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Li-Ion Battery Recycling via High-Intensity Milling Followed by Organic Acid Leaching for Preferential Lithium Extraction

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Abstract

Nowadays, spent batteries are considered a secondary and potential resource to meet the growing demand for lithium, a critical element widely used in the manufacturing of electric vehicles. Therefore, this work presents a hydrometallurgical method for extracting lithium from Nickel–Manganese–Cobalt (NMC) batteries. Citric ($C_6H_8O_7$) and oxalic ($C_2H_2O_4$) acids were used as leaching agents, both of which are cataloged as environmentally friendly organic compounds. To comprehend the chemical interactions between citrate (cit), oxalate (ox) and metallic ions, a thermodynamic analysis is presented. According to this analysis, both ions were effective in dissolving lithium; however, the experimental studies demonstrated that oxalate ensured a selective process and achieved complete lithium dissolution under the experimental conditions 1 M $C_2H_2O_4$, 50 g/L, 60 °C, and 60 min, with a mechanically treated sample (milling time 8 min at 1000 rpm). In this process, the other metals present in the sample, such as cobalt, nickel, and manganese, formed insoluble species with oxalate, allowing their recovery in subsequent stages. Therefore, this investigation provides a proficient methodology for battery recycling, emphasizing sustainable practices.

Keywords: battery recycling; lithium extraction; eco-friendly leaching; circular materials

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1. Introduction

The automotive industry faces a significant challenge in reducing the use of virgin materials in the production of electric vehicles (EVs) [1]. One such material is lithium, a critical metal essential for EV manufacturing. The projected lithium demand is expected to grow by 18.9% (approximately USD 55.52 billion) by 2032 [2]. Therefore, the recycling sector is developing new strategies to recover it from various types of electronic waste, such as lithium-ion batteries (LIBs).

Current LIB recycling processes use hydrometallurgical techniques to recover and reincorporate not only lithium but also manganese, nickel, copper, and aluminum as recycled raw materials. This technology offers advantages such as minimal operation stages, acceptable product purity, high recovery efficiency, and moderate energy consumption [3]. Furthermore, numerous strategies have been employed to develop more environmentally friendly leaching processes. One notable approach is the extensive research on the use of organic acids as leaching agents [4,5].

Organic acids are chemical compounds formed by hydroxyl (HO^-) or carboxyl ($-COOH$) functional groups and carbon-carbon double bonds in their structures. Organic acids are generally classified as weak acids and can form complexes with metallic species. Properly, these complexes are a combination of a Lewis acid (the central metal atom) and the Lewis bases, ligands (organic acid dissociated) [6,7]. Therefore, acids such as lactic ($C_3H_6O_3$), malic ($C_4H_6O_5$), ascorbic ($C_6H_8O_6$), tartaric ($C_4H_6O_6$), aspartic ($C_4H_7NO_4$), formic (CH_2O_2), citric ($C_6H_8O_7$), oxalic ($C_2H_2O_4$), succinic ($C_4H_6O_4$), etc., have been studied for metal extraction from batteries with different chemistries: $LiCoO_2$ (LCO), $LiFePO_4$ (LFP), $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$, (NMC), $LiMn_2O_4$ (LMO), etc. [4,5,8].

High leaching efficiencies (~100%) of cobalt, lithium, nickel, and manganese have been reported, specifically in combined systems, where additional species, reductants (hydrogen peroxide, H_2O_2, D-glucose, $C_6H_{12}O_6$, stannous chloride, $SnCl_2$, etc.), are incorporated into organic leaching solutions [9–11]. Traditionally, during these processes, the temperature is typically raised, allowing leaching yields to exceed 90%. For example, 99% Co and 98.67% Li were obtained from LCO batteries using oxalic acid (0.46 M) and $SnCl_2$ (0.3 M) at 100 °C for 40 min [12]. While at 90 °C with citric acid (1.25 M), 0.2 M salicylic acid ($C_7H_6O_3$), and hydrogen peroxide (6 vol%) for 90 min, 99.5% Co and 97% Li were extracted [13]. In NMC treatment, leaching yields of cobalt, lithium, nickel, and manganese between 95% and ~99% have been obtained employing malic, acetic, and citric acid combined with H_2O_2 in a temperature range of 60–90 °C [8].

Multiple combinations of organic acids and reductants, and how temperature affects these systems, are widely reported [14], demonstrating that organic compounds are a potential alternative to substitute inorganic acids, which are considered the optimal leaching agents. Nevertheless, this novel proposal requires improvement. For example, reductants are essential for enhancing leaching yields; however, they can degrade organic acids at temperatures above 30 °C [15]. On the other hand, metallic impurities, such as copper, iron, aluminum, etc., can also be dissolved, and their presence in leaching liquors affects the selectivity of valuable metal extraction [16,17]. Therefore, it is necessary to continue analyzing these organic species for the treatment of spent batteries.

This investigation proposes a methodology for extracting lithium from NMC 111 batteries, using citric and oxalic acids as leaching agents, operating at low temperatures (<100 °C) and without reductants. Citric and oxalic acids have been selected due to their ability to act as multiligand chelators, allowing them to coordinate with different metallic species in the black mass [5]. The capacity to form stable complexes with metallic ions is because both organic species dissociate in aqueous media in successive steps (Equations (1)–(5) [18,19]).

Successive dissociation steps of citrate:

$$H_3cit \rightleftharpoons H^+ + H_2cit^- \tag{1}$$

$$H_2cit^- \rightleftharpoons H^+ + Hcit^{2-} \tag{2}$$

$$Hcit^{2-} \rightleftharpoons H^+ + cit^{3-} \tag{3}$$

Successive dissociation steps of oxalate:

$$H_2C_2O_4 \rightleftharpoons H^+ + HC_2O_4^- \tag{4}$$

$$HC_2O_4^- \rightleftharpoons H^+ + C_2O_4^{2-} \tag{5}$$

On the other hand, mechanical activation (in one-step milling) is proposed as a pretreatment to enhance lithium extraction by liberating it from the solid material. Therefore, this work investigated the influence of milling time, speed, leaching agent concentration, and temperature on the process. Moreover, a thermodynamic analysis is presented to comprehend the chemical interaction between organic and metallic ions. This theoretical study offers valuable insights, such as the pH range for selective lithium extraction.

2. Materials and Methods

2.1. Black Mass Characterization

A discharged batch of 150 kg EV lithium-ion battery packs, NMC-111, provided by Volvo Cars AB, Gothenburg, Sweden was tested. The packs were first dismantled (Stena Recycling AB, Gothenburg, Sweden); then, they were crushed, mechanically sieved, and magnetically separated by Akkuser Oy (Nivala, Finland).

The chemical analysis of the black mass with a particle size of $\leq$500 µm was performed. 0.2 g of solid was dissolved in 30 mL of aqua regia prepared with hydrochloric acid (HCl, 37%, Sigma-Aldrich, St. Louis, MO, USA) and nitric acid (HNO_3, 65–67%, Sigma-Aldrich) (HCl:HNO_3, 3:1 v/v). The experiments were conducted in triplicate at 80 °C for 6 h, using magnetic agitation (200 rpm). Afterward, the solutions were cooled to room temperature, filtered, and diluted with 0.1 M nitric acid (HNO_3, 65–67%, Sigma-Aldrich). All samples were analyzed by Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES, Thermo Fisher Scientific, Greenville, NC, USA, Model iCAP^{TM} 6000 Series).

2.2. Physical and Chemical Treatments: Mechanical Activation and Acid Leaching

The milling stage was carried out using a Vibratory Disc Mill RS 200 (Retsh®, Haan, Germany). The experimental parameters were 700 and 1000 rpm and 2 and 8 min. After the physical treatment, each treated sample was analyzed chemically in triplicate using aqua regia, as described in the previous section.

The leaching tests were performed in triplicate using a solid–liquid ratio of 50 g/L; the solutions were prepared with deionized water (Millipore Milli-Q, Q-POD-Merck KGaA, Darmstadt, Germany) and citric acid ($C_6H_8O_7$, $\geq$99.5%, Sigma-Aldrich) or oxalic acid ($C_2H_2O_4$, $\geq$99%, Sigma-Aldrich), 1 and 2 M. The leaching systems were double-wall PVDF reactors (MEAB®, Gothenburg, Sweden), of 100 mL capacity, and mechanical agitation was employed (300 rpm). The temperature was controlled using an Optima™ heated circulating bath model T100-P12 (Grant Instruments, Cambridge, UK), and the pH of the solutions was measured using a MeterLab-PHM240 electrode (Radiometer Analytical SAS, Villeurbanne, France). Finally, all samples were analyzed by ICP-OES (Thermo Fisher Scientific, Model iCAP^{TM} 6000 Series).

2.3. Solid Residue Characterization: X-Ray Diffraction (XRD) and Scanning Electron Microscope Combined with Energy Dispersive X-Ray Spectroscopy (SEM-EDS)

The solid residue after the leaching process was analyzed by XRD and SEM-EDS techniques. XRPD Siemens D5000 diffractometer (LabX, Austin, TX, USA) was used, which operated under the following conditions: a radiation source in a two-theta interval of 10–70° (Cu λ = 1.54 Å) with a rotational speed of 15 rpm, and a voltage of 40 mA and 40 kV. The analytical interpretation of the diffractograms was performed using EVA (V6.0.0.7.) software and the JCPDS database.

SEM-EDS tests were conducted using Scanning Electron Microscopy coupled with Energy Dispersive X-ray Spectroscopy (SEM-EDS, FEI Quanta 200 FEG SEM with an Oxford Instruments X-Max EDS detector, HiTechTrader, Coatesville, PA, USA).

3. Results

3.1. Thermodynamic Analysis of the Metal-Citrate (M-Cit) System

Make Equilibrium Diagrams Using Sophisticated Algorithms (MEDUSA V.1) software [20], was used to construct species distribution diagrams for the proposed chemical system, metal-citrate (M-cit). In this analysis, the metal composition of the untreated sample (Table 1) was considered, where lithium, cobalt, nickel, manganese, copper, and aluminum were detected. It is important to highlight that the physically treated samples were also analyzed, and their metallic content is reported in Table A1 (Appendix A).

Table 1. Average metallic content of the untreated black mass sample (≤500 µm).

Element	Li	Co	Ni	Mn	Cu	Al
wt%	3.70 ± 0.10	12.00 ± 0.30	9.30 ± 0.20	8.10 ± 0.10	3.00 ± 0.10	0.80 ± 0.10

Figure 1 shows the species distribution diagrams for the different elements. In the first case (Figure 1a), lithium in ionic form is prevalent from pH value 1 to 4, and it can form a soluble compound with citrate ion ($\mathrm{Li(cit)^{2-}}$) from pH ~4 to 14. On the other hand, two cobalt-citrate species are formed ($\mathrm{Co(cit)^{-}}$, $\mathrm{Co(OH)_{2(s)}}$); where $\mathrm{Co(cit)^{-}}$ is dominant from pH 2 to ~10, and the solid becomes prevalent at pH values ≥ 10 (Figure 1b). A similar case is observed for nickel-citrate (Figure 1c), where a hydroxide species is predominant in alkaline conditions. When the pH is below 10, only soluble nickel-citrate compounds are formed. Finally, the citrate ion forms soluble species with manganese practically in the entire pH range (Figure 1d). Therefore, this analysis demonstrates that lithium, cobalt, and manganese are soluble in acidic and alkaline conditions (1 > pH~10). Nevertheless, impurity dissolution (copper and aluminum) is expected at this pH range (Figure 1e,f).

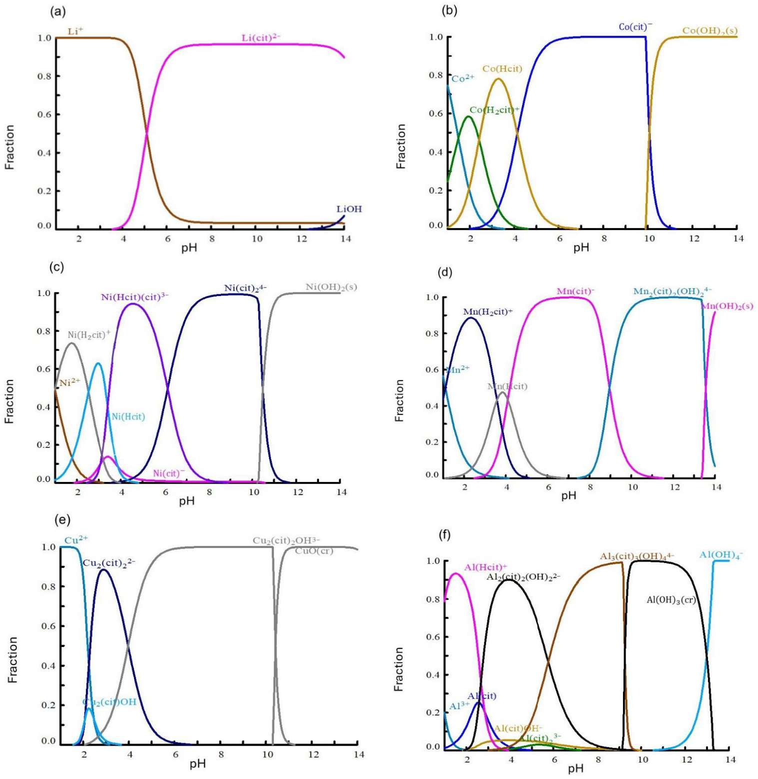

Figure 1. Species distribution diagrams for the metal-citrate (M-cit) system: (**a**) Li-cit, (**b**) Co-cit, (**c**) Ni-cit, (**d**) Mn-cit, (**e**) Cu-cit, and (**f**) Al-cit. All cases with 1 M cit at room temperature (25 °C). Elaborated using MEDUSA software [20].

According to the previous thermodynamic study, experimental tests were designed to leach the untreated and treated mechanically samples (Table 1 and Table A1, Appendix A) in citric acid solutions (1 M and 2 M) at room temperature (25 °C), and without adjusting the pH value to analyze the leaching behavior of valuable metals (lithium, cobalt, nickel, and manganese) and impurities (aluminum and copper).

3.2. The Effect of Citric Acid ($C_6H_8O_7$) Concentration on the Leaching of the Untreated and Treated Samples

Figure 2 shows the metal leaching yield at 1 M and 2 M of citric acid. The tendencies in both cases are similar, and the leaching efficiencies are also comparable; for example, with 1 M $C_6H_8O_7$ (Figure 2a), 60% Al, 33% Cu, 18% Li, 12% Co, 12% Mn, and 11% Ni were extracted. With 2 M $C_6H_8O_7$ (Figure 2b), 62% Al, 32% Cu, 20% Li, 12% Co, 12% Mn, and 12% Ni were leached. These results indicate that the citrate coordinated all metal ions, as indicated by the thermodynamic analysis (Figure 1). Despite the preferential dissolution of copper and aluminum, their concentrations were ~400 mg/L Cu and ~200 mg/L Al, while the concentrations of valuable metals in both systems were approximately 300 mg/L Li, 700 mg/L Co, 460 mg/L Mn, and 500 mg/L Ni, respectively.

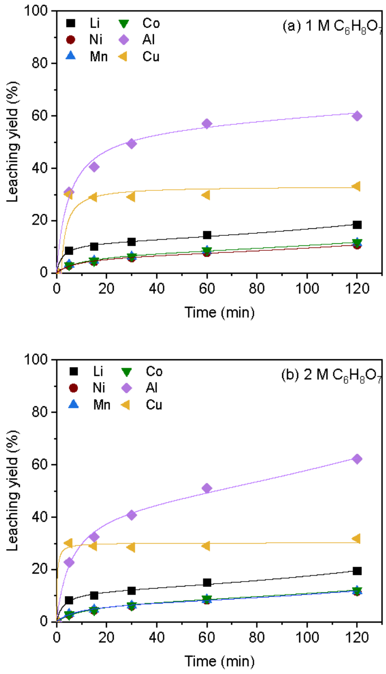

Figure 2. Metal leaching efficiency for the untreated sample. Experimental conditions: (**a**) 1 M $C_6H_8O_7$ and (**b**) 2 M $C_6H_8O_7$. With 50 g/L, 300 rpm (mechanical agitation) at 25 °C in both systems.

Previous results (Figure 2) demonstrated that increasing the concentration of the leaching agent did not improve metal extraction or achieve selectivity. Thus, experimental tests were conducted using mechanically treated samples to study the effect of physical treatment on metal leachability.

Figure 3 shows the leaching test results with 1 M and 2 M of $C_6H_8O_7$ at room temperature (25 °C). In both cases, the sample was milled at two speeds and times (700 and 1000 rpm; 2 and 8 min). Compared to the systems in Figure 2, the leaching yield improved for all metals when mechanical activation was employed, particularly with the higher speeds and longer milling times, 1000 rpm for 8 min (Figure 3).

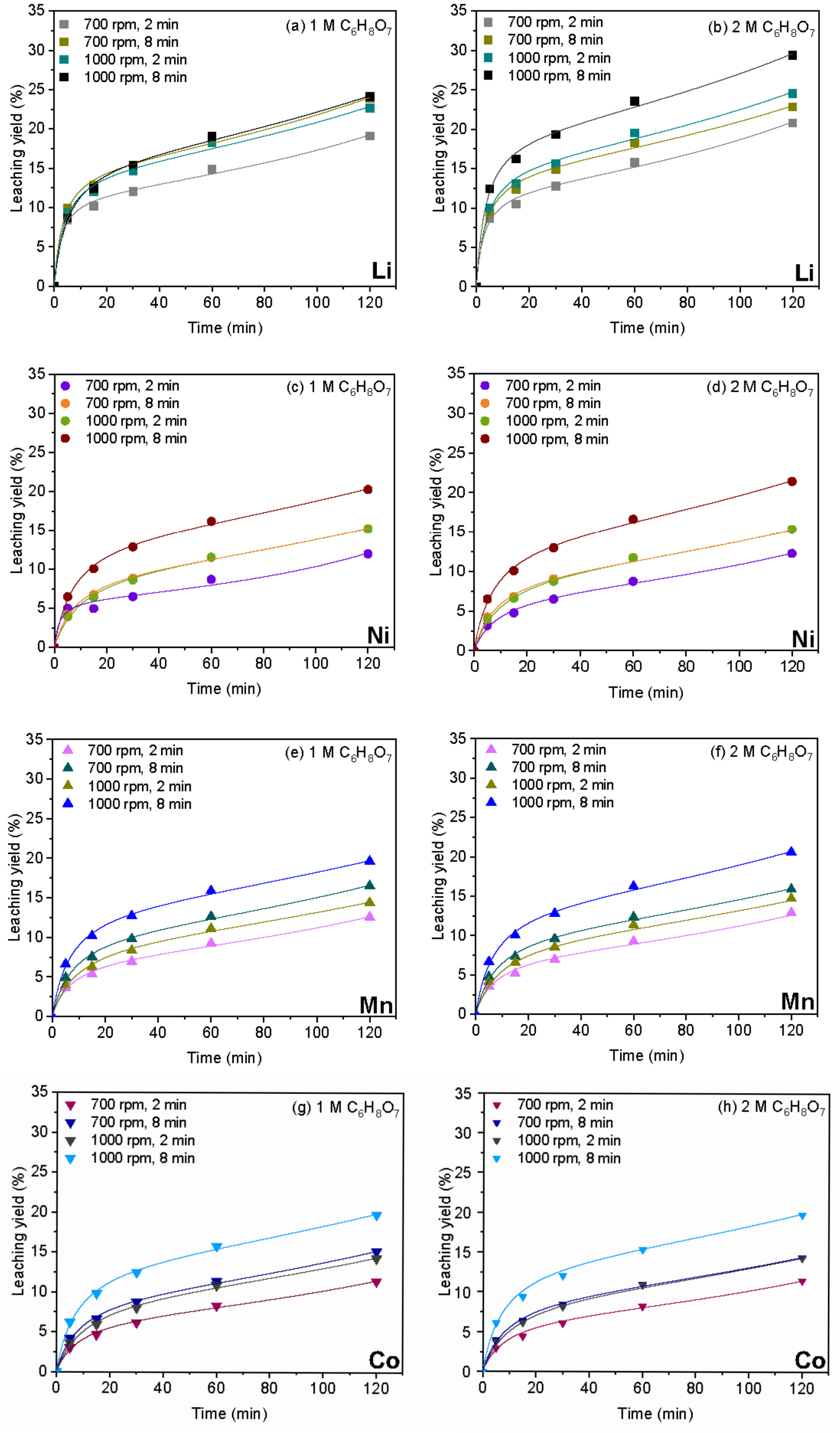

Figure 3. Leaching efficiency of Li, Ni, Mn, and Co for the different mechanically activated samples. Experimental conditions: 1 M $C_6H_8O_7$ (**a**,**c**,**e**,**g**) and 2 M $C_6H_8O_7$, (**b**,**d**,**f**,**h**). All cases with 50 g/L, 300 rpm (mechanical agitation) at 25 °C.

Although the leaching efficiencies of lithium, cobalt, and nickel remained low (<30%), slight differences were observed at varying speeds and milling times. For instance, at

700 rpm for 2 min of milling, lithium efficiency was 19% (343 mg/L), and 29% (485 mg/L) was obtained when the sample was milled at 1000 rpm for 8 min (Figure 3a). When the citric acid concentration increased, 2 M $C_6H_8O_7$, the leaching efficiency of lithium did not enhance significantly; in this case, 21% (364 mg/L) and ~30% (500 mg/L) were obtained at 700 rpm and 2 min, and 1000 rpm and 8 min, respectively, compared to the previous case (1 M $C_6H_8O_7$).

On the other hand, the leaching efficiencies for nickel, manganese, and cobalt were similar (Figure 3c–h). Even so, a milling time of 8 min at 1000 rpm improved the dissolution of the three metals compared to the untreated sample, regardless of the leaching agent concentration. Under these experimental conditions, 20% Ni (900 mg/L), 20% Mn (800 mg/L), and 20% Co (1150 mg/L) were obtained.

Finally, aluminum and copper were also analyzed; according to Figure 4, both metals were extracted over 30%, and their dissolution also improved when speed and milling time increased. The maximum dissolution rate of aluminum was ~80% (273 mg/L) in both chemical systems (Figure 4a,b), and for copper, 70% (960 mg/L) with 1 M $C_6H_8O_7$ and 64% (868 mg/L) with 2 M $C_6H_8O_7$, respectively (Figure 4c,d).

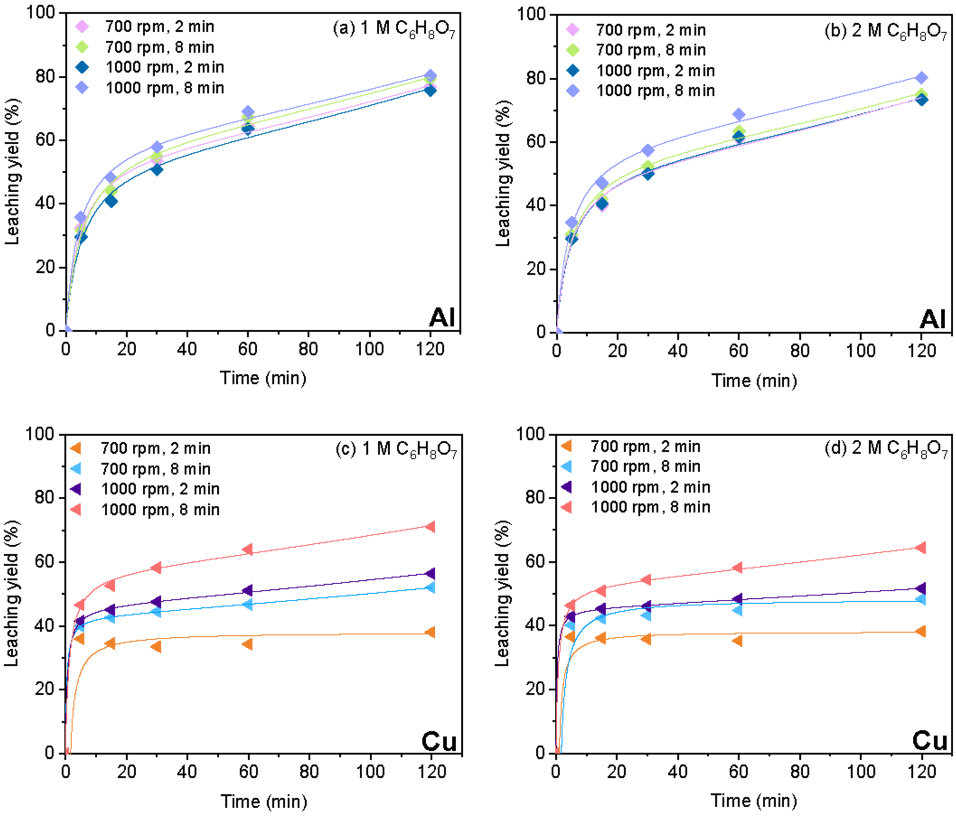

Figure 4. Leaching efficiency of Al and Cu for the different mechanically activated samples. Experimental conditions: 1 M $C_6H_8O_7$ (**a**,**b**) and 2 M $C_6H_8O_7$ (**c**,**d**). All cases with 50 g/L, 300 rpm (mechanical agitation) at 25 °C.

Figures 3 and 4 show that mechanical activation positively affected metal extraction, especially at 1000 rpm and 8 min of milling. This physical pretreatment helps break down agglomerates of black mass, reducing particle size, increasing surface area, and improving the contact between solid and liquid phases [21,22].

Another important aspect is pH, which plays a crucial role in enhancing the formation of M-citrate complexes. In all cases described above (Figures 2–4), the pH was monitored throughout the experiments, with registered values ranging from ~1 to ~2. In this pH range, lithium is in ionic form, while the rest of the metals can coordinate with citrate ion (CoH_2cit^-, Co(Hcit), $Ni(H_2cit)^-$, Ni(Hcit), $Mn(H_2cit)^+$, $Cu_2(cit)_2^{2-}$, and $Al(Hcit)^+$) based on the species distribution diagrams reported in Figure 1.

3.3. Leachability of Valuable Metals in Citric Acid ($C_6H_8O_7$) Solutions at 60 °C

To evaluate how temperature affects metal leaching efficiency, experimental tests were conducted at 60 °C, using 1 M and 2 M $C_6H_8O_7$, with 50 g/L physically treated sample (milling conditions: 1000 rpm for 8 min). In some published works [23–25], it has been reported that citric acid solutions at 60 °C to 95 °C enhance the extraction of valuable metals from spent batteries by over 90%. However, it has also been determined that citrate is chemically unstable at temperatures >50 °C [26]. Additionally, in the presence of certain reductants, like hydrogen peroxide, citrate degradation occurs above 30 °C [15]. Therefore, in this work, a temperature of 60 °C was tested, which is the lower limit of the reported optimal range for promoting metal dissolution.

Raising the temperature (Figure 5) enhanced the leaching efficiency for all metals, obtaining 73% Li (1300 mg/L), 60% Mn and Ni (2300 mg/L Mn and 2700 mg/L Ni), 55% Co (3000 mg/L), 100% Al (400 mg/L), and 90% Cu (1200 mg/L) independently of the citric acid concentration.

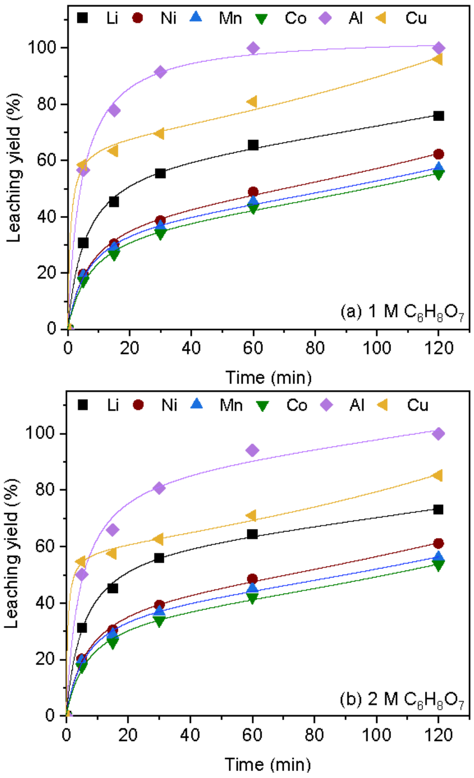

Figure 5. Metal leaching efficiency for the sample mechanically treated at 1000 rpm for 8 min. Experimental conditions: (**a**) 1 M $C_6H_8O_7$ and (**b**) 2 M $C_6H_8O_7$. With 50 g/L, 300 rpm (mechanical agitation) at 60 °C, in both systems.

The temperature modification partially benefits lithium dissolution, but temperatures above 60 °C will require further testing to demonstrate the absence of citric acid degradation. For this reason, this work investigated an additional leaching agent, focusing on developing a methodology that operates at low temperatures (≤60 °C) and avoids the decomposition of organic species.

3.4. Oxalate System as an Alternative to Enhance Lithium Dissolution

Results using citrate solutions (Figures 2–5), demonstrated the possibility of extracting different metals from black mass; however, the leaching process is incomplete and non-selective. For this reason, a second alternative was proposed; oxalic acid ($C_2H_2O_4$) was employed as the leaching agent. A preliminary approximation of this study involved a thermodynamic analysis (Figure 6). According to Figure 6, the oxalate ion (ox) can coordinate all metallic ions similarly to citrate. Nevertheless, oxalate has the particularity of forming insoluble compounds with cobalt ($\mathrm{Co(ox)_s}$), nickel ($\mathrm{Ni(ox)_s}$), manganese ($\mathrm{Mn(ox)_s}$), and copper ($\mathrm{Cu(ox)_s}$) in acid media, pH from 1 to ≥3 (Figure 6b–e), while lithium and aluminum are in ionic or soluble form (Figure 6a,f). This behavior suggests that lithium can be extracted selectively, keeping the other valuable metals in the remaining solid.

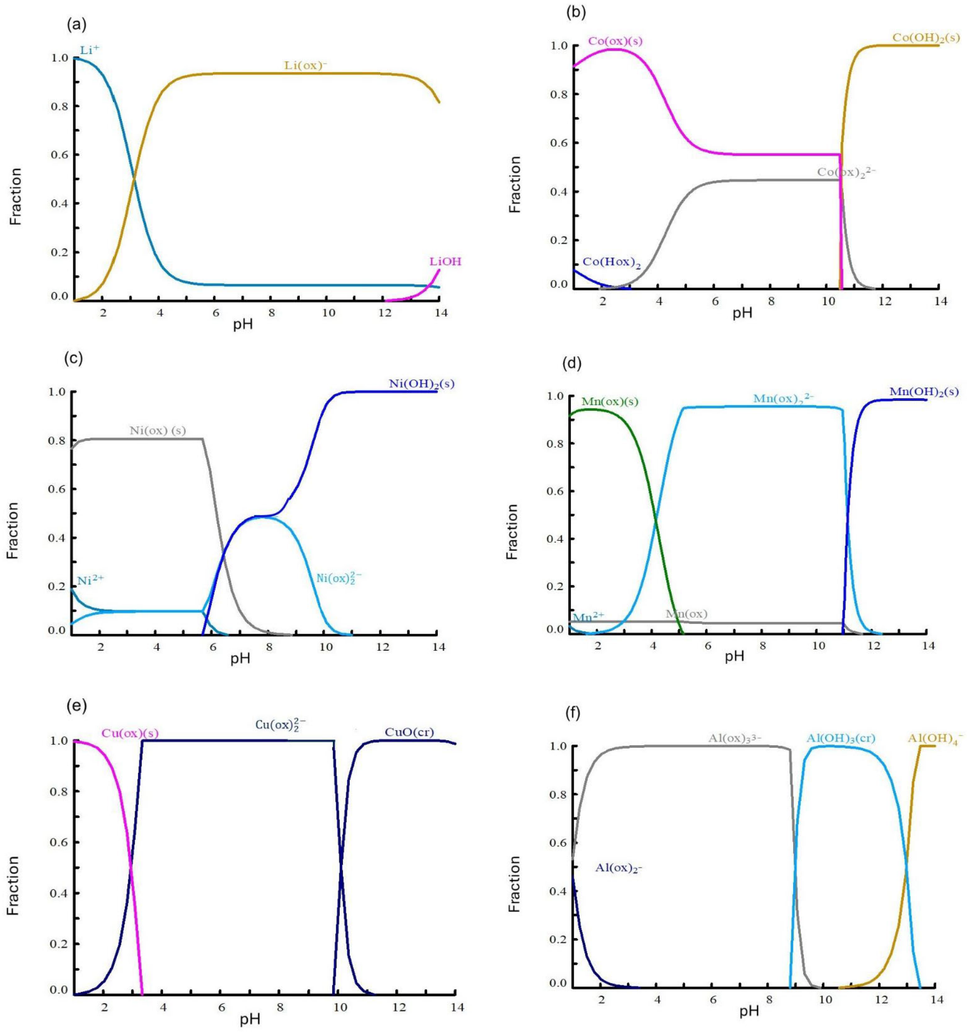

Figure 6. Species distribution diagrams for the metal-oxalate (M-ox) system: (**a**) Li-ox, (**b**) Co-ox, (**c**) Ni-ox, (**d**) Mn-ox, (**e**) Cu-ox, and (**f**) Al-ox. All cases with 1 M ox at room temperature (25 °C). Elaborated using MEDUSA software [20].

The species distribution diagrams (Figure 6) were constructed employing reported data at 25 °C [20]. However, the experiments were conducted at 60 °C, using the same experimental conditions as in the citrate system (sample mechanically treated for 8 min at 1000 rpm, a solid–liquid ratio of 50 g/L, and 1 M of leaching agent), ensuring consistency between the two chemical systems and allowing comparison.

Figure 7 shows the leaching efficiency of the treated sample at different milling conditions (speed and time). Lithium, aluminum, manganese, and copper were dissolved.

Nickel was not extracted, and cobalt efficiency was ≤0.5% in all cases. When the sample was physically treated, lithium extraction after 60 min was complete (100%, 2020 mg/L), and the system reached equilibrium (Figure 7a). Moreover, a clear difference was observed when the sample without mechanical activation was used; in this case, the lithium leaching efficiency was 76% (1660 mg/L).

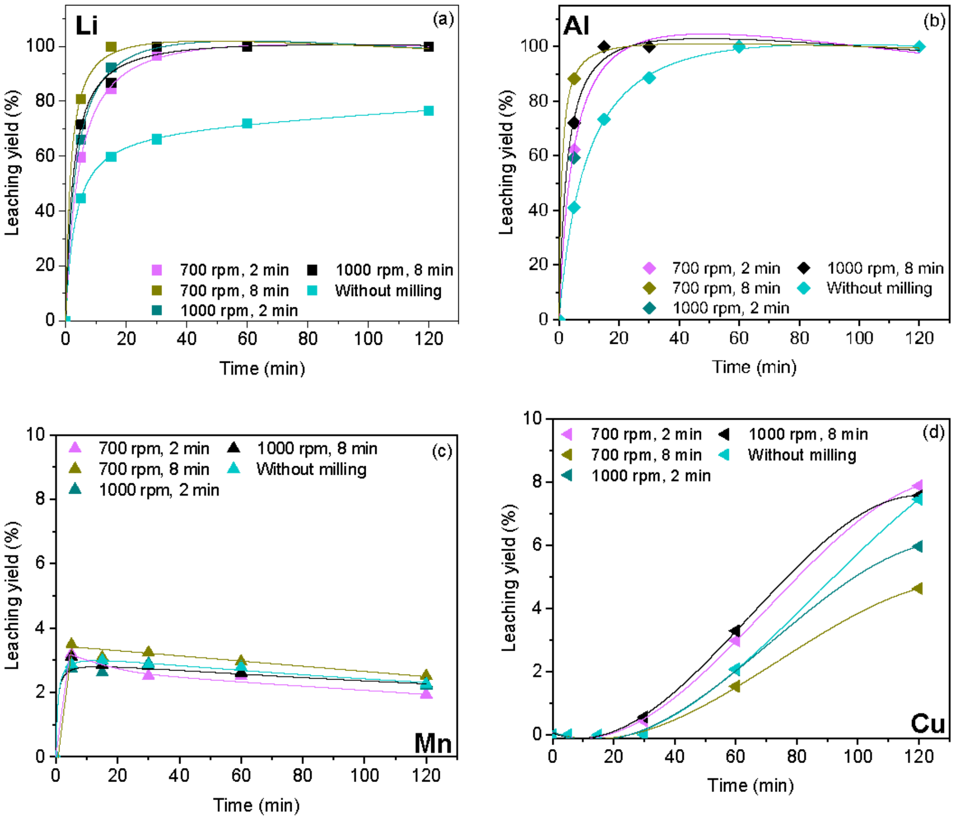

Figure 7. Metal leaching efficiency (**a**) Li, (**b**) Al, (**c**) Mn, and (**d**) Cu for the sample mechanically treated at 1000 rpm for 8 min. Experimental conditions: 1 M $C_2H_2O_4$, 50g/L, 300 rpm (mechanical agitation) at 60 °C.

Aluminum was also entirely extracted in all cases (Figure 7b); despite its high extraction (100%, 400 mg/L), the composition in the original samples is not significant compared to that of valuable metals (Table 1 and Table A1, Appendix A). On the other hand, manganese and copper traces were dissolved, but their leaching efficiency was ≤8% (≤100 mg/L) (Figure 7c,d). Increasing the leaching time favored only copper dissolution. Thus, in the oxalate system, 60 min is enough to achieve complete lithium extraction, which is not affected by other impurities in the leaching system.

Besides, the pH value in oxalate solutions was practically constant (pH~2) throughout the leaching experiments; this behavior promotes the formation of insoluble compounds between oxalate and nickel, manganese, and cobalt ions primarily (Figure 6).

After the leaching process, the solid residues were characterized by XRD and SEM-EDS to analyze the remaining elements. The diffractogram (Figure 8a) showed Co-ox, Ni-ox, and Mn-ox salts, as well as graphite and copper. These results demonstrate structural modifications compared to the diffractogram of the original and untreated black mass sample (Figure A1, Appendix B). On the other hand, SEM analysis (Figure 8b) evidenced polyhedral shapes, suggesting the formation of a mixture of nickel, cobalt, and oxalate [27]. The oxalate compounds correspond to the species reported by Schmitz et al. [28], who used oxalate as a precipitant for the treatment of spent NMC batteries. Finally, the elemental mapping (Figure 8c–h) confirmed that cobalt, manganese, nickel, copper, carbon, and oxygen were the main elements in the solid residue.

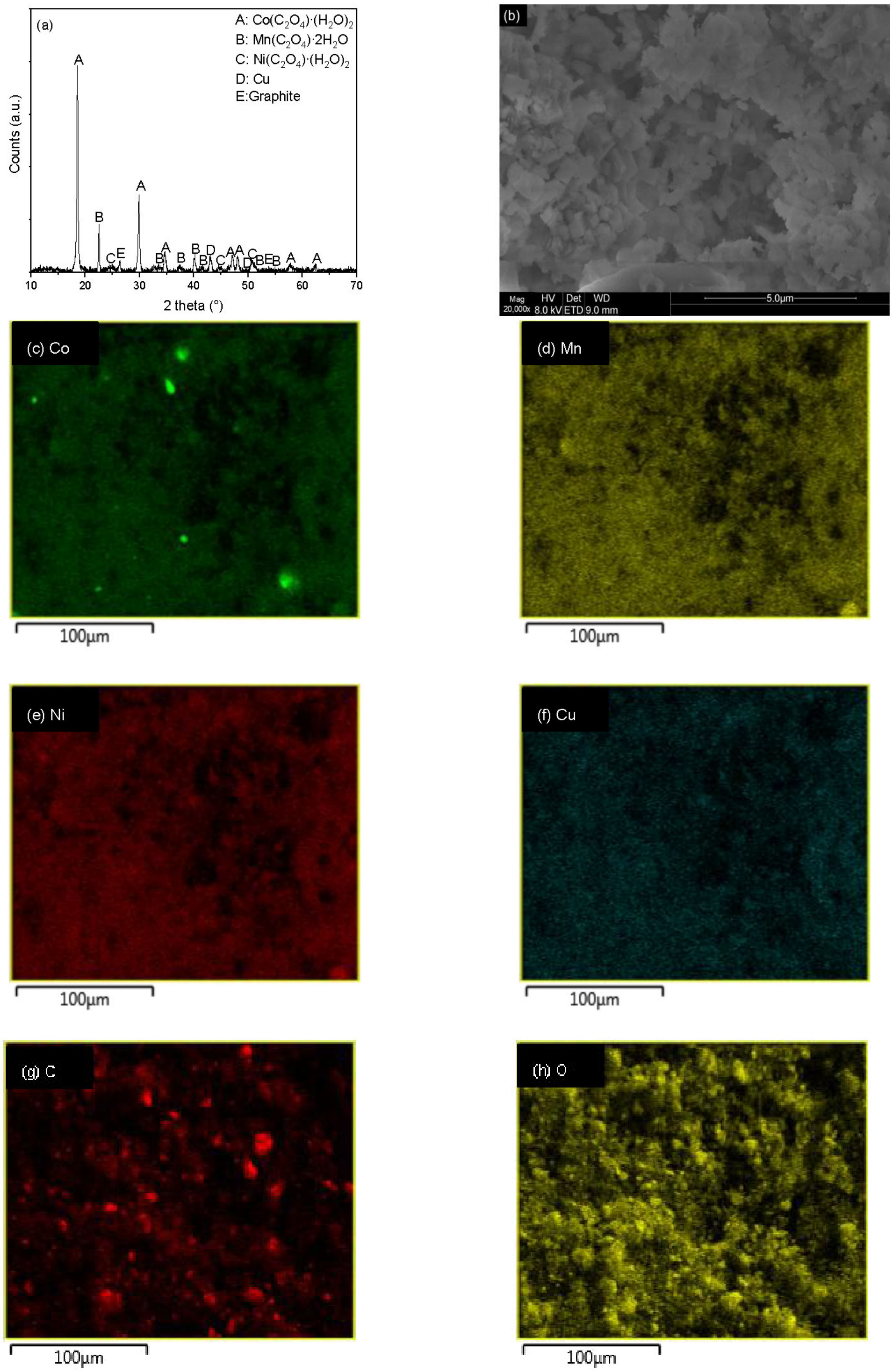

Figure 8. Solid residue characterization (after leaching with oxalic acid: 1 M $C_2H_2O_4$, 50g/L, 300 rpm, and 60 °C). (**a**) XRD analysis ($Co(C_2O_4)\cdot(H_2O)_2$: PDF 04-016-6937, $Mn(C_2O_4)\cdot 2H_2O$: PDF 00-057-0602, $Ni(C_2O_4)\cdot(H_2O)_2$: PDF 04-016-6938, graphite: PDF 04-013-0292, and copper: COD 9013016). (**b**) SEM, (**c**–**h**) EDS elemental mapping.

4. Conclusions

This investigation analyzed the ability of organic acids, particularly citric and oxalic, to extract lithium from spent NMC batteries preferentially. A theoretical and experimental study was described, in which thermodynamic analysis proved useful for understanding the chemical interactions between ionized organic species and metallic ions. Leaching experiments demonstrated that oxalic acid promoted lithium extraction selectively compared to citric acid. On the other hand, mechanical activation was used as a physical treatment to improve lithium dissolution. When the sample was mechanically treated for 8 min at 1000 rpm, and then leached using 1 M $C_2H_2O_4$, 50 g of black mass per liter of solution, 60 °C for 60 min, 100% (2020 mg/L) Li was extracted. The other metals, such as cobalt, nickel, and manganese, formed solid compounds with oxalate, remaining in the solid residue. Although aluminum was also entirely dissolved, its concentration was 400 mg/L; these traces did not affect lithium dissolution.

The proposed methodology is an alternative process for recycling spent lithium-ion batteries, utilizing eco-friendly compounds as leaching agents to achieve high lithium leaching yields. Moreover, additional recovery stages must be incorporated to recover lithium from Pregnant Leach Solution (PLS) and other valuable metals from the solid residue. Finally, this methodology serves as a starting point for improving automotive waste management and recycling techniques that adhere to circular economic principles.

Author Contributions: Investigation, methodology, writing—original draft, data curation, formal analysis, B.S.-B.; investigation, methodology, supervision, L.R. and N.V.; investigation, methodology, validation, K.B. and C.M.; conceptualization, funding acquisition, project administration, writing—review and editing, resources, M.P. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author.

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Conflicts of Interest: Author Nathália Vieceli was employed by the company Northvolt Revolt AB. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Appendix A

The same batch of NMC black mass was treated using an inorganic system; these results have been published [29]. Consequently, Tables 1 and A1 report the same elemental composition as in the published work.

Table A1. Average metallic content of the mechanically treated black mass sample (≤500 µm).

Element (wt%)	Co	Ni	Mn	Li	Cu	Al
Mechanical activation conditions (time and speed of milling)						
2 min, 700 rpm	12.09 ± 0.28	9.43 ± 0.26	8.30 ± 0.14	3.48 ± 0.07	3.08 ± 0.12	0.64 ± 0.04
2 min, 1000 rpm	11.84 ± 0.15	9.29 ± 0.12	8.80 ± 0.12	3.43 ± 0.05	2.87 ± 0.04	0.67 ± 0.01
8 min, 700 rpm	12.25 ± 0.21	9.62 ± 0.16	8.39 ± 0.11	3.63 ± 0.03	3.05 ± 0.03	0.68 ± 0.04
8 min, 1000 rpm	11.62 ± 0.84	8.88 ± 0.63	8.09 ± 0.65	3.45 ± 0.30	2.67 ± 0.19	0.69 ± 0.02

Appendix B

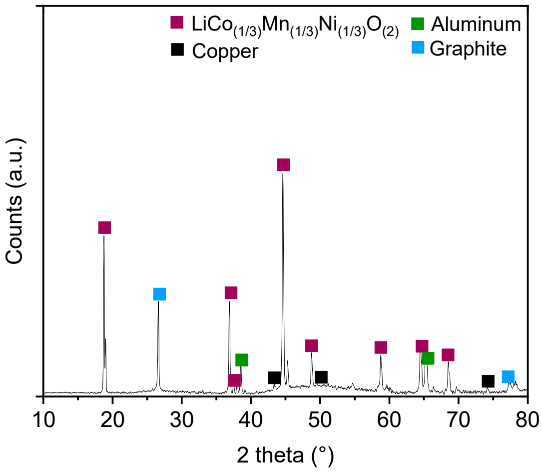

Figure A1. XRD analysis of the original and untreated NMC black mass sample. The identified peaks were $LiCo_{(1/3)}Mn_{(1/3)}Ni_{(1/3)}O_{(2)}$ (PDF 04-013-4379), copper (PDF 00-62-0420), aluminum (COD 90-08-460), and Graphite (PDF 04-016-6937).

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