IOCM
2025
Conference

The 4th International Online Conference on Materials



03-05 November 2025 | Online

Tailoring Novel Cathode Materials with High Potential to Combine Performance with Reduced Content of Critical Raw Materials

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INTRODUCTION & AIM

Conventional cathode active materials (CAM) contain in their composition critical raw materials (CRM), which due to the high economic impact and criticality, make their long-term use impossible. The aim of this work is to obtain a new cathode material, which does not have CRMs in its composition, by two different methods: combustion synthesis (SCS) and co-precipitation. A second objective of this study was to analyse the two methods from an energy and raw material consumptions perspective. This research focused on the requirements needed to obtain a well-defined amount of the new complex oxide material.

METHOD

The oxide material was obtained through combustion synthesis (SCS), due to the exothermic reaction between fuel (glycine) and metal nitrides. For LiAl $_{0.25}$ Fe $_{0.25}$ Mn $_{0.25}$ Ni $_{0.25}$ O $_2$ composition, the used precursors were LiNO $_3$, Al(NO $_3$) $_3$ ·9H $_2$ O, Fe(NO $_3$) $_3$ ·9H $_2$ O, Mn(NO $_3$) $_2$ ·4H $_2$ O and Ni(NO $_3$) $_2$ ·6H $_2$ O. Stoichiometric amounts of nitrides and glycine were dissolved in 100 mL of distilled water and heated to 60°C until a viscous gel formed. The temperature was then increased, leading to autocombustion and powder formation. The resulting powder was calcinated at 600°C for 3h.

The co-precipitation process involves the formation of a metallic precursor complex $[M(NH_3)]^{2+}$ from a mixture of $Al_2(SO_4)_3*18H_2O$, $Fe(SO_4)_7H_2O$, $MnSO_4_H_2O$, and $NiSO_4_6H_2O$ in the presence of sodium hydroxide and ammonium. The precursor is then reacted with lithium hydroxide. The resulted mixture was calcinated at 800°C for 6 h to obtain the desired oxide material.

An inductively coupled plasma spectrometry (ICP-OES) of type Agilent 725 spectrometer was used to determinate the chemical composition of the samples.

The samples were analysed using FEI Quanta 3D FEG microscope.

RESULTS

Table 1. Results of chemical analysis, expressed in weight percentages.

| | Li | Al | Fe | Mn | Ni | 0 |
|---------------------------------|------|------|-------|-------|-------|-------|
| Nominal | 7.88 | 7.66 | 15.86 | 15.60 | 16.66 | 36.34 |
| Experimental - SCS | 8.03 | 7.36 | 15.1 | 14.6 | 14.6 | 40.21 |
| Experimental – Co-precipitation | 3.08 | 7.30 | 9.6 | 13.5 | 13.7 | 52.82 |

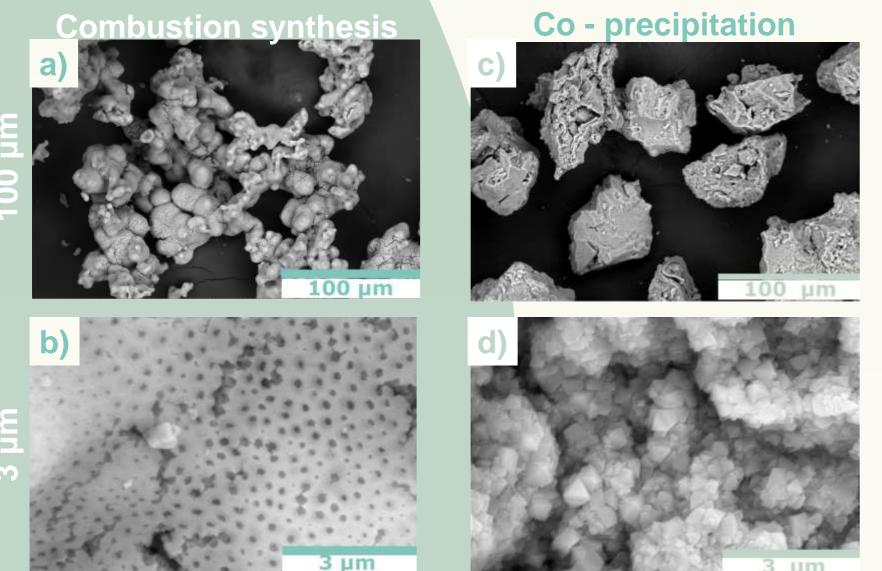


Fig 1. SEM images of the samples obtained by combustion synthesis at a) 100 μ m and b) 3 μ m; and co-precipitation at c) 100 μ m and d) 3 μ m

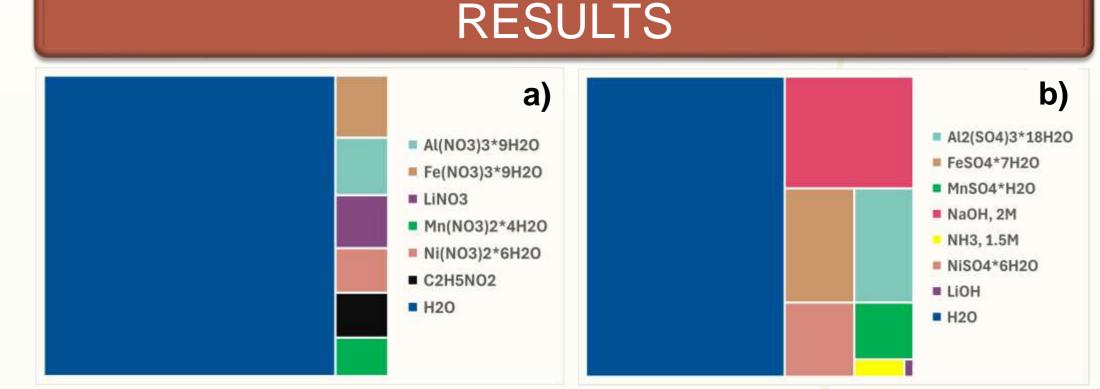
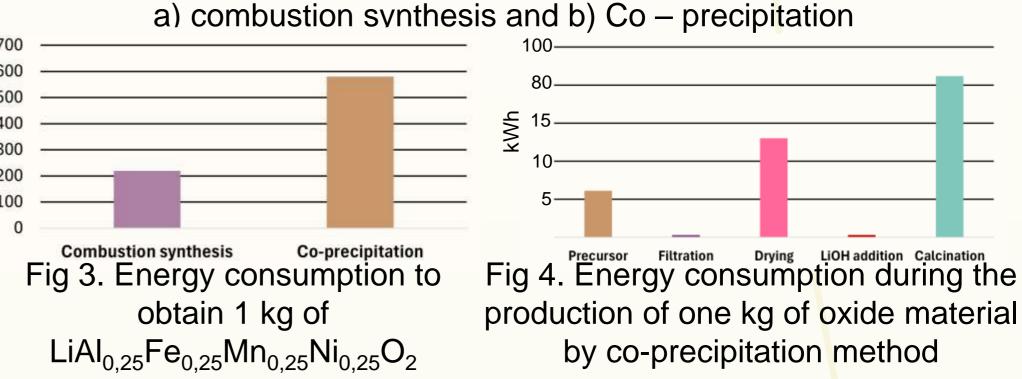


Fig 2. Graphical representation of the raw materials quantities that are necessary to obtain one kilogram of LiAl_{0,25}Fe_{0,25}Mn_{0,25}Ni_{0,25}O₂, through



DISCUSSION & CONCLUSION

From Table 1, it can be observed that using SCS method, a sample with a chemical composition close to the nominal one was obtained. The energy required to produce 1 kg of the oxide material is almost 60% lower in the case of SCS compared to co-precipitation. The latter method has a higher energy consumption, especially during the calcination stage. Even though the co-precipitation process has a lower CRM consumption compared to the SCS method, the differences between the experimental and nominal compositions indicate that an additional amount of LiOH could have been used. Both processes require optimization to reduce energy and material consumption, as well as to achieve the targeted composition. As a result of the co-precipitation process, large amounts of wastewater are generated, which must undergo a treatment process. In a future study, the environmental impact of the both methods will be analysed, to have a wider perspective on the life cycle assessment and the possibilities to extend it.

Acknowledgement

This work was supported by a grant of the Ministry of Research, Innovation and Digitization, CNCS/CCCDI – UEFISCDI, project number ERANET – ERAMIN - 3 – HETMOLION, within PNCDI IV; the Core Program within the National Research Development and Innovation Plan 2022-2027, financed by MCID, project no. PN 23 25 01 03 and by the European Union, from the Horizon Europe program, grant agreement number 101069789: project RELIEF – Recycling of Lithium from secondary raw materials and further. Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union or the European Climate, Infrastructure and Environment Executive Agency (CINEA). Neither the European Union nor the granting authority can be held responsible for them.

