THE BATTERY CHRONICLES

STREAMS
SUSTAINABLE TECHNOLOGIES FOR REDUCING EUROPE'S BATTERY RAW MATERIALS DEPENDANCE

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Advancing the STREAMs of LFP Cathode Recycling through Regeneration for the benefit of European Automotive and BESS Battery Programs

My name is Roman Derkach, and I am a PhD student and Junior Researcher, working at the Institute for Sorption and Problems of Endoecology of the National Academy of Sciences of Ukraine (ISPE).

I joined the international project "Sustainable Technologies for Reducing Europe's bAttery raw MaterialS dependence" (STREAMS). Within the framework of this project, I am fortunate to be working on a comprehensive study aimed at developing methods for the direct regeneration of spent lithium iron phosphate ($LiFePO_4$, LFP) cathode materials. The primary objective of this research is to fully restore the stoichiometric composition, crystal structure, and electrochemical performance of the regenerated material to the level of commercial-grade LFP.

A comparative analysis of existing approaches has shown that, for LFP-based systems, direct regeneration is considered one of the most promising economically viable and environmentally sustainable methods compared with conventional pyrometallurgical and hydrometallurgical recycling technologies. Unlike layered oxide cathodes (NMC and NCA), LFP generally retains its stable olivine-type crystal framework during battery operation despite the formation of lithium vacancies, Li–Fe antisite defects, partial oxidation of Fe^{2+} to Fe^{3+}, and local crystal lattice distortions.

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This makes it possible to perform relithiation without complete chemical decomposition of the material into its precursor components followed by energy-intensive regeneration of LFP, for its reuse in the newly manufactured batteries used in EVs and BESS systems alike. Owing to the preservation of the fundamental crystal structure, the regeneration process can be primarily focused on compensating for lithium losses, restoring the oxidation state of iron, and eliminating structural defects that develop during long-term battery operation.

A critical stage of the technological process remains the separation of the active material from the aluminum current collector. Although mechanical separation is relatively effective on a laboratory scale, its industrial implementation is associated with considerable labor costs, increased energy consumption, and the risk of contamination by aluminum particles, which adversely affect the quality of the regenerated cathode material. As an alternative, a method based on the selective dissolution of the polymeric binder by soaking electrode sheets in solutions of lithium salts of organic acids (particularly lithium citrate, lithium ascorbate, and lithium oxalate) has been proposed. The use of these solutions enables gentle degradation of the polymer binder without significantly affecting either the crystal structure of LFP or the surface of the aluminum foil. This approach simultaneously addresses two important technological objectives: it ensures the careful separation of the active material while preserving the integrity of the aluminum foil for subsequent recycling, and it pre-enriches the processing solution with lithium ions, thereby creating a reaction medium suitable for subsequent hydrothermal relithiation. Of course as part of the STREAMS project we are also working with LFP recovered from aluminum foil through a Direct Recycling process implemented on the industrial scale at a partner facility of American Energy technologies Company, located in Chicago, Illinois, USA. Their LFP comes in as free-flowing powder without any residual aluminum and it can be easily processed through ISPE's regeneration systems. I am proud to say that we have joint publications between ISPE and AETC on this subject.

For the hydrothermal relithiation process, lithium salts of organic acids (namely lithium citrate, lithium ascorbate, and lithium oxalate) were employed during ISPE's experimental studies. These compounds exhibit different reducing abilities, complex-forming properties, and stability in aqueous solutions. It is assumed that their use provides the required concentration of lithium ions in the reaction medium while creating conditions for the reduction of structural Fe^{3+} to Fe^{2+}, which is essential for restoring the electrochemical activity of the material. The subsequent stage of the research is focused on investigating combinations of lithium salts of organic acids to achieve precise control over the reduction kinetics, lithium diffusion rate, and surface recrystallization processes. Since each lithium salt possesses distinct reducing and complexing properties, their balanced combination may promote more efficient relithiation of the structure, reduce the concentration of Li–Fe antisite defects, and improve the crystallinity of the regenerated material. In turn, this is expected to be one of the key factors governing the recovery of the electrochemical performance of the regenerated cathode material, including its charge/discharge rate capability, specific capacity, cycling stability, and long-term durability of lithium iron phosphate batteries.

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The developed regeneration methods can be utilized by STREAMS project partners, including TUBITAK, IMN, ULIEGE, UOULU, AETC, and others, for the direct regeneration of spent lithium iron phosphate cathode materials.

I am very happy that our Ukrainian team is able to contribute with this technology to the success of STREAMS project and to the overall success of European Union's lithium-ion battery program for use in EVs and BESS.

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