Journal of Power Sources 594 (2024) 234025

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Innovative methodology for advanced battery recycling research demonstrated with electrochemical extraction

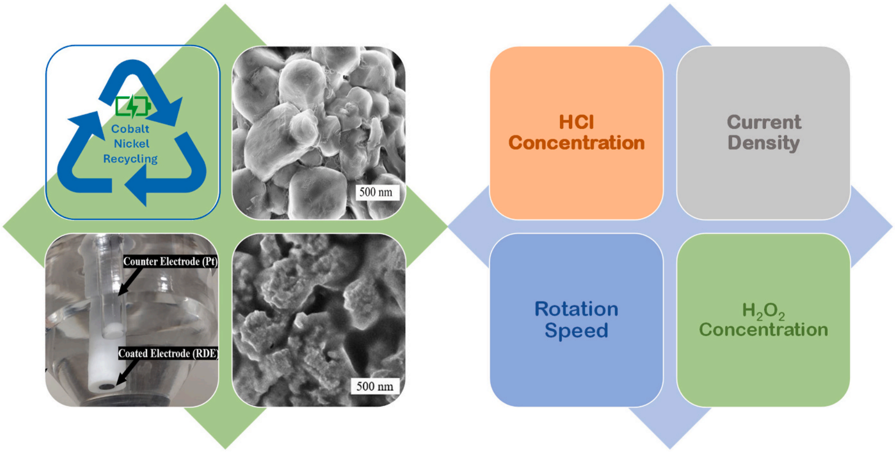
Bryce Watson [a], Brett Carlson [a], Alevtina Smirnova [b,*]

[a] Materials Science and Engineering Program, South Dakota Mines, Rapid City, SD, 57701, USA
[b] Department of Chemistry, Biology, and Health Sciences, South Dakota Mines, Rapid City, SD, 57701, USA

HIGHLIGHTS

- A new methodology for electrochemical metal extraction is proposed.
- Rotating disc electrode was utilized to test methodology.
- Concentration of HCl and H_2O_2, rotation rate, and current density were studied.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Battery recycling
Hydrometallurgy
Mass balance

ABSTRACT

Environmentally friendly processes to recapture critical metals and supplement markets are vital to overall sustainability in the energy sector. This work outlines a precise methodology for the electrochemical study of extraction performance in hydrometallurgical recycling. To demonstrate this method, the battery cathode material NMC532 is exposed to hydrochloric acid solutions at varying concentrations, rotation rates, current densities, and hydrogen peroxide contents. A dispersion of NMC532 and Nafion™ in water is deposited onto a rotating disc electrode surface to form a thin-film composite. The solution is sampled over time and relevant component concentrations are measured using inductively coupled plasma mass spectrometry (ICP-MS). The solution volume is maintained by replacing the sampled volume with initial solution and a correction equation is used to account for dilution. After electrochemical extraction, the NMC532 residue is collected for further analysis using scanning electron microscopy (SEM). This methodology requires minimal recyclable material to assess a wide variety of extraction conditions and provide various high-precision results. It can also facilitate the development of advanced electrochemical systems and provide valuable insight into key mechanisms for various hydrometallurgical and electrochemical processes.

---

* Corresponding author.
E-mail address: alevtina.smirnova@sdsmt.edu (A. Smirnova).

https://doi.org/10.1016/j.jpowsour.2023.234025
Received 25 August 2023; Received in revised form 15 December 2023; Accepted 26 December 2023

1. Introduction

As lithium-ion batteries become dominant, the need to recover valuable transition metals through recycling is critical. Many strategies have been employed to extract valuable metals from battery components including pyrometallurgical, hydrometallurgical, and more recently direct recycling. Most of these methods require pre-processing that involves battery discharging and shredding while hydrometallurgy involves processes to recover specific components such as magnetic separation for steel casings and flotation of plastics and graphite [1–8]. Direct recycling aims to utilize advanced component separation techniques to directly reuse the components in new battery manufacturing without breaking down the structure as is typically performed in metallurgical extraction processes [9–16]. Pyrometallurgy utilizes high temperatures to either oxidize or melt metals of interest, producing an alloy with transition metals (TM) and a slag containing various oxides. This process has benefits for battery recycling such as simplicity, speed, and convenience as the anode material graphite acts as a reducing agent for the oxides in the cathode. However, such methods require high energy consumption, cause atmospheric pollution from CO_2 and burned plastic byproducts, and possess intrinsic process hazards from molten metal transport and toxic gas generation [1,2,4,17–19]. Hydrometallurgy utilizes aqueous solutions with lixiviants, often strong oxidizing acids, to dissolve metal components for selective recovery by precipitation or selective solvent extraction. This process has benefits for battery recycling such as relatively low energy requirements, the ability to recover individual elements, and improved process safety with fewer hazards in processing. However, this process also includes drawbacks such as slower processing, larger industrial footprint, high water usage, and potential pollution in the form of solvents and lixiviants released through spillage and evaporation [1,2,4,20–25].

Recent developments in the recycling of lithium batteries have investigated electrochemical methods that may address some of the issues known for hydrometallurgical processing and improve various recycling and extraction processes altogether [26–36]. Electro-extraction is a rather modern adaptation to hydrometallurgical processing, particularly in environmental remediation and TM recovery from sea nodules but has not been widely implemented due to its complexity [37,38]. Recent studies provide evidence of electrochemical methods' benefits for recovering metals from secondary raw materials via electro-extraction and electrodeposition [39]. An electric field is commonly used to enhance the extraction of organic and inorganic compounds [40], phenolics [41], metals, such as nickel [42] and uranium from seawater [43], or in chromatography sample preparation to improve mass transport and selectivity [44]. In each of these applications, the electric field initiates different mechanisms and results in different reaction products. For example, the application of an electric field results in production of hydrogen peroxide which acts as a reducing agent [45–49].

Electrolytic reduction of mixed metal oxides facilitates metal extraction through re-oxidation in presence of solubilizing anions [50,51]. Finally, the charged surface of an electrode may concentrate charged ions in the hydrodynamic layer and facilitate adsorption and subsequent reaction on materials surfaces [52]. This would increase the effective reagent concentration without requiring an increase in bulk concentration. Some particular advantages of electrochemically enhanced extraction processes include use of eco-friendly reducing agents [53], precise process control, increased extraction rate, and improved environmental sustainability [54–58].

Proposed in this study, an advanced methodology using specified evaluation parameters would facilitate fast and high precision assessment of extraction performance from a minimal amount of waste. This methodology introduces an innovative recyclability evaluation that could be applied to other materials for the development of safe and environmentally friendly recycling processes. To demonstrate the proposed high-precision electrochemical methodology, a lithium-ion battery cathode, specifically $LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2$ (NMC532) [59] was chosen as a baseline. This material was investigated in a dilute inorganic acid extraction system in combination with hydrogen peroxide, a common additive for metal extraction from mixed metal oxides. While elevated temperature analysis is possible in the future, the current experimentation was performed at ambient temperature.

2. Experimental methods

2.1. Materials

Cathode NMC532 powder was purchased from MSE Supplies. Trace metal grade concentrated hydrochloric acid (HCl) and hydrogen peroxide (H2O2) were purchased from Fisher Scientific. Dupont DE521 1100 5 % NafionTM solution was purchased from Ion Power Inc. High purity liquid chromatography (HPLC) water was used for dilutions and rinsing.

2.2. Electrochemical measurements

The electrochemical studies of the metal extraction from the NMC532 powder were performed using the Rotating Disk Electrode (RDE) setup from Pine Instruments (Fig. 1A). Application of NMC532 to the RDE surface involved preparing a dispersion of the NMC532 powder in water mixed with Nafion solution and drying a droplet on the Glassy Carbon (GC) electrode surface. The dispersion was made by combining 10 mL of 1 mg/mL NMC532 in water and 50 μL of 5 % Nafion solution. The dispersion was mixed on a stir plate using a magnetic stir bar at 700 rpm and applied to the electrode by depositing 30 μL onto the GC surface. The electrode was then covered with a beaker and left in a fume hood to dry for at least 12 h forming a uniform film on the GC surface (Fig. 1C and D).

A GC rotating disc electrode in Teflon casing was used to reach a specified current density. The rotation rate was controlled using a Pine Instruments AFMSRCE rotator and preset to the specified speed for each experiment then adjusted manually to account for frictional losses after assembling the cell. The electrode potential was controlled using the Pine Instruments AFCBP1 Bipotentiostat with the associated Pine Research Aftermath software. A platinum counter electrode and an Ag/AgCl, Cl-reference electrode and a salt bridge filled with 0.1 M KCl were used in these experiments. For experiments with applied current, the current was set to 1 mA for a reduction current density of 5 mA/cm2 and 5 mA for a current density of 25 mA/cm2. Other settings were kept as default except for auto-ranging functions.

2.3. Preparation of samples for metal extraction and ICP-MS analysis

A 10 mL pipette was used to add relevant volumes of concentrated trace metal grade reagents to the flask which was then diluted to 100 mL using HPLC water. Each experiment required 300 mL of electrolyte to be prepared; 100 mL for the initial cell filling and at least 140 mL for replenishing solution.

Electrolyte samples were collected at the following extraction times for each experiment: 5, 10, 15, 20, 30, 40, 50, 60, 90, 120, 150, and 180 min. At the start of each experiment, an initial liquid electrolyte and replenishing solution were sampled. At the end of each experiment, another sample of the replenishing solution was collected to account for potential contamination during the experiment.

The TM concentrations (Mn, Co, Ni) were analyzed using the Aligent 7900 ICP-MS system with Masshunter 4.1 instrument control and analysis software. The system was calibrated using a blank of 2 % trace metal grade HCl diluted with HPLC water and PlasmaCAL SPC28AES standard solution at concentrations of 1, 10, 100 ppb and 1, 10 ppm. Quality control samples using HPS ICP-MSCS-PE3-A standard solution at concentrations of 100, 10, and 1 ppb were analyzed to confirm that instrument maintained acceptable accuracy.

The measured concentrations were corrected using equation (1) generalized for systems of different cell and sample volumes, and analyte elements.

$$C_{C,a,n} = C_{M,a,n} + \left[(V_S / V_c) \left(\left(\sum_{i=0}^{n-1} C_{M,a,i} \right) - (C_{R,a} * n) \right) \right] - C_{M,a,0} \tag{1}$$

where $C_{C,a,n}$ – corrected concentration of analyte for a numbered sample; $C_{M,a,n}$ – measured concentration of analyte in numbered sample; V_S – sample volume; V_C - cell volume; $\sum_{i=0}^{n-1} C_{M,a,i}$ - sum of measured concentrations from the first sample to the previous sample; $C_{R,a}$ - average concentration of the replenishing solution; n - number of samples collected; and $C_{M,a,0}$ initial concentration of the analyte. This equation allows for total ionic balance analysis considering that (a) solution is sufficiently mixed; (b) sampling/replenishing volumes remain constant throughout the experiment; (c) replenishing volume is equivalent to the sampling volume; (d) extracted ions remain in solution.

Inductively coupled plasma mass spectrometry (ICP-MS) measurements were processed using the dilution correction equation. The median of extraction completion rates calculated for all samples was compared between the experiments. For extraction of metals from NMC532, sample concentrations of nickel, manganese, and cobalt were measured and used for further calculations. Equation (2) was used to calculate the extraction completion rate for each sample. This equation was generalized for solutions of different electrolyte density, cell volume, starting material composition and amount, measured analyte elements, and elapsed extraction time for a given sample:

$$R_n = \big[(100 * \rho_e * V_C * MM_m) / (t_n * m_{0,m} * a) \big] * \left\{ \sum\nolimits_i^f \big[(C_{c,a_i,m}) / (MC_{a_i,m} * MM_{a_i}) \big] \right\} \tag{2}$$

where R_n – extraction completion rate for sample n; ρ_e – density of liquid electrolyte; V_C - cell volume; MM_m – molar mass of extracted material m; t_n – extraction time for sample n; $m_{0,m}$ – initial mass of material m; a – number of analytes used to estimate extraction completion rate; $\sum_i^f$– summation for measured analytes ai through af; $C_{c,a_i,m}$ - corrected concentration of analyte ai extracted from material m; $MC_{a_i,m}$ – molecular coefficient of analyte ai in material m; and MM_{a_i} – molar mass of analyte ai. This equation was used in assumption that the change in electrolyte density is negligible, and an algebraic average of analyte extraction completion rates is sufficient to describe the overall extraction rate.

The expected mass of the NMC532 in the NMC532/Nafion deposited layer was not always constant. For experiments where the extraction was incomplete, the expected mass of deposited NMC532 was assumed to be the average from all depositions. Finally, the extraction completion rate was calculated by averaging the slopes over time for each sample. The median value of sampled extraction completion rates yields a singular value which was used to compare the extraction efficiency between experiments. By averaging median extraction rates of experiments that share an identical setting, response curves were formed to compare the impact of each parameter at each relative level.

2.4. Morphological and chemical analysis before and after extraction

For morphological analysis, NMC532 cathode samples before and after electrochemical extraction were collected using double sided conductive carbon tape. The SEM images were collected from the samples where full extraction was not reached. Imaging was performed on a Helios 5 CX system and controlled by ThermoFisher XT Microscope Control software.

3. Results and discussion

3.1. Evaluation of the metal extraction efficiency by ICP-MS

The data regarding TM extraction from the NMC532 cathode material (Fig. 2) demonstrates the effect of the four chosen parameters (Table 1) on the electrochemical extraction efficiency. In some cases (e.g. Exp. 4, 8, 7, 6, and 3, Table 1) the majority of NMC532 particles were extracted in the first 60 min. However, in absence of hydrogen peroxide (Exp. 5 and 9, Table 1), the extraction completion rate dropped drastically after the first 60 min. In Exp. 2 performed in 0.1 M HCl solution the rate was slow but was steadily increasing until 120 min. This figure also highlights the exceptionally low extraction performance when all parameters are at their lowest levels (Exp. 1, Table 1).

The experimental results are presented using Taguchi design [60–63] with four parameters including HCl concentration, rotation rate, applied current density, and hydrogen peroxide concentration. The selected parameters (Table 1) include HCl concentrations (0.1, 1.0, and 2.0 M), rotation rates (400, 900, and 1600 rpm), reduction current densities (0, 5, and 25 mA/cm2), and hydrogen peroxide concentrations (0, 0.1, and 1.0 %).

The impact from each of the four extraction parameters on the overall metal extraction performance (Fig. 3) is presented using the median extraction completion rate results. The points on this figure are

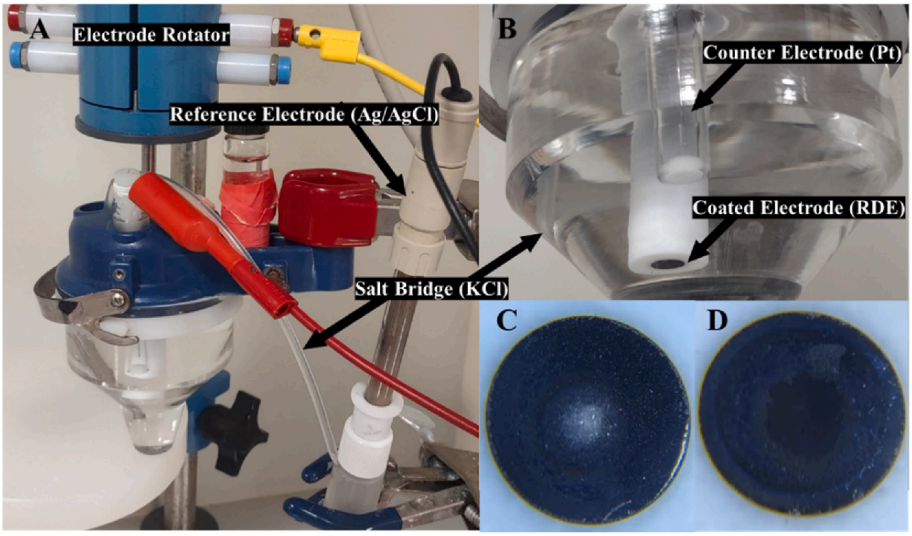


Fig. 1. (A, B) Electrochemical cell with RDE, platinum wire counter electrode, and KCl salt bridge submerged in the liquid electrolyte; (C, D) Images of the NMC532/Nafion coating on the GC surface before (C) and after metal extraction (D).

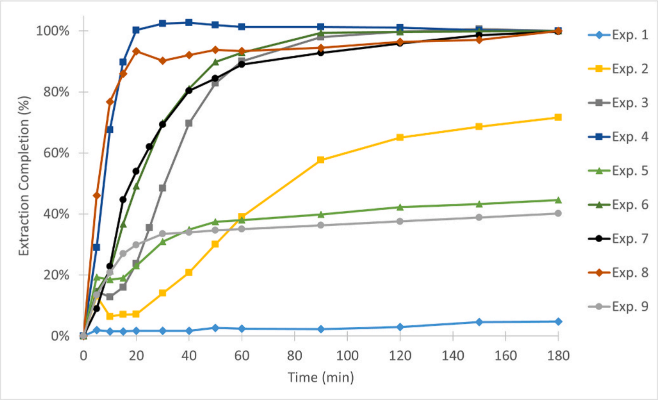


Fig. 2. ICP-MS results showing the metal extraction from NMC532 cathode powder at different extraction times for nine experiments presented in Table 1.

averages of median extraction rates for 3 experiments with an identical level of the specified parameter. For example, the level 1 point on the HCl curve is an average of the median extraction rates of experiments 1, 2, and 3 as they share the same HCl concentration of 0.1 M. This figure shows that the hydrogen peroxide concentration increases the median extraction rate. Conversely, rotation rate shows the least impact on median extraction rate. Furthermore, the HCl concentration demonstrated a significant impact between 0.1 M and 1.0 M but much lower impact in the range of 1.0 M–2.0 M. The current density also shows minimal impact at low level but managed to improve extraction at its higher values.

Summarizing the overall trends in NMC cathode material recycling (Fig. 3) it is evident that H_2O_2 concentration has the highest impact on metal extraction. Compared to H_2O_2, HCl concentration has also strong effect on extraction performance, however this effect decreases at concentrations above 1.0 M. Regarding the current density, this effect was minimal at lower currents, but more beneficial at higher currents. Similar to the current density parameter, the rotation rate is also minimally impactful and higher rotation rates appear to have a negative effect on the extraction performance. Further study should be undertaken to understand these trends and their synergistic effects in terms of mass transport limitations, for example correlation between rotation rate vs. extraction efficiency and how this extraction at microscale can be correlated with industry-relevant extraction processes.

The effect of measured potential required to hold the electrode at a specific current throughout experiments is presented in Fig. 4. In effect, a lower reduction potential can be correlated with higher conductivity of thin film NMC532 based composite. The potential decreased in the first 30–60 min before reaching a constant value. On the other hand, Exp. 2 and 7 show a wide range of variation which is directly correlated with an observed formation of gas bubbles at the electrode surface. Exp. 3 and 5 were similar to each other and required the highest potential to reach the high-level current density.

3.2. Morphological and structural properties before and after electrochemical extraction

SEM imaging was used to investigate the morphological changes in NMC532 cathode powder as a result of the electrochemical extraction process. To accomplish this, the NMC532 was imaged before and after an extraction experiment. The pristine NMC532 sample (Fig. 5) shows closely packed grains with smooth boundaries. On the contrary, the sample obtained in Exp. 5 (Table 1 and Fig. 6) demonstrates rough grain surfaces and wide gaps between grains. This observation provides evidence that the applied parameters, such as concentration of the acid, rotation rate, current density, and hydrogen peroxide concentration (Table 1), are important for understanding the specific effects caused by each of them in reaching high recycling efficiency.

In the proposed methodology, at least five potential sources of error should be highlighted. One of the major sources of error was a variation in mass of the NMC532 particles deposited on the GC surface. To address this, the 100 % extraction completion was based on the highest corrected concentration attained for experiments that appeared to reach completion (Exp. 3, 4, 6, 7, 8, Table 1). For experiments that did not reach completion, the 100 % extraction completion was based on the average of that of the experiments that reached completion. Another source of error involves generation of H_2 gas caused by electrolysis in acidic solution. This was observed in Exp. 7 (Table 1) where the slow rotation rate was coupled with high HCl and H_2O_2 concentration, and high current density. These gas bubbles caused blockage of the composite NMC532 layer and its contact with HCl-based electrolyte and deviations of the potential during chronopotentiometry tests (Fig. 4). Another potential source of error involves delamination of Nafion™ - NMC532 composite from the GC surface. This source of error could be caused by an interaction of H_2O_2 with Nafion, though this effect is negligible at low concentrations. Finally, there is some expected error involved with ICP-MS analysis at low concentrations of analytes. Even

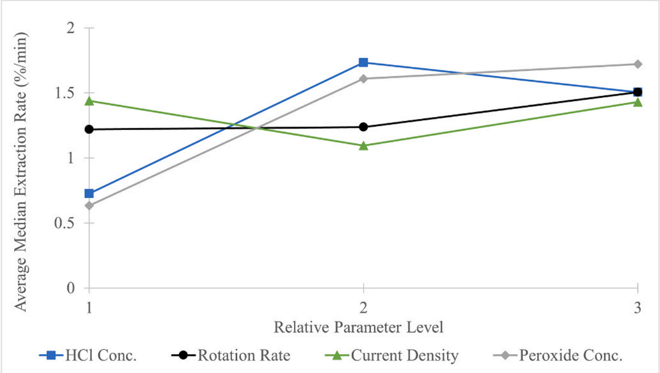


Fig. 3. The effect of the four experimental parameters on the efficiency of metal extraction from NMC532 lithium-ion battery cathode powder. Each point is the average of median extraction rates from 3 experiments of equivalent parameter setting.

Table 1
The performed experiments considering four testing parameters and the corresponding median extraction rates as performance indicators for each set of parameters. Four parameters were tested at three levels. For example, the three levels of HCl concentration are 0.1 M, 1.0 M, and 2.0 M.

Experiment #	HCl concentration (M)	Rotation rate (rpm)	Current density (mA/cm^2)	H_2O_2 concentration (vol. %)	Median extraction completion rate (%/min)
1	0.1	400	0	0	0.06
2	0.1	900	5	0.1	0.70
3	0.1	1600	25	1.0	1.42
4	1.0	400	5	1.0	1.81
5	1.0	900	25	0	1.07
6	1.0	1600	0	0.1	2.32
7	2.0	400	25	0.1	1.80
8	2.0	900	0	1.0	1.94
9	2.0	1600	5	0	0.78

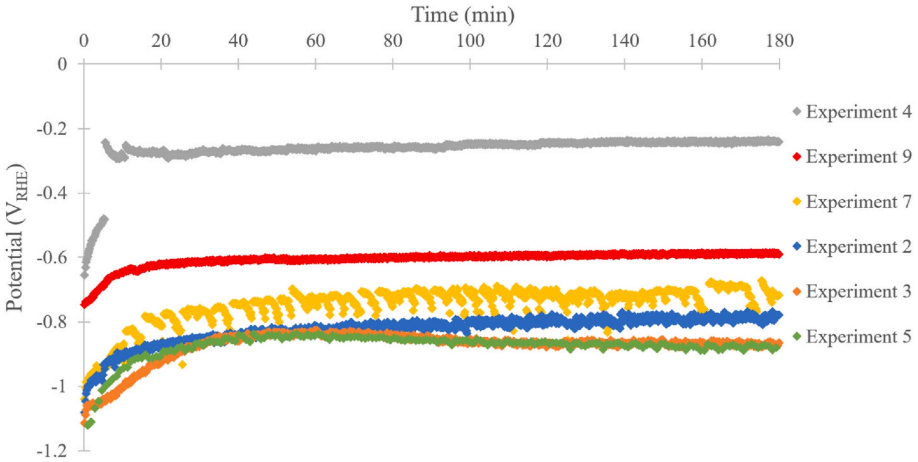


Fig. 4. The effect of the electrode potential vs. time for the NMC532 cathode powder (Exp. 2, 3, 5,and 7, Table 1).

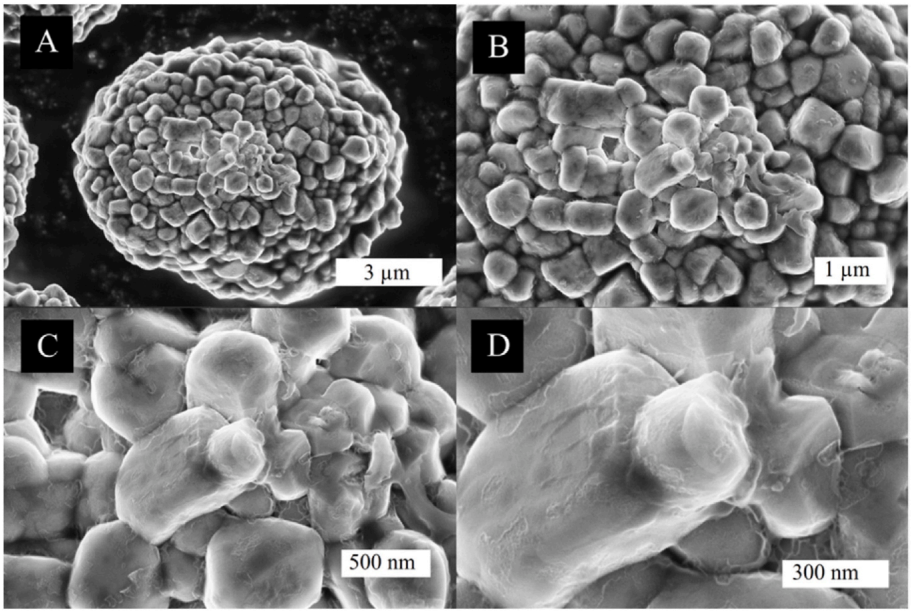


Fig. 5. SEM images of commercial NMC532 particles before electrochemical extraction at magnifications 10,000× (A), 20,000× (B), 50,000× (C), & 100,000× (D).

minor contamination during sample collection and analysis can significantly impact results.

4. Conclusions

As new energy storage technologies are developed, more advanced recycling processes should be developed to sustainably recover and reuse materials. This work outlines a precise methodology to study the effect of hydrochloric acid concentration, hydrogen peroxide concentration, current density, and rotation rate on the extraction of Ni, Mn, and Co from NMC532 cathodes used in lithium-ion and lithium metal batteries. The concentration of hydrogen peroxide was found to have the greatest impact while rotation rate had negligible impact on the extraction. Hydrochloric acid concentration significantly improved extraction rates when increased from 0.1 M to 1.0 M but this improvement was less significant when increased from 1.0 M to 2.0 M. The applied current density did not significantly increase the extraction rate but provided some improvement at 25 mA/cm^2, high enough to initiate water electrolysis. While these results do not support the hypothesis that an applied electrochemical current can improve extraction rates in this particular system, the method used may be applied to other experimental systems to generate precise data regarding the extraction of valuable wastes. This method can be considered effective for investigating electrochemical phenomena through high precision characterization of both the electrolyte and solid materials. While this method can benefit from improvements such as better control of particle deposition onto the electrode surface and prevention of gaseous bubble interference at the interface, it holds promise to closely monitor and investigate a variety of advanced recycling techniques. The proposed study demonstrates an accurate methodology for the electrochemical study of extraction properties in hydrometallurgical recovery using NMC352 as an example. Advanced recycling processes provide a path forward to eco-friendly extraction of lithium-ion battery cathodes.

CRediT authorship contribution statement

Bryce Watson: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Validation, Visualization, Writing – original draft, Writing – review & editing. **Brett Carlson:** Supervision, Writing – review & editing. **Alevtina Smirnova:** Conceptualization, Funding acquisition, Project administration, Supervision, Visualization, Writing – review & editing.

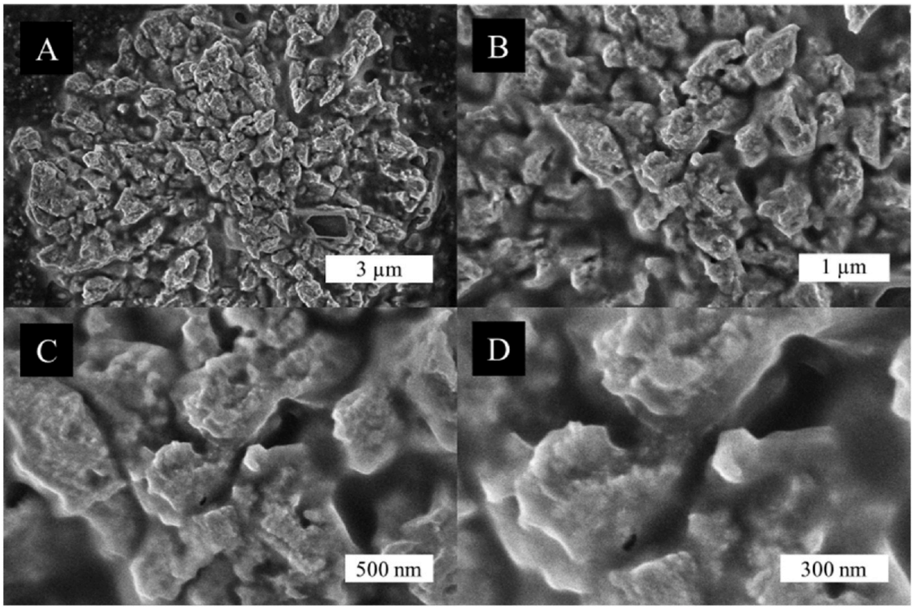


Fig. 6. The SEM images of the sample after electrochemical extraction (Exp. 5, Table 1) at magnifications 10,000× (A), 25,000× (B), 50,000× (C), & 100,000× (D).

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Alevtina White Smirnova reports financial support was provided by SDBoR Governor's Research Center. Alevtina White Smirnova reports financial support was provided by South Dakota School of Mines and Technology. Alevtina White Smirnova reports financial support was provided by National Science Foundation. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgments

The authors acknowledge financial support from the NSF IUCRC program for the "Center for solid-state electric power storage" (#2052631) and support from the South Dakota Board of Regents for the "Governor's Research Center (GRC) for electrochemical energy storage".

References

[1] L.F. Zhou, D. Yang, T. Du, H. Gong, W. Bin Luo, The current process for the recycling of spent lithium ion batteries, Front. Chem. 8 (2020), https://doi.org/10.3389/fchem.2020.578044.

[2] C. Liu, J. Lin, H. Cao, Y. Zhang, Z. Sun, Recycling of spent lithium-ion batteries in view of lithium recovery: a critical review, J. Clean. Prod. 228 (2019), https://doi.org/10.1016/j.jclepro.2019.04.304.

[3] M. Grützke, S. Krüger, V. Kraft, B. Vortmann, S. Rothermel, M. Winter, S. Nowak, Investigation of the storage behavior of shredded lithium-ion batteries from electric vehicles for recycling purposes, ChemSusChem 8 (2015), https://doi.org/10.1002/cssc.201500920.

[4] T. Or, S.W.D. Gourley, K. Kaliyappan, A. Yu, Z. Chen, Recycling of mixed cathode lithium-ion batteries for electric vehicles: current status and future outlook, Carbon Energy 2 (2020), https://doi.org/10.1002/cey2.29.

[5] H. Ali, H.A. Khan, M. Pecht, Preprocessing of spent lithium-ion batteries for recycling: need, methods, and trends, Renew. Sustain. Energy Rev. 168 (2022), https://doi.org/10.1016/j.rser.2022.112809.

[6] D. Thompson, C. Hyde, J.M. Hartley, A.P. Abbott, P.A. Anderson, G.D.J. Harper, To shred or not to shred: a comparative techno-economic assessment of lithium ion battery hydrometallurgical recycling retaining value and improving circularity in LIB supply chains, Resour. Conserv. Recycl. 175 (2021), https://doi.org/10.1016/j.resconrec.2021.105741.

[7] J. Marshall, D. Gastol, R. Sommerville, B. Middleton, V. Goodship, E. Kendrick, Disassembly of Li ion cells—characterization and safety considerations of a recycling scheme, Metals 10 (2020), https://doi.org/10.3390/met10060773.

[8] H. Pinegar, Y.R. Smith, End-of-Life lithium-ion battery component mechanical liberation and separation, JOM 71 (2019), https://doi.org/10.1007/s11837-019-03828-7.

[9] L. Gaines, Q. Dai, J.T. Vaughey, S. Gillard, Direct recycling R&D at the recell center, Recycling 6 (2021), https://doi.org/10.3390/recycling6020031.

[10] M. Ganesan, S.R. Ravishankar, Ola Electric Technologies Private Limited, Direct Regeneration of NMC Cathode Material by Using Non-toxic Solvent, Technical Disclosure Commons, Def. Pub. Series, 2023.

[11] B. Liu, J. Huang, J. Song, K. Liao, J. Si, B. Wen, M. Zhou, Y. Cheng, J. Gao, Y. Xia, Direct recycling of spent LiNi0.5Co0.2Mn0.3O2 cathodes based on single oxalic acid leaching and regeneration under mild conditions assisted by lithium acetate, Energy Fuels 36 (2022) 6552–6559.

[12] S.E. Sloop, J.E. Trevey, L. Gaines, Advances in Direct Recycling of Lithium-Ion Electrode Materials, ECS Meeting Abstracts, 2018, https://doi.org/10.1149/ma2018-01/4/607. MA2018-01.

[13] P. Xu, Q. Dai, H. Gao, H. Liu, M. Zhang, M. Li, Y. Chen, K. An, Y.S. Meng, P. Liu, Y. Li, J.S. Spangenberger, L. Gaines, J. Lu, Z. Chen, Efficient direct recycling of lithium-ion battery cathodes by targeted healing, Joule 4 (2020), https://doi.org/10.1016/j.joule.2020.10.008.

[14] Y. Bai, N. Muralidharan, J. Li, R. Essehli, I. Belharouak, Sustainable direct recycling of lithium-ion batteries via solvent recovery of electrode materials, ChemSusChem 13 (2020), https://doi.org/10.1002/cssc.202001479.

[15] P. Xu, Z. Yang, X. Yu, J. Holoubek, H. Gao, M. Li, G. Cai, I. Bloom, H. Liu, Y. Chen, K. An, K.Z. Pupek, P. Liu, Z. Chen, Design and optimization of the direct recycling of spent Li-ion battery cathode materials, ACS Sustain. Chem. Eng. 9 (2021), https://doi.org/10.1021/acssuschemeng.0c09017.

[16] X. Fan, C. Tan, Y. Li, Z. Chen, Y. Li, Y. Huang, Q. Pan, F. Zheng, H. Wang, Q. Li, A green, efficient, closed-loop direct regeneration technology for reconstructing of the LiNi0.5Co0.2Mn0.3O2 cathode material from spent lithium-ion batteries, J. Hazard Mater. 410 (2021), https://doi.org/10.1016/j.jhazmat.2020.124610.

[17] B. Makuza, Q. Tian, X. Guo, K. Chattopadhyay, D. Yu, Pyrometallurgical options for recycling spent lithium-ion batteries: a comprehensive review, J. Power Sources 491 (2021), https://doi.org/10.1016/j.jpowsour.2021.229622.

[18] S. Windisch-Kern, A. Holzer, L. Wiszniewski, H. Raupenstrauch, Investigation of potential recovery rates of nickel, manganese, cobalt, and particularly lithium from nmc-type cathode materials (LiNixMnyCozO2) by carbo-thermal reduction in an inductively heated carbon bed reactor, Metals 11 (2021), https://doi.org/10.3390/met11111844.

[19] R.E. Ciez, J.F. Whitacre, Examining different recycling processes for lithium-ion batteries, Nat. Sustain. 2 (2019), https://doi.org/10.1038/s41893-019-0222-5.

[20] E. Asadi Dalini, G. Karimi, S. Zandevakili, M. Goodarzi, A Review on Environmental, Economic and Hydrometallurgical Processes of Recycling Spent Lithium-Ion Batteries, Mineral Processing and Extractive Metallurgy Review, 2020, https://doi.org/10.1080/08827508.2020.1781628.

[21] W.S. Chen, H.J. Ho, Recovery of valuable metals from lithium-ion batteries NMC cathode waste materials by hydrometallurgical methods, Metals 8 (2018), https://doi.org/10.3390/met8050321.

[22] C.A. Nogueira, F. Margarido, Battery recycling by hydrometallurgy: evaluation of simultaneous treatment of several cell systems, in: Energy Technology 2012:

Carbon Dioxide Management and Other Technologies, 2012, https://doi.org/10.1002/9781118365038.ch28.
[23] Y. Zhou, Z. Chen, J. Xu, Progress and prospect of recycling spent lithium battery cathode materials by hydrometallurgy, Huagong Xuebao/CIESC J. 73 (2022), https://doi.org/10.11949/0438-1157.20210901.
[24] W. Xuan, A. de Souza Braga, C. Korbel, A. Chagnes, New insights in the leaching kinetics of cathodic materials in acidic chloride media for lithium-ion battery recycling, Hydrometallurgy 204 (2021), https://doi.org/10.1016/j.hydromet.2021.105705.
[25] N. Vieceli, P. Benjamasutin, R. Promphan, P. Hellström, M. Paulsson, M. Petranikova, Recycling of lithium-ion batteries: effect of hydrogen peroxide and a dosing method on the leaching of LCO, NMC oxides, and industrial black mass, ACS Sustain. Chem. Eng. 11 (2023), https://doi.org/10.1021/acssuschemeng.3c01238.
[26] B.I. Bairachnyi, Y.A. Zhelavskaya, V.N. Skorikova, Study of electrochemical extraction of cobalt from spent catalysts, Russ. J. Appl. Chem. 70 (1997).
[27] J. Coyle, X. Li, S. Santhanagopalan, A. Burrell, Recycle of End-Of-Life NMC 111 Cathodes by Electrochemical Relithiation, ECS Meeting Abstracts, 2019, https://doi.org/10.1149/ma2019-02/5/449. MA2019-02.
[28] M.B.J.G. Freitas, E.M. Garcia, Electrochemical recycling of cobalt from cathodes of spent lithium-ion batteries, J. Power Sources 171 (2007), https://doi.org/10.1016/j.jpowsour.2007.07.002.
[29] H. Wang, M. Li, S. Garg, Y. Wu, M. Nazmi Idros, R. Hocking, H. Duan, S. Gao, A. J. Yago, L. Zhuang, T.E. Rufford, Cobalt electrochemical recovery from lithium cobalt oxides in deep eutectic choline Chloride+Urea solvents, ChemSusChem 14 (2021), https://doi.org/10.1002/cssc.202100954.
[30] E.J. Calvo, Electrochemical methods for sustainable recovery of lithium from natural brines and battery recycling, Curr. Opin. Electrochem. 15 (2019), https://doi.org/10.1016/j.coelec.2019.04.010.
[31] E.A.A. Aboelazm, N. Mohamed, G.A.M. Ali, A.S.H. Makhlouf, K.F. Chong, Recycling of cobalt oxides electrodes from spent lithium-ion batteries by electrochemical method, in: Topics in Mining, Metallurgy and Materials Engineering, 2021, https://doi.org/10.1007/978-3-030-68031-2_4.
[32] H. Li, H. Li, C. Li, J. Liang, H. Yan, Z. Xu, Study on the behavior of electrochemical extraction of cobalt from spent lithium cobalt oxide cathode materials, Materials 14 (2021), https://doi.org/10.3390/ma14206110.
[33] H. Li, Y. Fu, J. Liang, C. Li, J. Wang, H. Yan, Z. Cai, Electrochemical mechanism of recovery of nickel metal from waste lithium ion batteries by molten salt electrolysis, Materials 14 (2021), https://doi.org/10.3390/ma14226875.
[34] K. Liu, S. Yang, F. Lai, H. Wang, Y. Huang, F. Zheng, S. Wang, X. Zhang, Q. Li, Innovative electrochemical strategy to recovery of cathode and efficient lithium leaching from spent lithium-ion batteries, ACS Appl. Energy Mater. 3 (2020), https://doi.org/10.1021/acsaem.0c00395.
[35] M.M. Thackeray, P.J. Johnson, L.A. de Picciotto, P.G. Bruce, J.B. Goodenough, Electrochemical extraction of lithium from LiMn2O4, Mater. Res. Bull. 19 (1984), https://doi.org/10.1016/0025-5408(84)90088-6.
[36] C. Lupi, M. Pasquali, A. Dell'Era, Nickel and cobalt recycling from lithium-ion batteries by electrochemical processes, Waste Manag. (2005), https://doi.org/10.1016/j.wasman.2004.12.012.
[37] L.G. Santos Sobral, R.L.C. Santos, Electroleaching process for remediation of mercury contaminated soils, Contam. Soil '95 (1995), https://doi.org/10.1007/978-94-011-0421-0_141.
[38] A. Kumari, K.A. Natarajan, Electroleaching of polymetallic ocean nodules to recover copper, nickel and cobalt, Miner. Eng. 14 (2001), https://doi.org/10.1016/S0892-6875(01)00090-5.
[39] N. Leclerc, S. Legeai, M. Balva, C. Hazotte, J. Comel, F. Lapicque, E. Billy, E. Meux, Recovery of metals from secondary raw materials by coupled electroleaching and electrodeposition in aqueous or ionic liquid media, Metals 8 (2018), https://doi.org/10.3390/met8070556.
[40] M. Gavahian, Y.H. Chu, S. Sastry, Extraction from food and natural products by moderate electric field: mechanisms, benefits, and potential industrial applications, Compr. Rev. Food Sci. Food Saf. 17 (2018), https://doi.org/10.1111/1541-4337.12362.
[41] C. Delsart, R. Ghidossi, C. Poupot, C. Cholet, N. Grimi, E. Vorobiev, V. Milisic, M. M. Peuchot, Enhanced extraction of phenolic compounds from merlot grapes by pulsed electric field treatment, Am. J. Enol. Vitic. 63 (2012), https://doi.org/10.5344/ajev.2012.11088.
[42] V.E. Serga, L.D. Kulikova, B.A. Purin, Extraction of nickel by liquid membranes in an electric field, Separ. Sci. Technol. 35 (2000), https://doi.org/10.1081/ss-100100158.
[43] N. Li, L. Yang, R. Su, N. Shi, J. Wu, J. Zhao, L. Wen, Z. Wang, Selective extraction of uranium from seawater on amidoximated MXene/metal-organic framework architecture under an electric field, Desalination 566 (2023), https://doi.org/10.1016/j.desal.2023.116940.
[44] H. Chen, R.K. Anand, Sample Preparation by Electric Field–Assisted Extraction, vol. 40, LC-GC North America, 2022, https://doi.org/10.56530/lcgc.na.rg2889z4.
[45] X. Zhang, X. Zhao, P. Zhu, Z. Adler, Z.-Y. Wu, Y. Liu, H. Wang, Electrochemical oxygen reduction to hydrogen peroxide at practical rates in strong acidic media, Nat. Commun. 13 (2022).
[46] O.M. Cornejo, I. Sirés, J.L. Nava, Cathodic generation of hydrogen peroxide sustained by electrolytic O2 in a rotating cylinder electrode (RCE) reactor, Electrochim. Acta 404 (2022), https://doi.org/10.1016/j.electacta.2021.139621.
[47] I. Diouf, O. Dia, M.B. Diedhiou, P. Drogui, A.O. Toure, S.M. Lo, M. Rumeau, C. G. Mar/Diop, Electro-generation of Hydrogen Peroxide Using a Graphite Cathode from Exhausted Batteries: Study of Influential Parameters on Electro-Fenton Process, Environmental Technology (United Kingdom), 2020, p. 41, https://doi.org/10.1080/09593330.2018.1537309.
[48] N. Wang, S. Ma, P. Zuo, J. Duan, B. Hou, Recent progress of electrochemical production of hydrogen peroxide by two-electron oxygen reduction reaction, Adv. Sci. 8 (2021), https://doi.org/10.1002/advs.202100076.
[49] K.H. Wu, D. Wang, X. Lu, X. Zhang, Z. Xie, Y. Liu, B.J. Su, J.M. Chen, D.S. Su, W. Qi, S. Guo, Highly selective hydrogen peroxide electrosynthesis on carbon: in situ interface engineering with surfactants, Chem 6 (2020), https://doi.org/10.1016/j.chempr.2020.04.002.
[50] S. Bonefas, C. Morneau, B. Kyer, E. Baker, Low Temperature Electrolysis of Mixed Metal Oxides, Worcester, 2017.
[51] X. Zou, S. Gu, X. Lu, X. Xie, C. Lu, Z. Zhou, W. Ding, Electroreduction of iron(III) oxide pellets to iron in alkaline media: a typical shrinking-core reaction process, Metall. Mater. Trans. B 46 (2015), https://doi.org/10.1007/s11663-015-0336-8.
[52] J.J. Howard, J.S. Perkyns, B.M. Pettitt, The behavior of ions near a charged wall-dependence on ion size, concentration, and surface charge, J. Phys. Chem. B 114 (2010), https://doi.org/10.1021/jp9108865.
[53] F. Wu, X. Liu, G. Qu, P. Ning, A critical review on extraction of valuable metals from solid waste, Sep. Purif. Technol. 301 (2022), https://doi.org/10.1016/j.seppur.2022.122043.
[54] V. Rai, D. Liu, D. Xia, Y. Jayaraman, J.C.P. Gabriel, Electrochemical approaches for the recovery of metals from electronic waste: a critical review, Recycling 6 (2021), https://doi.org/10.3390/recycling6030053.
[55] Q. Song, Y. Liu, L. Zhang, Z. Xu, Selective electrochemical extraction of copper from multi-metal e-waste leaching solution and its enhanced recovery mechanism, J. Hazard Mater. 407 (2021), https://doi.org/10.1016/j.jhazmat.2020.124799.
[56] Y. Jang, C.H. Hou, S. Park, K. Kwon, E. Chung, Direct electrochemical lithium recovery from acidic lithium-ion battery leachate using intercalation electrodes, Resour. Conserv. Recycl. 175 (2021), https://doi.org/10.1016/j.resconrec.2021.105837.
[57] A. Battistel, M.S. Palagonia, D. Brogioli, F. La Mantia, R. Trócoli, Electrochemical methods for lithium recovery: a comprehensive and critical review, Adv. Mater. 32 (2020), https://doi.org/10.1002/adma.201905440.
[58] L.A. Diaz, M.L. Strauss, B. Adhikari, J.R. Klaehn, J.S. McNally, T.E. Lister, Electrochemical-assisted leaching of active materials from lithium ion batteries, Resour. Conserv. Recycl. 161 (2020), https://doi.org/10.1016/j.resconrec.2020.104900.
[59] A. Fahimi, H. Solorio, R.K. Nekouei, E. Vahidi, Analyzing the environmental impact of recovering critical materials from spent lithium-ion batteries through statistical optimization, J. Power Sources 580 (2023), 233425, https://doi.org/10.1016/j.jpowsour.2023.233425.
[60] K. Krishniah, P. Shahabudeen, K. Krishnaiah, P. Shahabudeen, Applied Design of Experiments and Taguchi Methods, 2012.
[61] J.A. Ghani, H. Jamaluddin, M.N.A. Rahman, B.M. Deros, Philosophy of taguchi approach and method in design of experiment, Asian. J. Sci. Res. 6 (2013), https://doi.org/10.3923/ajsr.2013.27.37.
[62] P. Woolf, S. Fraley, J. Zalewski, M. Oom, B. Terrien, 14.1: design of experiments via taguchi methods - orthogonal arrays, in: Chemical Process Dynamics and Controls, 2022.
[63] V.K. Modi, D.A. Desai, Review of taguchi method, design of experiment (doe) & analysis of variance (anova) for quality improvements through optimization in foundry, J. Emerg. Technol. Innovat. Res.(JETIR) (2018) 5.