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Recycling of Li-Ion Batteries: The Effects of Mechanical Activation on Valuable Metals Leachability from the Black Mass of NMC 111

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Abstract

The electric vehicle (EV) market is expected to grow in the coming years, driven by the use of green energy. Nevertheless, this technological advance also leads to an increase in battery waste generation. This work presents a suitable solution to improve recycling techniques. A coupled method involving mechanical activation and acid leaching was evaluated to process a sample of black mass (chemistry nickel-manganese-cobalt, NMC 111). This study analyzed the effect of milling time, leaching agent concentration, and temperature. Around 98% of lithium and ≥ 80% of cobalt, manganese, and nickel were extracted using 1000 rpm and 60 min of milling time, 2 M H_2SO_4, 60 ºC, and 120 min. The experimental conditions also influenced the extraction of copper and aluminum traces; however, they did not affect the dissolution of valuable metals, especially lithium.

Keywords Lithium-ion battery recycling · Mechanical activation · Valuable metals extraction · Acid leaching

1 Introduction

Over the past decade, the lithium-ion batteries (LiBs) market has significantly grown, driven by the increasing use of batteries in electric vehicles (EVs) [1]. The Nordic area has one of the world's highest market shares of EVs, and projections indicate that by 2030, around 4 million electric vehicles will operate in this region. Consequently, current recycling methods are challenged to meet the European Union's priorities of promoting an environmentally friendly battery value chain [2].

So far, hydrometallurgical methods can achieve high recovery rates of valuable metals such as lithium, cobalt, manganese, and nickel and produce battery-grade purity compounds. Before this chemical treatment, discharged batteries usually undergo pretreatment where non-metallic components (plastic, separator, polymer tapes, and binder) and metallic materials (steel casing, nickel and aluminum tabs, and copper, and aluminum current collector) are removed [3]. Traditionally, separation is usually done by sieving, a mechanical method employed after the disassembly and crushing of discharged spent batteries [4]. Also, additional mechanical treatments can be employed to improve valuable metal dissolution. For example, mechanical activation is considered a proficient physical method that benefits the extraction of cobalt and lithium from various types of batteries [5, 6].

From a chemistry point of view, mechanical activation allows the breakage of atomic bonds in the crystal lattice of solids, promoting a decrease in the activation energy and increasing the leaching efficiency [7]. This technique is also considered a competent treatment because it reduces the particle size, increases the surface area, and enhances the mass transport phenomena; however, high energy consumption is considered the main drawback of this physical process [8, 9].

Yan et al. studied the effect of two-step crushing followed by hammer crushing, obtaining 100% dissociation of battery compounds in 10 s [10]. While Guan et al. proposed a methodology for recovering cobalt and lithium from lithium-cobalt oxide (LCO) batteries by mechanical activation and acid leaching [11]. The physical treatment was performed using a planetary ball mill, and the leaching system was sulfuric acid and hydrogen peroxide ($H_2SO_4 + H_2O_2$). The

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optimal experimental conditions were 700 rpm, 60 min, 1 M H_2SO_4, 2 vol% H_2O_2, and room temperature (25 °C), obtaining 93% Co and 100% Li. This work demonstrated that high speeds and time (over 700 rpm and 120 min) contribute to the agglomeration of particles. This phenomenon can impact leaching efficiency in the second phase, and prolonged milling time leads to increased energy consumption.

On the other hand, Yu et al. analyzed lithium extraction from lithium iron phosphate (LFP) batteries using a ball-milling method (300 rpm for 60 min). After mechanical treatment, the LFP sample was leached in two stages. The first one was useful for aluminum removal, using sodium hydroxide solutions (NaOH, 2 M). In the second leaching stage, the solid residue was leached with 0.3 M of sulfuric acid (H_2SO_4) and 0.45 M of sodium persulphate ($Na_2S_2O_8$) at 60 °C for 1.5 h. 97.3% Li and < 5% of iron and phosphorus were extracted [12]. This investigation evidenced that traces of aluminum, iron, and phosphorus directly impact lithium dissolution, and they should be removed from the leaching liquors.

Sulfuric acid solutions have shown effective metal extraction, particularly from nickel-manganese-cobalt (NMC) batteries [13], Eq. 1). In most cases, this reagent has been combined with different reductants, such as hydrogen peroxide (H_2O_2), hydrazine sulfate ($N_2H_6SO_4$), ferrous ion (Fe^{2+}), ammonium chloride (NH_4Cl), etc. These chemical systems have allowed high extraction efficiencies (> 90%) of cobalt, manganese, nickel, and lithium [14–18].

$$6LiNi_{1/3}Mn_{1/3}Co_{1/3}o_{2(s)} + 9H_2SO_{4(aq)} \rightarrow 2NiSO_{4(aq)} + 2MnSO_{4(aq)} + 2CoSO_{4(aq)} + 3LiSO_{4(aq)} + 9H_2O_{(aq)} + 3/20_{2(g)} \quad (1)$$

In NMC battery treatment, the extraction of manganese, cobalt, and nickel is more challenging than lithium dissolution. It has been established that lithium can be released from the crystal structure and interacts with hydronium ions (H^+). Afterward, the other metals react, and the crystal structure can be progressively destroyed [14]. Therefore, this work investigates the effect of mechanical activation (milling) on the liberation of valuable metals and uses sulfuric acid to extract cobalt, manganese, nickel, and lithium from the black mass (NMC 111). The influence of physical treatment on the dissolution of impurities, like copper and aluminum, is also analyzed. This work presents an alternative solution for developing more efficient recycling techniques.

2 Materials and Methods

2.1 Solid Sample Characterization

150 kg of EV discharged lithium-ion battery packs (NMC 111) were provided by Volvo Cars AB (Sweden) and then dismantled down to the cell level by Stena Recycling AB (Sweden). Afterward, the cells (120 kg) were processed through three stages: crushing, mechanical sieving, and magnetic separation by Akkuser Oy (Finland). The fine fraction (black mass) obtained represented 58.5% of the initial weight and was constituted from the cathode and anode with the current foils and separator.

The black mass was further sieved at ≤ 500 µm to obtain a homogeneous powder and to simulate an industrial sample of today's standards. The sample chemical analysis was carried out (in triplicate) employing 30 mL of aqua regia prepared with hydrochloric acid (HCl, 37%) and nitric acid (HNO_3, 65–67%) (HCl:HNO_3, 3:1 v/v), where 0.2 g solid powder was introduced and reacted for 6 h at 80 ºC, using magnetic agitation (200 rpm). The solution was cooled to room temperature overnight, then filtered (PTFE syringe filters with a particle size retention of 0.45 µm), diluted (with 0.1 M HNO_3, 65–67%), and analyzed by Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES, Thermo Fisher Scientific, Model iCAP™ 6000 Series).

To study the potential deformations of the crystal lattice structure due to milling, the black mass without and with mechanical activation was analyzed using X-ray crystallography (XRPD Siemens D5000 diffractometer): Cu ($\lambda = 1.54$ Å) radiation source in a 2θ range of 10°–80° with a rotational speed of 15 rpm. The operating current and voltage were 40 mA and 40 kV, respectively. EVA software and the JCPDS database were used for analytical interpretation.

2.2 Mechanical Activation: Milling

The milling process was conducted using the Vibratory Disc Mill RS 200 by Retsch®, which operates through circular horizontal vibrations powered by a 1.5 kW three-phase motor. The equipment features a jar with a capacity of 100 g and is fitted with two concentric grinding rings. A centrifugal force acts on the rings, creating extreme pressure, which allows for effective sample fractionation. The experiments were carried out using 50 g of black mass per tested batch. This amount of sample ensures a homogeneous powder and minimizes material loss. The samples were ground for 2, 8, and 60 min at two speeds of 700 and 1000 rpm.

2.3 Acid Leaching

The black mass was processed in sulfuric acid solutions (H_2SO_4, 97%), employing three double-wall PVDF reactors MEAB® (100 mL capacity) to guarantee repeatability. For temperature control, an Optima™ heated circulating bath model T100-P12 was utilized. A solid–liquid ratio of 50 g/L was used, and mechanical agitation was employed (300 rpm). This work focused on the analysis of two parameters: temperature (25 °C and 60 °C) and H_2SO_4 concentration (1 M and 2 M). During the leaching process, samples were taken at 5, 15, 30, 60, and 120 min. Then, each sample was filtered (PTFE syringe filters with a particle size retention of 0.45 µm), diluted with 0.1 M HNO_3, 65–67%), and analyzed by ICP-OES (Thermo Fisher Scientific, Model iCAP™ 6000 Series).

3 Results and Discussion

3.1 Sample Characterization

Table 1 shows the average weight percentage (wt%) of metal composition in untreated (mechanically) and treated black mass samples. Cobalt, manganese, and nickel were quantified, and traces of lithium, copper, aluminum, and iron were also detected. The composition data reported in Table 1 presented slight differences; this fact can be attributed to the heterogeneity of the sample and the distribution of metals within the matrix.

The samples were characterized by X-ray diffraction to analyze the deformation of the crystalline structure due to the milling process. The diffractograms are presented in Fig. 1, which shows no significant crystal lattice structure modifications; only the peak intensity changes were identified at 26.2° and 77.1°.

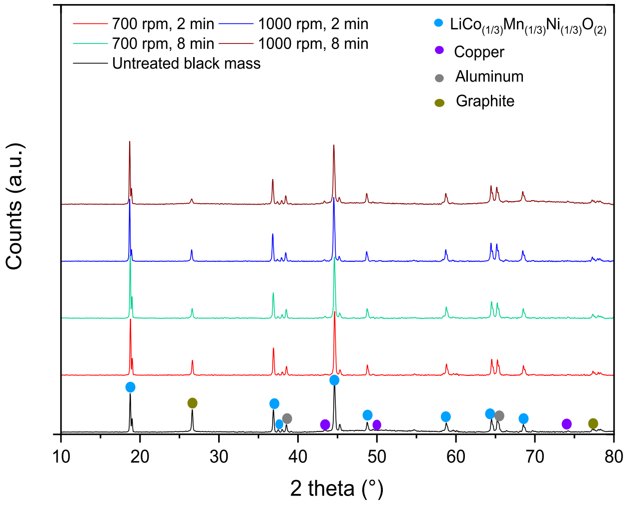

Fig. 1 XRD pattern for the untreated and mechanically treated black mass under different milling conditions. Peaks correspond to graphite (PDF 04–016-6937), NMC111-LiNi1/3Co1/3Mn1/3O2 (PDF 04–013-4379), aluminum (COD 90–08-460), and copper (PDF 00–62-0420)

3.2 Leaching of Valuable Metals from the Untreated Sample

The untreated sample was leached with H_2SO_4 solutions. Figure 2 shows the kinetic curves of metal extraction for the systems at 25 ºC and two concentrations of H_2SO_4 (1 and 2 M). With 1 M H_2SO_4 after 120 min, the leaching efficiency was 40% Co, 42% Ni and Mn, and 69% Li (Fig. 2a). The acid media guaranteed the preferential dissolution of lithium. To improve its dissolution, the H_2SO_4 concentration was modified to 2 M (Fig. 2b). The results demonstrated that the reagent concentration enhanced the leaching efficiency of lithium (~ 85%). However, cobalt, manganese, and nickel behaved similarly to the previous

Table 1 Elemental composition of the black mass sample (≤ 500 µm)

Mechanical activation conditions	Untreated sample	2 min, 700 rpm	2 min, 1000 rpm	8 min, 700 rpm	8 min, 1000 rpm
Element (wt%)					
Co	12.00 ± 0.30	12.09 ± 0.28	11.84 ± 0.15	12.25 ± 0.21	11.62 ± 0.84
Ni	9.30 ± 0.20	9.43 ± 0.26	9.29 ± 0.12	9.62 ± 0.16	8.88 ± 0.63
Mn	8.10 ± 0.10	8.30 ± 0.14	8.80 ± 0.12	8.39 ± 0.11	8.09 ± 0.65
Li	3.70 ± 0.10	3.48 ± 0.07	3.43 ± 0.05	3.63 ± 0.03	3.45 ± 0.30
Cu	3.00 ± 0.10	3.08 ± 0.12	2.87 ± 0.04	3.05 ± 0.03	2.67 ± 0.19
Al	0.80 ± 0.10	0.64 ± 0.04	0.67 ± 0.01	0.68 ± 0.04	0.69 ± 0.02
Fe	0.16 ± 0.05	0.20 ± 0.06	0.10 ± 0.02	0.19 ± 0.08	0.10 ± 0.02

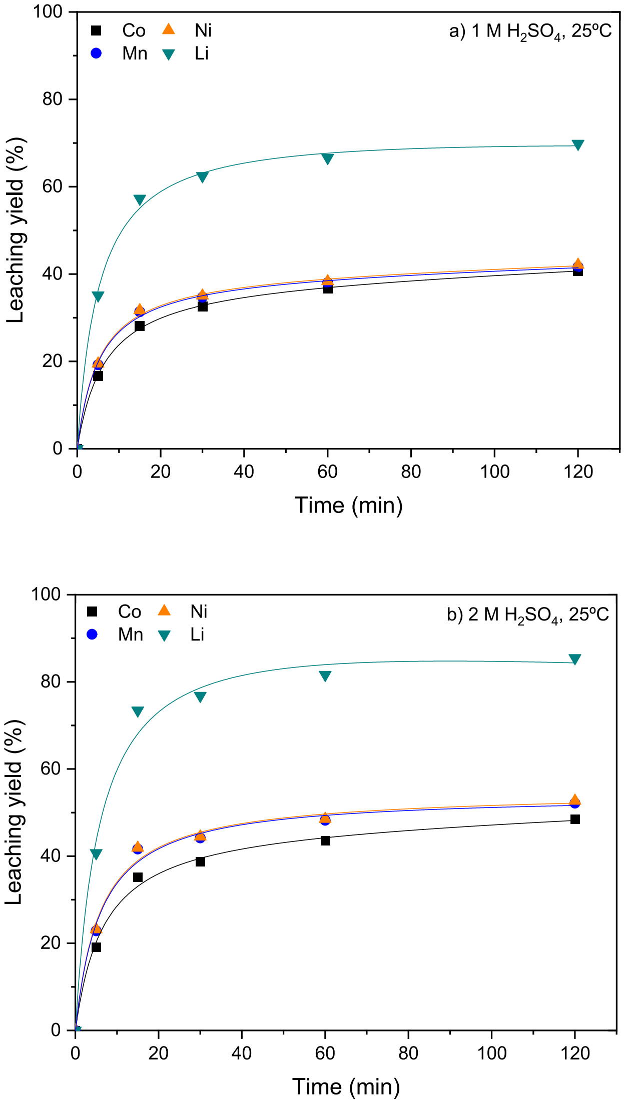

Fig. 2 Leaching efficiency of cobalt, manganese, nickel, and lithium. **a**) 1 M H_2SO_4 and **b**) 2 M H_2SO_4, both systems at 25 °C

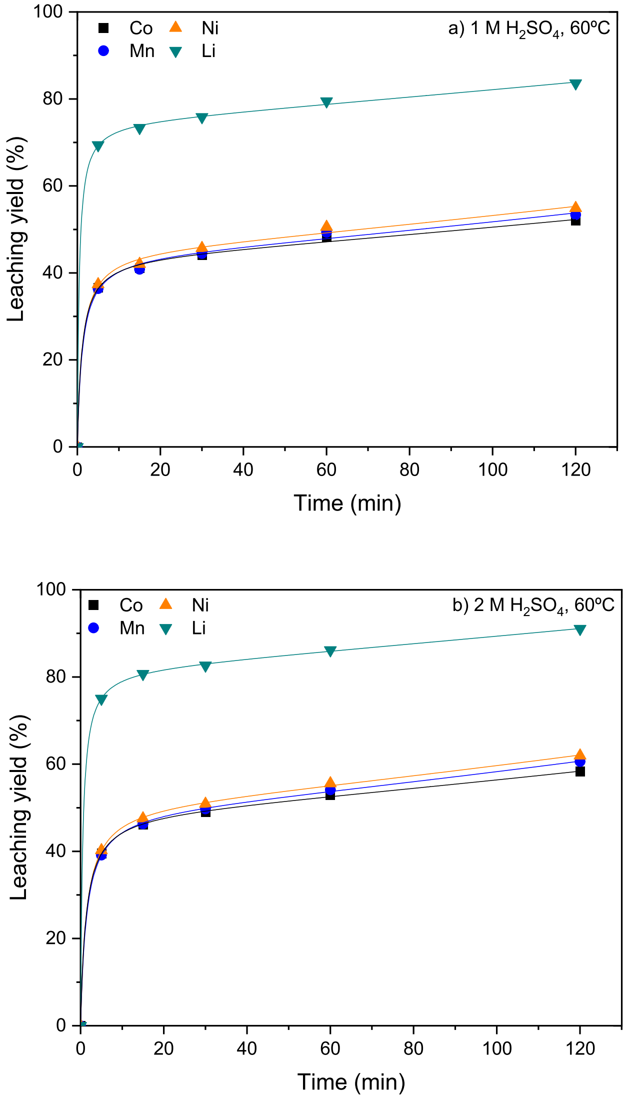

Fig. 3 Leaching efficiency of cobalt, manganese, nickel, and lithium. **a**) 1 M H_2SO_4 and **b**) 2 M H_2SO_4, both systems at 60 °C

system (1 M H_2SO_4); in this case, the leaching efficiency was 48% Co and, 52% Mn and Ni.

On the other hand, some studies report that improving lithium extraction in inorganic acid media requires increasing the temperature between 50–160 °C [4, 14, 19]. Therefore, the experiments were performed at 60 ºC. As the temperature increased, the metal leaching efficiency also increased in both cases, 1 M and 2 M H_2SO_4 (Fig. 3), particularly for lithium. Its leaching efficiency was 83%, with 1 M H_2SO_4 (Fig. 3a) and 91% using 2 M H_2SO_4 (Fig. 3b). The leaching yield for the rest of the metals was ~ 52% Co, 54% Ni and Mn (Fig. 3a), and 58% Co, 61% Ni and Mn (Fig. 3b).

Previous leaching systems at room temperature and 60 ºC demonstrated that the kinetic curves of cobalt, nickel, and manganese behaved similarly, while the leaching efficiency of lithium improved (91% Li) when the temperature and sulfuric acid concentration increased. Lin et al. proposed that cobalt, nickel, and manganese react partially with H_2SO_4, while the chemical reaction with lithium is favored because this metal releases from the solid structure as Li^+ and enhances its interaction with sulfate ion (SO_4^{2-}) if the temperature increases ($LiSO^-_{4(aq)}$, $\Delta G°$ −5.09 kJ/mol; ΔG (60 °C) = −4.93 kJ/mol,) [20, 21]; this fact explains the metal's leaching efficiency in Fig. 2 and Fig. 3.

3.3 Effect of Mechanical Activation on Valuable Metals Leachability

The effect of mechanical activation was studied considering two milling times and speeds: 2 and 8 min, 700 and 1000 rpm.

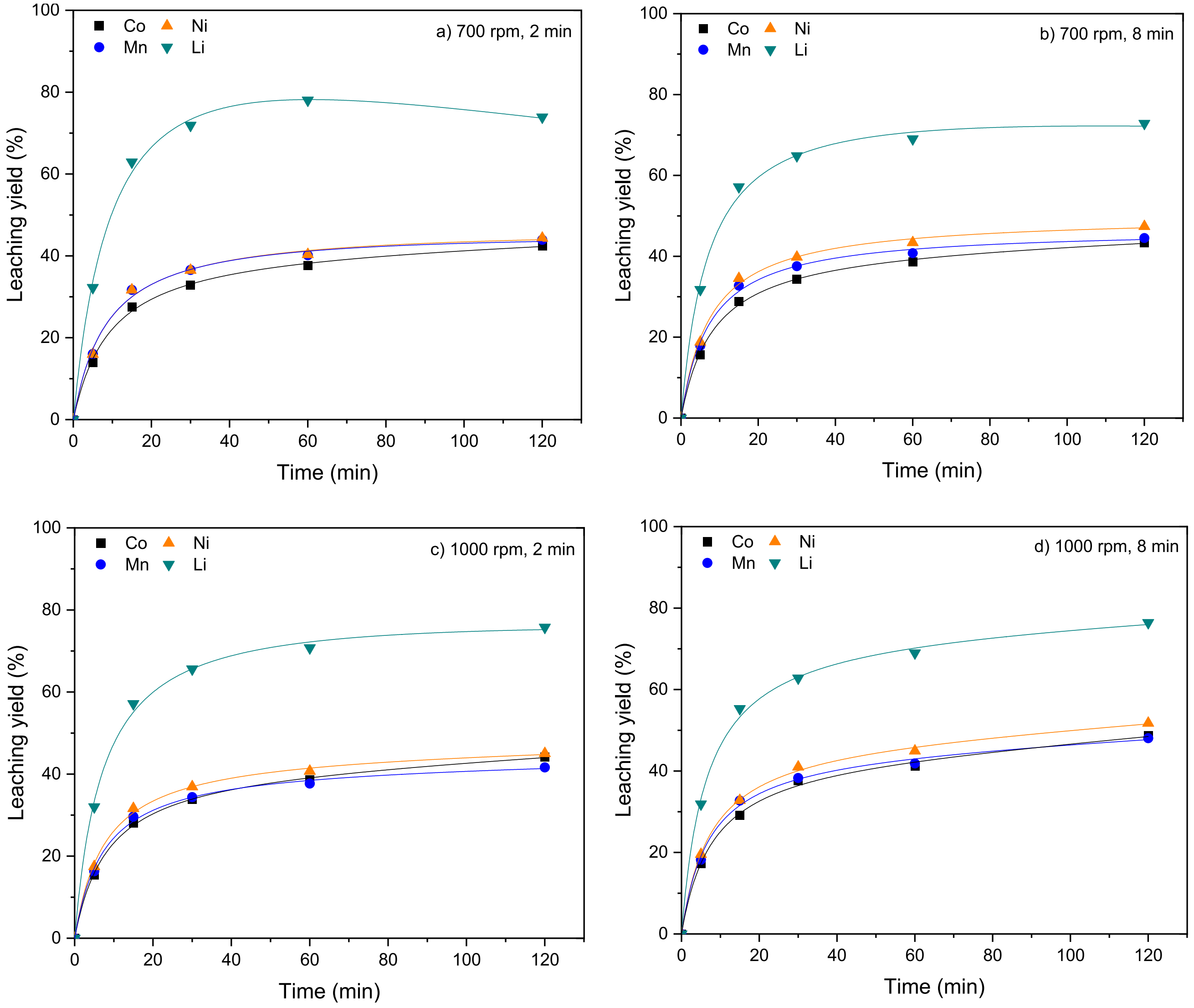

Fig. 4 Leaching efficiency of cobalt, manganese, nickel, and lithium. Mechanical activation conditions: **a**) 700, 2 min; **b**) 700, 8 min; **c**) 1000, 2 min; and **d**) 1000 rpm, 8 min. All systems with 1 M H_2SO_4 at 25 ºC

Figure 4 shows the leaching efficiency of cobalt, manganese, nickel, and lithium when the system was operated at room temperature and the Sulfuric acid concentration was 1 M. Lithium extraction was favored in all cases. Its maximum leaching efficiency was 76% when 1000 rpm and 8 min of milling were employed (Fig. 4d). These experimental conditions enhanced the metal extraction compared to the previous system (Fig. 2); however, the leaching yield for cobalt, nickel, and manganese was < 60% (48% Co and Ni, 52% Mn).

To comprehend the influence of sulfuric acid on metal dissolution, its concentration was adjusted to 2 M, and the tests were conducted under the same experimental conditions as those described above. The leaching efficiency of cobalt, manganese, and nickel (Fig. 5) did not present significant differences in comparison to the system at room temperature and 1 M H_2SO_4 (Fig. 4). Although the lithium dissolution was incomplete (72% Li), the results indicate that its extraction is directly influenced by high speed or high temperature (Fig. 3b, 4d, and 5d).

Based on the previous analysis, additional tests were carried out using 2 M H_2SO_4, 1000 rpm, 2 min, and the system operating at 60 ºC. In this case, the leaching yields were 98% Li, 55% Co, 51% Mn, and 54% Ni, respectively (Fig. 6). The experimental conditions favored lithium dissolution, achieving its extraction almost completely; however, this was not the case for the other metals.

The chemical systems described previously demonstrated that the extraction of cobalt, nickel, and

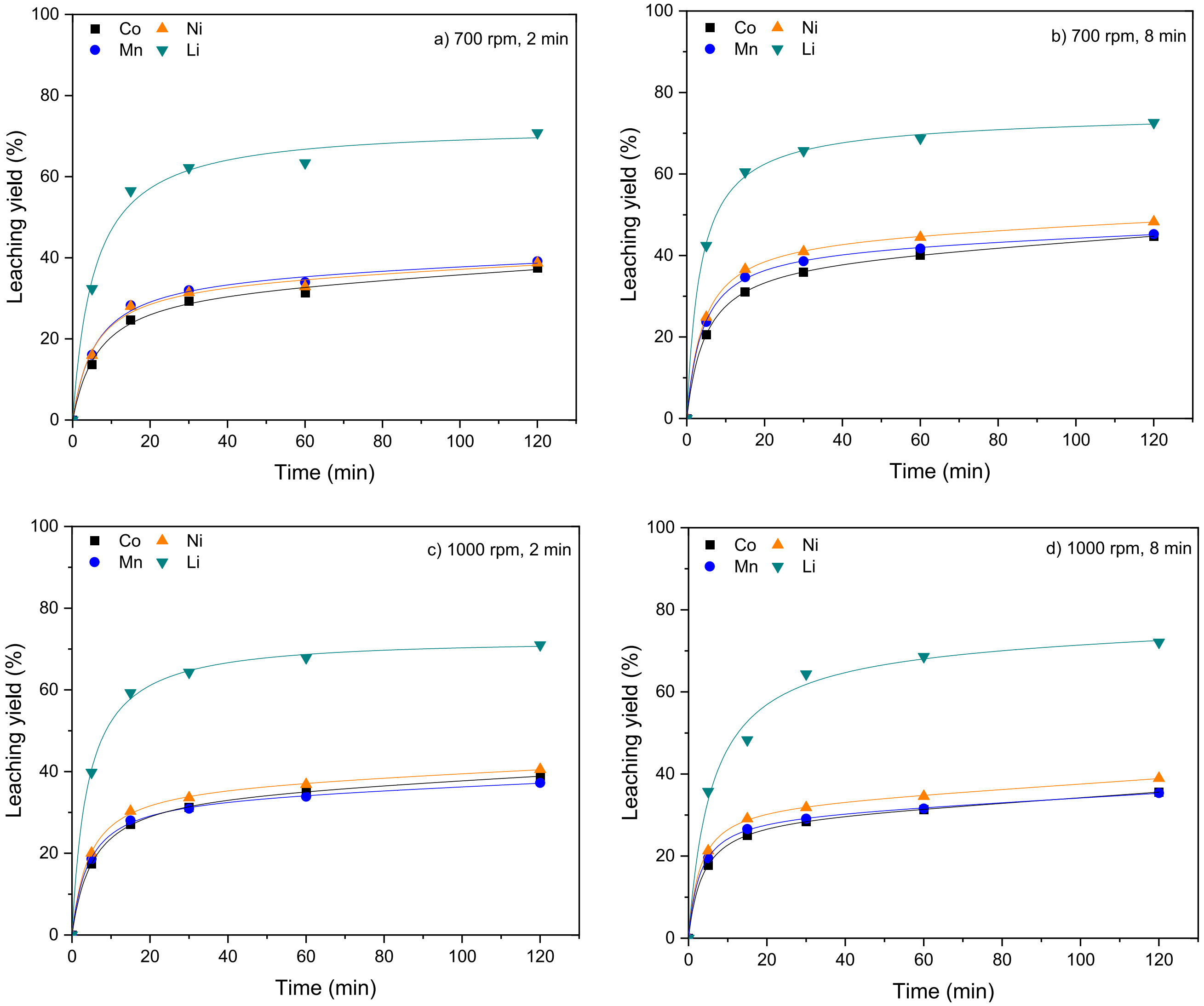

Fig. 5 Leaching efficiency of cobalt, manganese, nickel, and lithium. Mechanical activation conditions: **a**) 700, 2 min; **b**) 700, 8 min; **c**) 1000, 2 min; and **d**) 1000 rpm, 8 min. All systems with 2 M H_2SO_4 at 25 ºC

manganese is affected by temperature. In contrast, lithium dissolution is influenced by mechanical activation and temperature. Specifically, temperature enhances the leaching kinetics of lithium, even though the processing of the untreated and treated samples (Fig. 3b and Fig. 6). Despite mechanical activation being employed, cobalt, nickel, and manganese dissolution in the proposed systems did not benefit significantly. Guan et al. and Yu et al. Suggested 60 min as optimal milling time, using 700 and 300 rpm, respectively [11, 12]. Moreover, Guan et al. determined by XRD analysis that the intensity of the peaks decreased, and their Full Width at Half Maximum (FWHM) broadened, indicating that the particles were destroyed gradually and contributed to forming crystal defects, which improved the leaching process [11]. In this work, the sample was physically treated by increasing the milling time (60 min) at two tested speeds, 700 and 1000 rpm. After the physical treatment, the samples were analyzed by the XRD technique (Fig. 7) and chemically (Appendix). The diffractogram indicated that at high speed, 1000 rpm, the intensity of certain peaks decreased (specifically at 18.7°, 26.2°, 36.7°, 44.48°, 58.8°, 64.4°, and 65.1°), but no modifications of FWHM were identified.

Leaching tests were conducted using the treated sample with the new mechanical activation conditions: 60 min

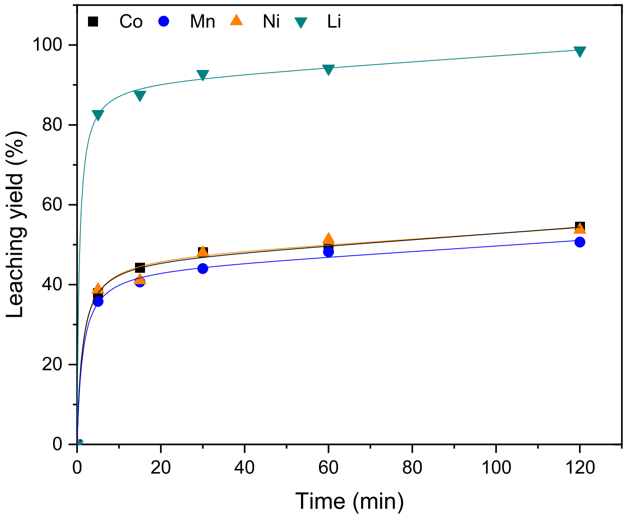

Fig. 6 Leaching efficiency of cobalt, manganese, nickel, and lithium. Experimental conditions: 1000 rpm, 8 min, with 2 M H_2SO_4 at 60 ℃

milling time at 700 and 1000 rpm, and 2 M H_2SO_4 at 60ºC (Fig. 8). The extended milling contributed positively to the metal dissolution, especially for cobalt, nickel, and manganese, compared to the results reported in Fig. 6. The leaching efficiency of the metals was ≥ 77% (700 rpm, 60 min: 77% Co, 82% Ni, and 80% Mn; 1000 rpm, 60 min: 81% Co, 84% Ni, and 80% Mn, Fig. 8a and Fig. 8b), and the extraction rate of lithium was 98% regardless of the milling speed (Fig. 8a and Fig. 8b). However, with the speed of 1000 rpm, the lithium extraction rate was faster, and after 30 min, its dissolution reached equilibrium (Fig. 8b). In contrast, the leaching yields of cobalt, nickel, and manganese continued to increase after 60 min, indicating a slower dissolution rate compared to lithium. Therefore, these experimental conditions demonstrated that increasing milling time improves metal liberation and results in high leaching yields (≥ 80%).

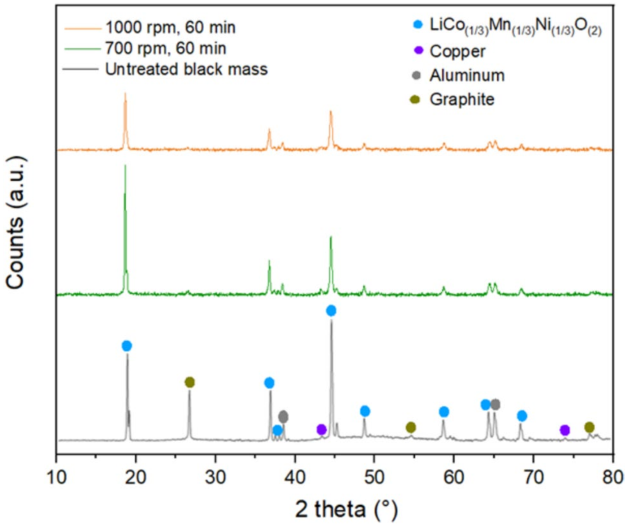

Fig. 7 XRD pattern for untreated and treated black mass, using 700 and 1000 rpm for 60 min. Peaks correspond to graphite (PDF 04–016-6937), NMC111-LiNi1/3Co1/3Mn1/3O2 (PDF 04–013-4379), aluminum (COD 90–08-460), and copper (PDF 00–62-0420)

3.4 Influence of Mechanical Activation on Metallic Impurity Dissolution

Battery recycling also involves the analysis of metallic impurities, like copper and aluminum, and their impact on leaching selectivity. Both metals are the most common impurities because they are essential components of LIBs, and their composition is usually Superior to that of other metals. For example, a generic battery module NMC 111 contains approximately 17.9% aluminum in the cell housing, 4.8% aluminum in the cathode, and 8.4% copper in the anode [22]. In the black mass sample, the content of these impurities is not significant compared to the valuable metals (Table 1 and Table 2), but copper and aluminum traces can also react with SO_4^{2-}, forming soluble species (21, Eq. 2 and 23, Eq. 3) and accumulate in the Pregnant Leach Solution (PLS).

$$2Al_{(s)} + 3H_2SO_{4(aq)} \rightarrow Al_2(SO_4)_{3(aq)} + 3H_{2(g)} \Delta G^\circ = -1029.81 kJ/mol \tag{2}$$

$$Cu_{(s)} + 2H_2SO_{4(aq)} \rightarrow CuSO_{4(aq)} + SO_{2(g)} + 2H_2O_{(aq)} \Delta G^\circ = -56.74 kJ/mol \tag{3}$$

The leaching efficiency of copper and aluminum in the different chemical systems was also analyzed. The temperature and a longer milling time positively affected metal dissolution, accomplishing an extraction of 100% (Fig. 9). For example, aluminum was dissolved entirely in the first system (1 M H_2SO_4, 700 rpm, 8 min of milling time, 60 ºC, Fig. 9a) after 30 min. Otherwise, the copper leaching rate was slower, but after 120 min, 96% Cu was obtained. The complete dissolution of aluminum is attributed to the increased surface area due to the milling effect, which enhances solid–liquid contact. Meanwhile, copper dissolution can be related to passivation phenomena.

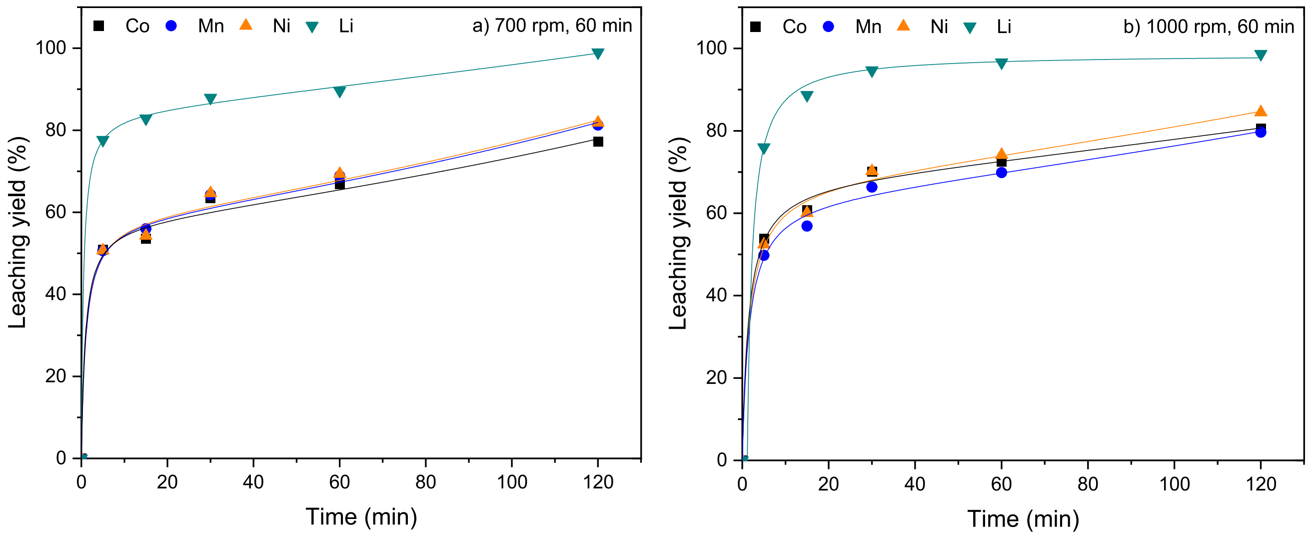

Fig. 8 Leaching efficiency of cobalt, manganese, nickel, and lithium. Experimental conditions: **a**) 700 rpm and **b**) 1000 rpm, 60 min milling time, and 2 M H_2SO_4 at 60 ℃

This leads to the formation of copper oxides, sulfides, or sulfates ($Cu_2O_{(s)}$, $CuO_{(s)}$, $Cu_2S_{(s)}$, $CuS_{(s)}$, and $CuSO_{4(s)}$) on the metal surface, impacting leaching efficiency [23]. When the milling speed was adjusted (Fig. 9c), the total leachability of both metals was guaranteed. These data are comparable when the H_2SO_4 concentration was modified (2 M) (Fig. 9b and 9 d), even with a longer milling time (60 min), the extraction kinetics of copper and aluminum improved. Therefore, these data demonstrated the influence of mechanical activation on impurity dissolution.

4 Implications of Mechanical Activation and Acid Leaching on Selective Valuable Metal Extraction

Strong acids and high temperatures (leaching process: 50–160 °C, pretreatment phases: 100–1500 °C) are commonly used to extract several metals from spent batteries [24]. In this investigation, mechanical activation has been used as an alternative to achieve the leachability of valuable metals, such as cobalt, nickel, lithium, and manganese. It has been demonstrated that the feasibility of combining milling and acid leaching to ensure the extraction of cobalt, nickel, manganese, and lithium from a black mass sample. ~ 100% Li and ≥ 80% of cobalt, nickel, and manganese were dissolved with 2 M H_2SO_4, 60 ℃, and 120 min; however, impurities dissolution was also reported.

The presence of undesirable metal ions in PLS requires additional effort during the refinement stages, where leaching liquors must be treated in multiple purification phases. For example, copper, aluminum, and other impurities can be removed using electrowinning, solvent extraction, cementation, precipitation, etc. Afterward, the same techniques can be applied to recover valuable metals [25]. Basically, a considerable number of stages involve higher energy and chemical consumption.

The study of metal leachability in spent NMC 111 batteries is crucial for conceptualizing a global process, determining pretreatment, extraction, and recovery stages, analyzing chemical consumption, and assessing economic and environmental feasibility. Moreover, this analysis can contribute to the optimization of current hydrometallurgical technologies. For example, recently, mechanochemical processes that combine mechanical activation and acid leaching in one step are being developed, achieving high lithium and cobalt leaching yields of over 90%, especially for lithium-cobalt oxide and lithium-iron-phosphate batteries processing [26, 27]; however, extending this study to other types of spent batteries is necessary.

5 Conclusions

This work studied the effect of coupling mechanical activation and leaching processes for extracting valuable metals from a black mass (NMC 111) sample. Furthermore,

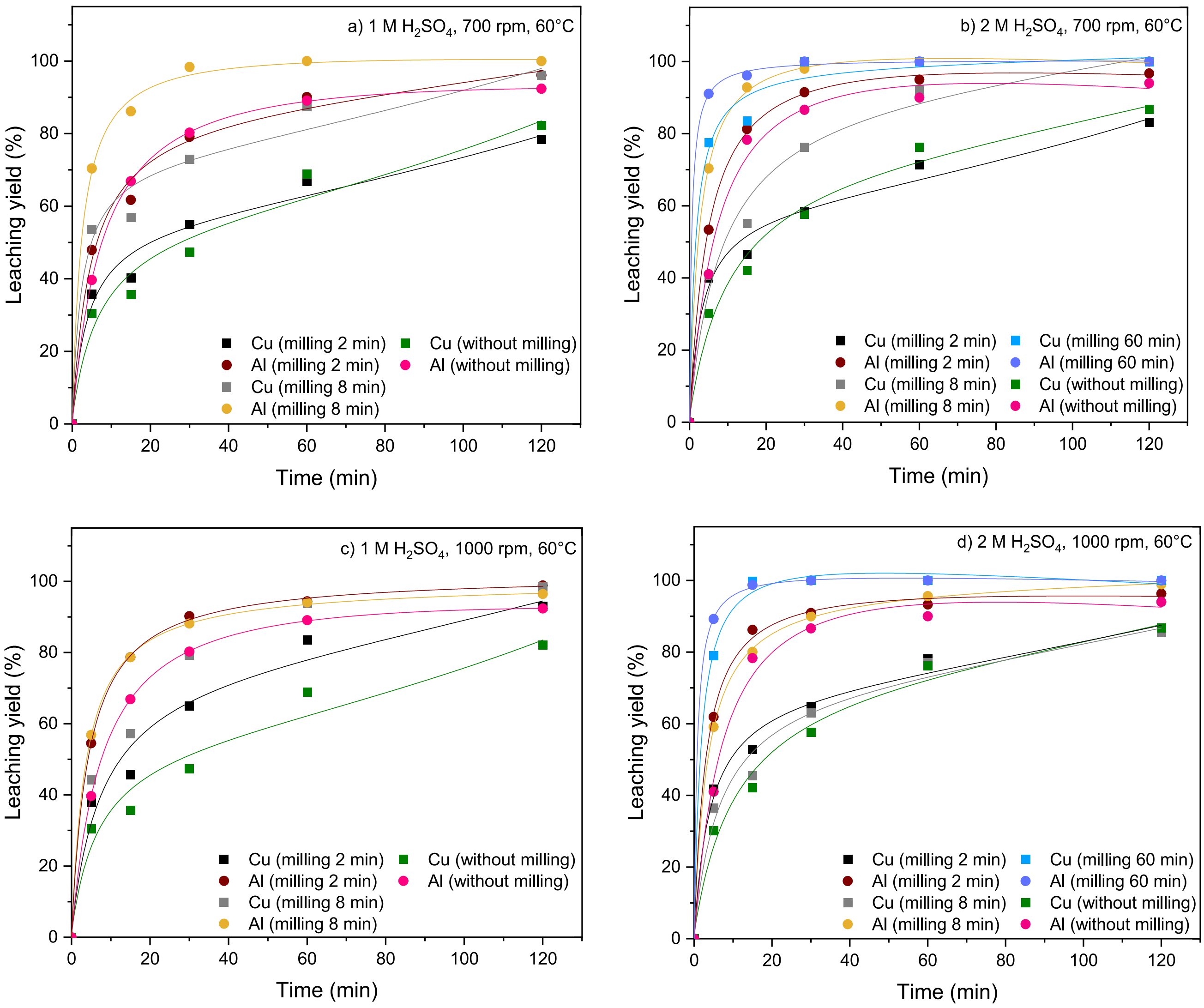

Fig. 9 Leaching efficiency of copper and aluminum: 700 rpm with 1 M H_2SO_4 (**a**) and 2 M H_2SO_4 (**b**); 1000 rpm with 1 M H_2SO_4 (**c**) and 2 M H_2SO_4 (**d**), all cases at 60 °C

the effect of the combined system on the extraction of metallic impurities was analyzed. The experimental conditions, 1000 rpm and 60 min of milling time, 2 M H_2SO_4, 60 ºC, and 120 min, allowed almost complete lithium dissolution (98%) and $\geq$ 80% of cobalt, manganese, and nickel (81% Co, 80% Mn, and 84% Ni). Although copper and aluminum traces were also extracted (100% Al and 100% Cu), they did not affect the valuable metal leaching.

The proposed methodology demonstrated that mechanical activation and temperature improved the extraction of lithium, cobalt, nickel, and manganese from NMC 111 black mass, enhancing leaching kinetics. Therefore, the proposed methodology serves as a basis for understanding metal dissolution behavior and highlights the significance of coupled treatments in recycling processes. Finally, this methodology can be applied to the treatment of other types of batteries and evaluate the effect of metallic impurities on the efficient extraction of valuable and critical metals.

Appendix A

Table 2 Elemental composition of the black mass sample (≤500 µm). Mechanical activation conditions: 700 rpm, 1000 rpm, and 60 min

Element	Co	Ni	Mn	Li	Cu	Al	Fe
(wt%)							
60 min, 700 rpm	12.10 ± 0.49	8.63 ± 0.39	8.10 ± 0.34	3.68 ± 0.06	2.72 ± 0.11	0.80 ± 0.003	0.28 ± 0.01
60 min, 1000 rpm	12.34 ± 0.15	8.67 ± 0.12	8.00 ± 0.10	3.60 ± 0.07	3.00 ± 0.04	0.70 ± 0.002	0.20 ± 0.01

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Declarations

Competing interests The authors have no relevant financial or non-financial interests to disclose.

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