

Understanding of chemical and catalytic mechanism of oxidative precipitation process applied for the lithium-ion batteries metals of interest recovery

Roberto Barbano^{1,2}, Tom Santori¹, Emmanuel Billy¹, Gaëlla Frajer¹, Hervé Muhr²

¹ Univ. Grenoble Alpes, CEA, Liten, DTNM, 38000 Grenoble, France

² Laboratoire Réactions et Génie des Procédés (LRGP), CNRS Université de Lorraine, 54001 Nancy, France

Li-ion battery is the key technology to drive the energetic transition. The global world demand is expected around 4,700 GWh by 2030 according to McKinsey & company [1] but a recent report from the European Commission identified almost all the materials in the battery as critical raw materials [2]. At the same time, the Council of the European Union established new rules to enhance a circular economy, through new collection targets and recycling obligations [3]. The context of European legislation, market and sovereignty highlights the need for efficient and economically sustainable close loop recycling process for the EU's strategic autonomy and energetic transition. The output of a close loop recycling process is the production of battery grade precursors to synthetize new cathode materials. The use of hydrometallurgical processes is necessary for the selective separation of the elements of interest (Mn, Co, Ni, Li) and will determine the costs and impacts of the processes [4]. This study focuses on the separation of Mn and Co by oxidative precipitation, using the oxidant peroxymonosulphate (PMS; $\text{HSO}_5^-/\text{SO}_4^{2-}$ couple) [5]. This approach is attractive because it is simple to implement, requires no solvent and does not contain any chlorinated species. Precipitation tests on Mn-Co model solutions show catalytic activation of PMS in the presence of Co^{2+} ions, facilitating selective Mn oxidation for a specific pH range (Figure 1). The role of cobalt and PMS was evaluated as a function of pH and temperature. It revealed some modifications in performance and PMS degradation phenomena. The process involves a complex precipitation mechanism, its understanding and control make it possible to consider high efficiency and purity of the precipitated elements.

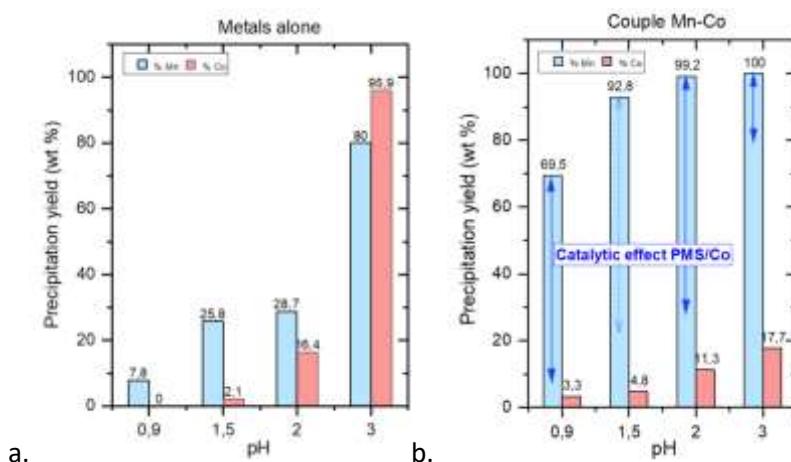


Figure 1 : Precipitation yield for a solution of H_2SO_4 / PMS for different pH with Co or Mn alone at 0.15 M, 30 °C, equivalence Co or Mn / oxidant = 1 (a) and Co and Mn mixed at 0.15 M each, 30 °C, equivalence Mn / oxidant = 1 (b)

[1]« Lithium-ion battery demand forecast for 2030 | McKinsey ». Consulté le: 8 juillet 2024. [En ligne]. Disponible sur: <https://www.mckinsey.com/industries/automotive-and-assembly/our-insights/battery-2030-resilient-sustainable-and-circular>

[2]Directorate-General for Internal Market, Industry, Entrepreneurship and SMEs (European Commission), M. Grohol, et C. Veeh, *Study on the critical raw materials for the EU 2023: final report*. Publications Office of

the European Union, 2023. Consulté le: 8 juillet 2024. [En ligne]. Disponible sur:
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[3]M. Ahuis, S. Doose, D. Vogt, P. Michalowski, S. Zellmer, et A. Kwade, « Recycling of solid-state batteries », *Nat. Energy*, vol. 9, n° 4, p. 373-385, avr. 2024, doi: 10.1038/s41560-024-01463-4.

[4]Y. Abe, R. Watanabe, T. Yodose, et S. Kumagai, « Cathode active materials using rare metals recovered from waste lithium-ion batteries: A review », *Heliyon*, vol. 10, n° 7, p. e28145, 2024, doi: 10.1016/j.heliyon.2024.e28145.

[5]E. Billy et S. Barthelemy, « Procede De Recyclage Des Batteries Li-Ion », 15 avril 2021 Consulté le: 8 juillet 2024. [En ligne]. Disponible sur: https://patentscope.wipo.int/search/fr/detail.jsf?docId=WO2021069822&_cid=P20-LYCQX0-82267-1