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Exploiting Deep Eutectic Solvents in $LiFePO_4$ Battery Recycling: from Cathode Leaching to Resynthesis

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Introduction

Deep Eutectic Solvents (DESs) can be applied in many areas, thanks to their modular composition, making them versatile and potentially green. Choline Chloride-based DESs have been used in cathodes' leaching, a step followed by the separation of metals through successive precipitation and purification processes to obtain compounds for cathode resynthesis.

Our project is based on the possibility of avoiding the separation of metals from the liquid phase, exploiting a **Choline Citrate-based DES**, using the organic matrix of the solvent itself as a support during direct resynthesis of the cathodes, reducing costs and working time.

Cathode Recycling

Pyrometallurgy

High-temperature roasting process

- ✓ Simple
- ✗ Li lost in slag
- ✗ Poor quality of recovered metals
- ✗ High energy consumption
- ✗ Emission of polluting gases

Hydrometallurgy

Leaching process of the metals, to be selectively precipitated and recovered

- ✓ High purity
- ✗ High water consumption
- ✗ Wastewater treatment
- ✗ Long recovery process

Our project

DES preparation and characterization (TGA DSC IR NMR Viscosity) → Leaching of the cathode (ICP-OES) → Direct resynthesis of the cathode (TGA, XRD, SEM)

Soft solvometallurgy employing **Deep Eutectic Solvents** without using Choline Chloride-based DES

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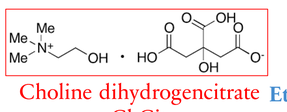
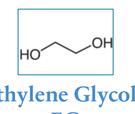
Two different DES compositions Choline Citrate-based

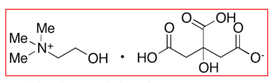


Choline dihydrogencitrate ChCit

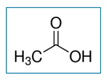


Ethylene Glycole EG

3:1 – 1:20 molar ratios
Stirring @ 90 °C → transparent liquid (60 min – 15 min)

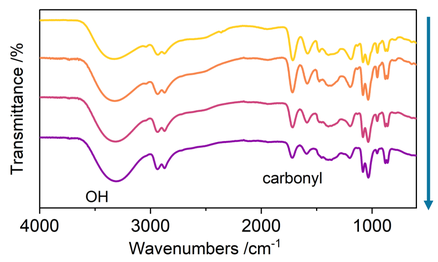


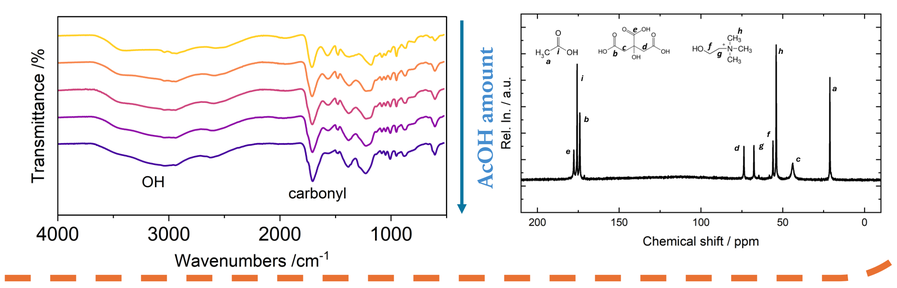
Choline dihydrogencitrate ChCit



Acetic Acid AcOH

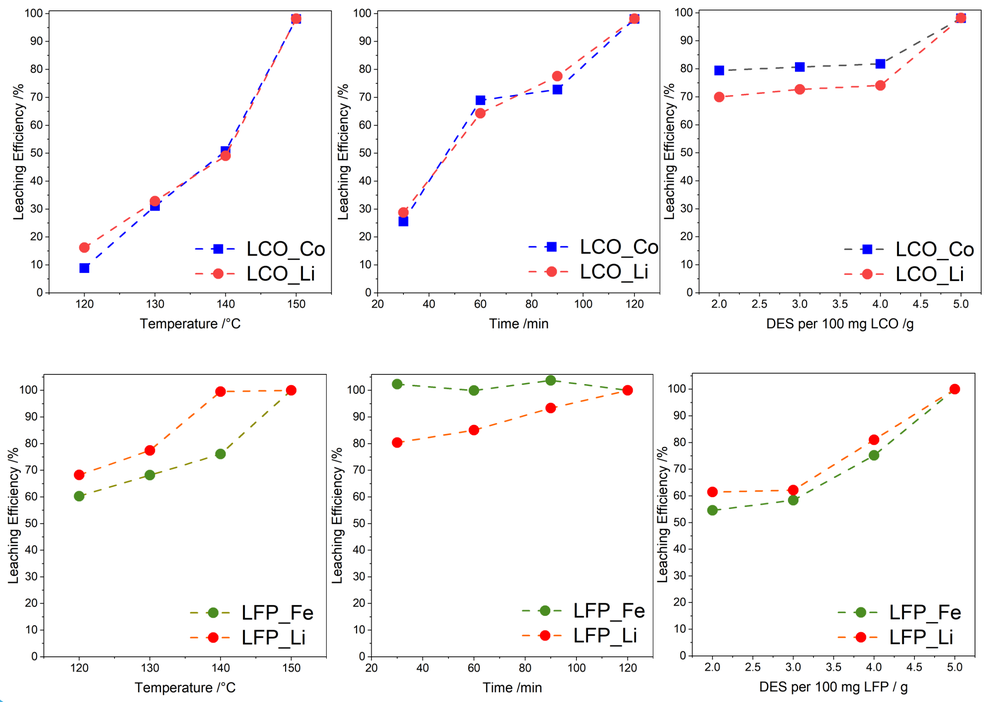
Samples	Water content (%)	Viscosity (Pa·s)
ChCit:EG_1:2	24.4	7.49
ChCit:EG_1:3	36.6	1.04
ChCit:EG_1:5	50.0	0.24
ChCit:EG_1:10	67.5	0.17
ChCit:AcOH_1:1	3.6	-
ChCit:AcOH_1:2	12.2	9.75
ChCit:AcOH_1:3	22.3	2.56
ChCit:AcOH_1:4	24.7	1.27
ChCit:AcOH_1:10	47.4	0.052





Influence of different variables:
- DES-to-cathode ratio
- Leaching temperature
- Leaching time

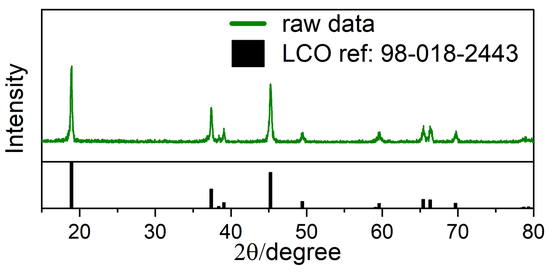
Best conditions:
- ✓ 5g of DES per 100 mg of cathode
- ✓ 150°C
- ✓ 2 hours

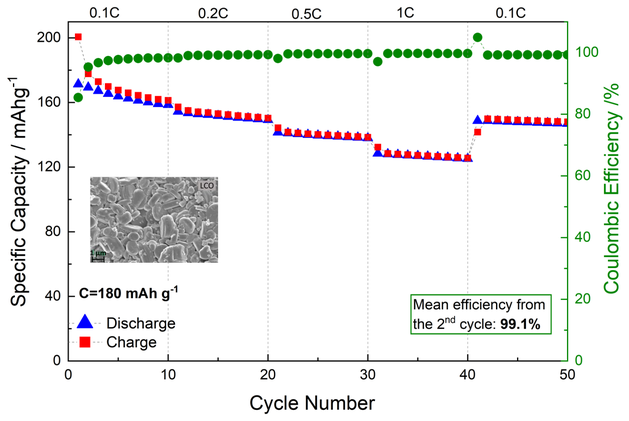


Sol-Gel Formation (160°C, 2 hours, under stirring) → **Calcination** → **Resynthesized active material**

Oxide-based cathodes

650°C, 10 hours in air in a muffle furnace

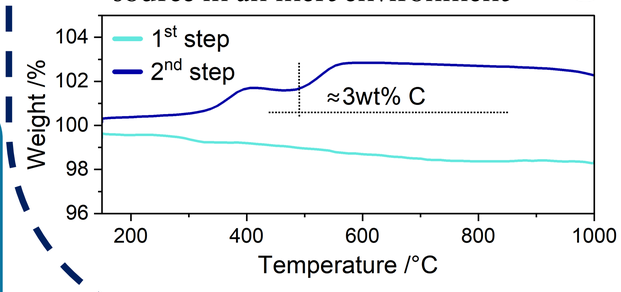


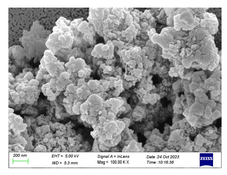


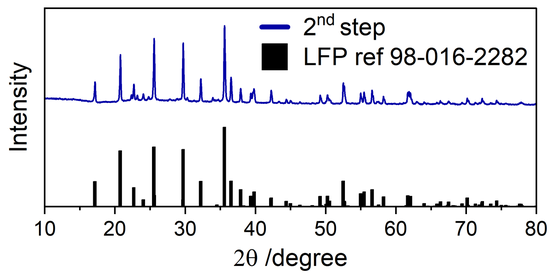
$LiFePO_4$: need to control the morphology and the carbon coating

1st: 400°C, 1 hour in a microwave

2nd: heat treatment with a carbon source in an inert environment







Conclusions

- ✓ **DES as efficient leaching medium**
- ✓ **Avoid the separation of CRMs**
- ✓ **Direct resynthesis**
- **Still need to implement the LFP resynthesis**

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