Chemistry & Engineering*, 6(2): 1504-1521. <https://doi.org/10.1021/acssuschemeng.7b03811>
- Martins, M.A.R., Pinho, S.P. and Coutinho, J.A.P. (2019). Insights into the nature of eutectic and deep eutectic mixtures. *Journal of Solution Chemistry*, 48: 962-982. <https://doi.org/10.1007/s10953-018-0793-1>
- Montgomery, D.C. (2017). *Design and Analysis of Experiments*, 9th edn. John Wiley & Sons, Hoboken, NJ.
- Pearson, R.G. (1963). Hard and soft acids and bases. *Journal of the American Chemical Society*, 85(22): 3533-3539. <https://doi.org/10.1021/ja00905a001>
- Rydberg, J., Cox, M., Musikas, C. and Choppin, G.R. (eds) (2004). *Solvent Extraction Principles and Practice*, 2nd edn. Marcel Dekker, New York.
- Smith, E.L., Abbott, A.P. and Ryder, K.S. (2014). Deep eutectic solvents (DESs) and their applications. *Chemical Reviews*, 114(21): 11060-11082. <https://doi.org/10.1021/cr300162p>
- Swain, B. (2017). Recovery and recycling of lithium: A review. *Separation and Purification Technology*, 172: 388-403. <https://doi.org/10.1016/j.seppur.2016.08.031>
- Tran, M.K., Rodrigues, M.-T.F., Kato, K., Babu, G. and Ajayan, P.M. (2019). Deep eutectic solvents for cathode recycling of Li-ion batteries. *Nature Energy*, 4: 339-345. <https://doi.org/10.1038/s41560-019-0368-4>
- van Osch, D.J.G.P., Parmentier, D., Dietz, C.H.J.T., van den Bruinhorst, A., Tuinier, R. and Kroon, M.C. (2016). Removal of alkali and transition metal ions from water with hydrophobic deep eutectic solvents. *Chemical Communications*, 52: 11987-11990.
- van Osch, D.J.G.P., Zubeir, L.F., van den Bruinhorst, A., Rocha, M.A.A. and Kroon, M.C. (2015). Hydrophobic deep eutectic solvents as water-immiscible extractants. *Green Chemistry*, 17: 4518-4521. <https://doi.org/10.1039/C5GC01451D>