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Industrial Recycling of Electric Vehicle Batteries

Industrielles Recycling von Elektro-Fahrzeug Batterien

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Abstract

For the introduction of Battery Electric Vehicles in high volume, it is mandatory to reduce the carbon footprint on battery production, which makes recycling as a strategic path. Due to large aluminium structures and mining activities to allow critical materials for high efficiency batteries, the carbon footprint on battery electric vehicles can be two to three times higher than combustion vehicles. In addition to that, the larger use of batteries in many applications is demanding higher volumes of key materials, which cannot be supplied as fast as demanded by new applications.

The use of recycling process on the end life batteries become strategic to improve critical material sourcing as well improving the carbon footprint for battery manufacturing. The most common recycling process is pyrometallurgy, employing high temperature, high energy consumption and higher CO_2 emissions. Even though, it allows a CO2e emission reduction of 34% lower than new batteries materials. Alternative recycling process is hydrometallurgy, which has stood out as a promising process with high efficiency, use of low temperatures, lower emissions, and lower energy consumption.

The process discussed in detail in this work is an innovative hydrometallurgy process where flexibility to cover different battery chemicals and geometries in the same batch was developed. This new feature compared to state of the art allows higher recycling efficiency keeping low energy consumption and high recovery rates.

Results presented come from different types of cells batteries (cylindrical, prismatic and pouch) and different chemicals (NCA – $LiNiCoAlO_2$; NMC – $LiNiMnCoO_2$; LCO – $LiCoO_2$). Physical separation and leaching recovered more than 90% of the minerals of interest with a reduction in energy consumption up to 65% compared to pyrometallurgical techniques. Demonstration plant is being built to 2024, in order to conclude development process up to TRL6-8 to allow modular plants for a global implementation.

Kurzfassung

Für die Einführung von batterieelektrischen Fahrzeugen in großen Stückzahlen ist es zwingend erforderlich, den CO_2-Fußabdruck bei der Batterieproduktion zu reduzieren, was das Recycling zu einem strategisch erforderlichen Weg macht. Aufgrund der großen Aluminiumstrukturen und der Bergbauaktivitäten, die kritische Materialien für hocheffiziente Batterien ermöglichen, kann der CO_2-Fußabdruck von batteriebetriebenen Elektrofahrzeugen zwei- bis dreimal höher sein als bei Fahrzeugen mit Verbrennungsmotor. Darüber hinaus erfordert der zunehmende Einsatz von Batterien in vielen Anwendungen höhere Mengen an Schlüsselmaterialien, die nicht so schnell geliefert werden können, wie es neue Anwendungen erfordern.

Der Einsatz von Recyclingprozessen für die End-Life-Batterien wird strategisch, um die Beschaffung kritischer Materialien zu verbessern und die CO_2-Bilanz für die Batterieherstellung zu verbessern. Das gebräuchlichste Recyclingverfahren ist die Pyrometallurgie, bei der wegen der hohen notwendigen Temperaturen, ein relativ hoher Energieverbrauch und hohe CO_2-Emissionen verursacht werden. Trotzdem führt es zu einem um 34 % niedrigeren CO_2-Emissionen im Vergleich zur Herstellung neuer Batterien. Ein alternatives Recyclingverfahren ist die Hydrometallurgie, das sich als vielversprechendes Verfahren mit hoher Effizienz, Verwendung niedriger Temperaturen, geringeren Emissionen und geringerem Energieverbrauch erwiesen hat.

Bei dem in dieser Arbeit ausführlich diskutierten Verfahren handelt es sich um ein innovatives hydrometallurgisches Verfahren, bei dem die Flexibilität entwickelt wurde, verschiedene Batteriechemikalien und Batteriegeometrien in derselben Charge abzudecken. Diese neue Funktion im Vergleich zu heute gebräuchlichen Verfahren ermöglicht eine höhere Recyclingeffizienz bei niedrigem Energieverbrauch und hohen Verwertungsraten.

Die vorgestellten Ergebnisse stammen von verschiedenen Arten von Zellbatterien (zylindrisch, prismatisch und beutelförmig) und verschiedenen Chemikalien (NCA – $LiNiCoAlO_2$; NMC – $LiNiMnCoO_2$; LCO – $LiCoO_2$). Durch die physikalische Trennung und Laugung wurden mehr als 90 % der interessierenden Mineralien gewonnen, wobei der Energieverbrauch im Vergleich zu pyrometallurgischen Techniken um bis zu 65 % gesenkt werden konnte. Die Demonstrationsanlage wird bis 2024 gebaut, um den Entwicklungsprozess bis TRL6-8 abzuschließen und um modulare Anlagen für eine globale Implementierung zu ermöglichen.

Introduction

The challenge of decarbonization on transportation sector is largely debated. The electrification is considered one important path to achieve low carbon footprint on transportation [1, 2, 3]. However, it depends on the energy matrix and specially the long chain of battery production. Considering a C type vehicle, literature [4, 5] indicates that the production footprint using a battery electric design can be two to three times bigger than the combustion version depending on the range, which is linked to size, of the batteries. Considering the same autonomy for both vehicles, the battery to the electric version would demand 26 thousand Liters of water to enable the extraction of necessary lithium. The mineral extraction process for one battery can reach 225 tons of earth movement. Moreover, large quantity of aluminium is necessary to provide structural safety to the batteries, which is also intensive on energy consumption. The use of recycling can support the target to reduce carbon footprint for the battery production. Compared to the mining of critical materials, pyrometallurgy recycling can reduce 34% of CO2e emissions and hydrometallurgy 70%. Estimations following hydrometallurgy recycling process indicate 28% reduced carbon footprint to the total battery, considering the current scenario of energy matrix in USA [6], see figure 1 out of [6].

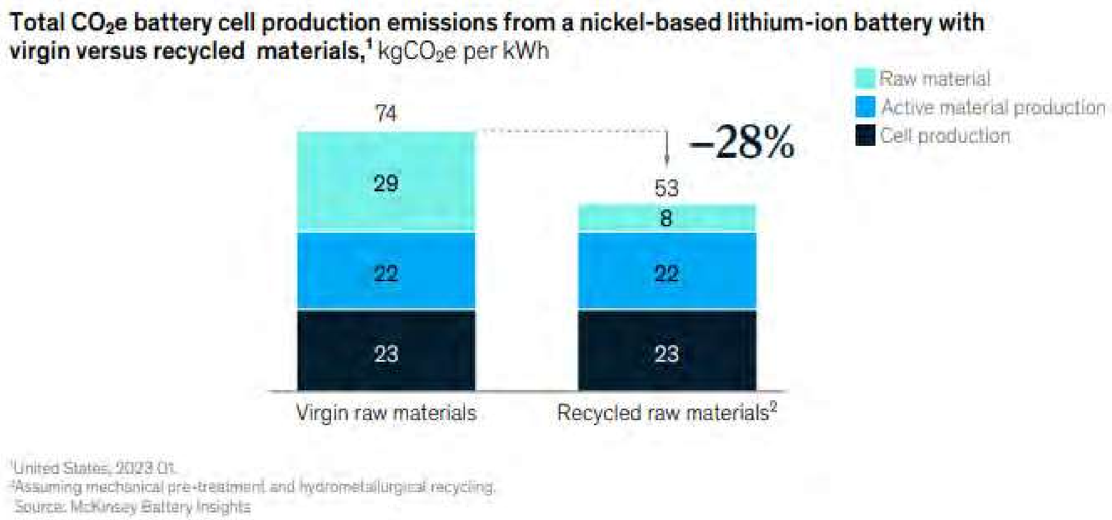

Fig.1. Vehicle production footprint and saving potential due to recycling [6].

Another important subject for batteries is the supply chain and the challenge to increase the production of critical minerals in the necessary speed for the expected vehicle electrification. The demands of new mines indicated in Figure 2 is a result from the study from *Benchmark Mineral Intelligence 2022*. However, it is important to consider that time-to-market is much longer for the mining sector than it is for the automotive sector. There are mine implementation plans that can take over 15 to 20 years to start production. Such condition will create a strong pressure on the mineral prices. Battery recycling can be strategic to balance the growing mineral demand for electric vehicles [7].

Fig. 2. Demand for new mines to attend critical minerals for EV production in the future (adapted from *Benchmark Mineral Intelligence 2022)*.

The geopolitical aspect involved on the battery manufacturing value chain is also a subject of concern due to the crisis of semi-conductors during pandemia time. The high concentration of volumes in very small number of countries are critical to the future decarbonization targets set by the countries. Figure 3 from IEA [8] illustrates the battery value chain concentration. Recycling decentralizes such supply chain when providing critical minerals on the correct purity level necessary for battery manufacturing.

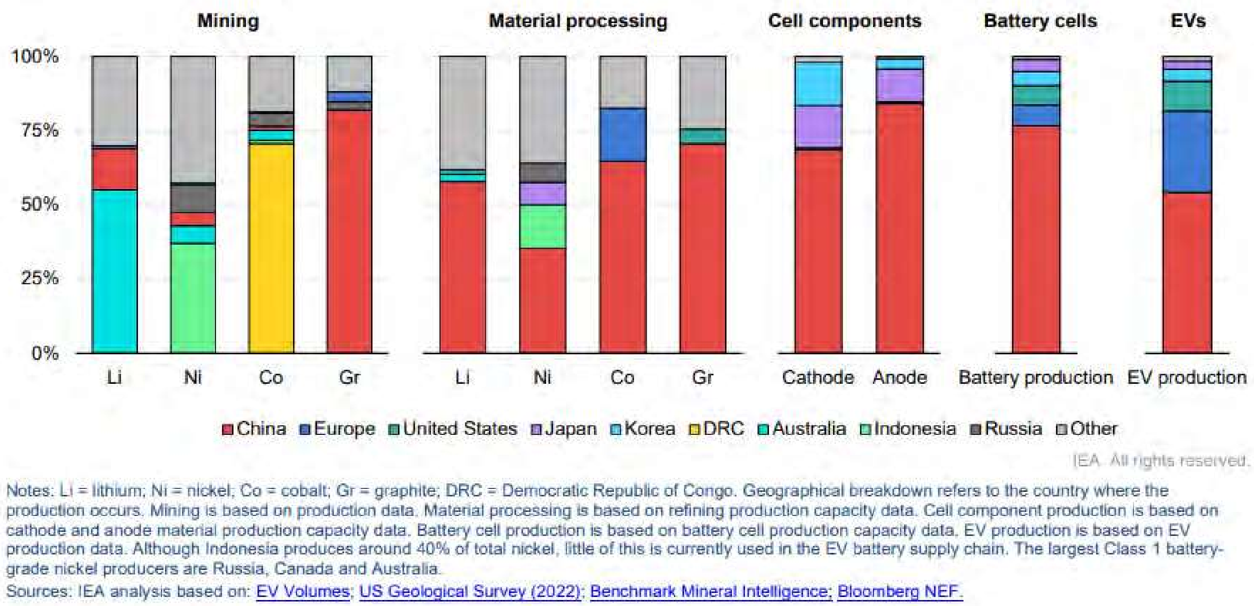

Fig. 3. Geographical distribution of the global EV battery supply chain [8]

Battery Recycling Technologies

The most traditional process for lithium-ion battery recycling is pyrometallurgy. In this process the battery is burnt in an oven up to 1,500 – 1,600 °C. Then by melting point, the materials of interest are separated. This process is energy intensive, generates toxic gases and has limited materials recovery. Recovery rates are between 60 to 80%, depending on the metal, with low levels of purity. Once Lithium is very light, it is lost in the slag and additional processes are necessary for actual Lithium recovery. In the same way, purity is very limited. However, this process presents good flexibility, once there is no big concern of battery disassembling levels, and low cost levels depending mainly on energy supply. Due to these characteristics, new battery recycling plants being announced in different regions are not using pyrometallurgical process.

The other process method applied to battery recycling is hydrometallurgy. Here it is necessary to disassemble the battery up to the cell level, which is ground forming a mixture usually called black mass. Such black mass is lixiviated on acid solution, which later in different steps are subjected to reagents to precipitate the material of interest. This process can present high recovery rates above 90% and helps to achieve high purity levels. Once the process happens on low temperature (120 – 150 °C), the levels of energy consumption and greenhouse gases are much lower than traditional method. The energy consumption on hydrometallurgy is concentrated on black mass processing, which is 8 to 9 times higher than energy consumed on hydrometallurgy chemical leaching and precipitation. Study [9] indicates that hydrometallurgy can reduce 70% of energy consumption compared to mining, while pyrometallurgy 34%. Once hydrometallurgy involves chemical consumables, it is expected to present higher cost. The disassemble necessity up to cell level combined to the chemical consumables can increase the cost of this alternative compared to traditional method. Even though, this process is the basis of all new recycling plants being announced in the last years worldwide.

The process explored in this paper is based on hydrometallurgy with innovative approach to enhance its flexibility, enabling the process in the same batch for different chemicals. Figure 4 illustrates Tupy-LAREX process.

Innovative Hydrometallurgy Process

The presented innovative hydrometallurgical process for the recycling of Lithium-ion batteries was developed in cooperation with the largest Latin America University, University of Sao Paulo, through the research group LAREX linked to the Chemical Engineering Department [10, 11]. For that reason, the process will be named in this paper Tupy-LAREX.

Being a hydrometallurgical process, the first concern was to cope with the limited flexibility. It was worked out enabling a process that can mixture different battery chemicals in the same process batch in high efficiency. It allows higher productivity once there is no need to limit process flow by battery availability. In the extreme level, batteries from EVs are possible to be processed with batteries from electronics. Certainly, the degree of freedom may vary depending on the value added materials that are integrated into the process.

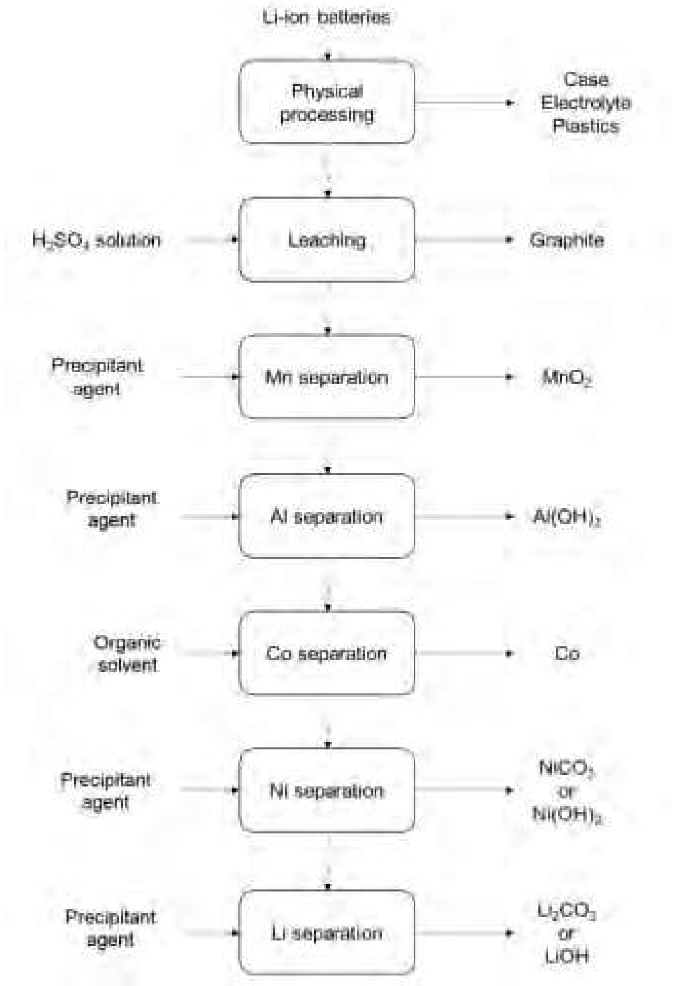

Fig. 4. Tupy-LAREX process illustration for Lithium-ion battery recycling [10].

The process can be carried out using different types of Lithium-ion batteries: cylindrical, pouch and prismatic. The batteries are discharged using electrical resistance in a controlled process. Evaluations indicate that discharge methods can influence the recycling process. After the voltage achieved zero (0V), the packs and modules are disassembled manually to release the cells. Further, the batteries were comminuted, and the cathode was separated by physical treatment.

The active material was leached using H_2SO_4 varying the parameters to achieve efficiencies above 95% for the most valuable metals. Then, the leach liquor is treated by separation and purification steps to obtain high purity products. It is used a precipitant agent for Mn recovery, precipitation for Al removal, solvent extraction for Co separation and further precipitation, and Ni and Li precipitation.

The experiments were carried out in laboratory scale. The experimental conditions were set considering the purity 90% and above for the interested products. The recycling process was designed to be flexible, simultaneously processing all types of Li-ion batteries. Separation and purification steps were carried out in glass reactor under temperature and pH control.

On modules, structures represent similar weight proportion and are composed mainly by aluminum structure. To characterize the cells obtained, after discharge, disassembly was performed separating cathode, anode, polymeric separator, case and electrolyte, see Table 1. Notice that distribution if very similar, especially on Cathode and Anode, that are more than 2/3 of cell weight.

Tab. 1. Cell composition evaluated [11].

Elements	Cylindrical	Pouch	Prismatic
Cathode	39%	32%	40%
Anode	31%	38%	32%
Electrolyte	5%	13%	9%
Case	17%	7%	14%
Polymeric Separator	8%	10%	5%

While anode is composed by graphite material with low lithium concentrations, cathode contains valuable metals, as Ni, Co, Li, Mn depending on the chemistry of active material. The chemicals evaluated were NCA – $LiNiCoAlO_2$; NMC – $LiNiMnCoO_2$; LCO – $LiCoO_2$; LFP – $LiFePO_4$. Separators are polymeric made and avoid short-circuit between cathode and anode. The case from pouch cells was lighter than others due to its unique structure mounted with aluminum foils. Electrolytes are a mix of organic compounds and Li salts, which was removed by evaporation at room temperature after disassembling.

Battery cells are firstly comminuted in knife mill or shredder, and components are separated by physical methods. The active material contained in cathode was leached reaching up to 99% of Li, Ni, Co, Mn, while 60% of Al was leached. Inert graphite and unreacted aluminum were retained in filter after solid/liquid separation. Pregnant leaching solution (PLS) was treated to separate and recover metals as oxides, carbonates and hydroxides by precipitation of Mn, Al, Li and Ni, while Co was separated by solvent extraction. Purity of products obtained was superior to 90% and recoveries of proposed process is at least 90% of cathode active material. The first metal removed from leach solution was Mn, in acid pH conditions and using a precipitant agent, it is possible to remove the Mn with precipitation efficiency of 99% with a purity of 96% (Co was in part co-precipitated). Al was removed from PLS by pH adjustment adding sodium carbonate or hydroxide, reaching efficiency of 100%, while Li, Ni and Co losses by co-precipitation were lower than 2%. Subsequently solvent extraction process separated Co from PLS, while Ni and Li remained in solution. Co was stripped from organic phase by an acid solution and then possibly recovered by oxalate and carbonate precipitation, or in metallic form by electrowinning. Li and Ni remaining in solvent extraction aqueous solution were precipitated as carbonate or hydroxide. Differently of pyrometallurgical process that reduce Co, Ni, Cu and Fe in alloys separating them from Li, Al and Mn in slag phase, hydrometallurgical process can recover metals individually.

In terms of energy consumption, literature [9 and 12] indicates average numbers for pyrometallurgy as 2.3 kWh/kg_cell and for hydrometallurgy 1.0 kWh/kg_cell. The Tupy-LAREX process presented in current TRL 5 stage values between 1.0 and 1.5 kWh/kg_cell. In this development step the process is handled on batch for each precipitation. In the development final steps, where process is starting now the continuous flow may present results expected to be between 0.8 and 1.1 kWh/kg_cell. It is today 46% lower than series pyrometallurgy and has potential to be 57% lower.

Figure 5 presents the conversion of energy consumption in equivalent CO_2 emission on the different processes, including comparison of nations with different power generation matrix (renewable and non renewable levels).

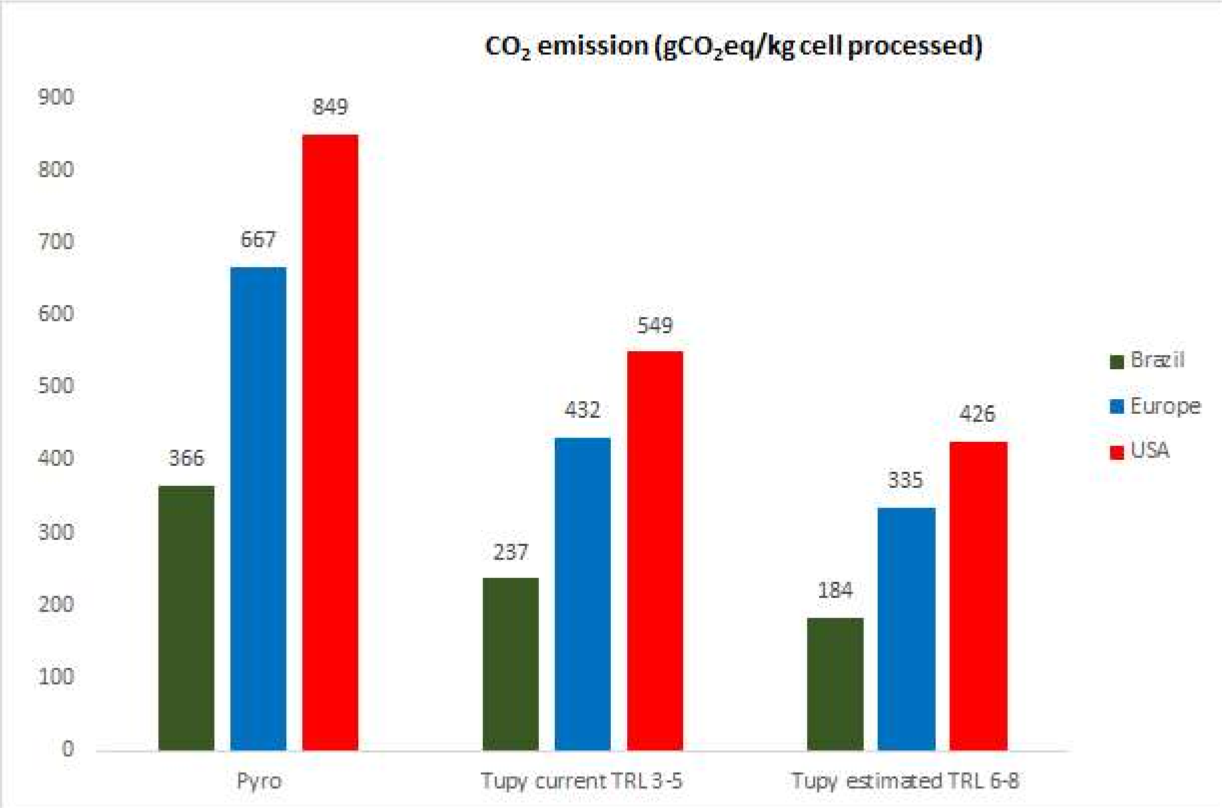

Fig. 5. CO_2 emission by recycling process and nation power grid [10].

Besides emission reduction, a comparison of material recovery from pyrometallurgical and hydrometallurgical processing of a NMC cell also demonstrates the impact of a hydro process for a circular economy. From Figure 5 for Brazilian power mix, pyrometallurgy presents almost 2 times higher CO_2 emission than hydrometallurgy for the same volume of processed cells. However, taking in consideration hydrometallurgy recovery is 40 to 50% higher than pyrometallurgy, the CO_2 emission for the same level of recovered materials presents CO_2 emissions more than 3 times higher for pyrometallurgy. Such analysis does not count the necessary extra processes needed for pyrometallurgy due to low purity levels at recovery materials.

Scale up activities

The new process development has been tested in different scales, first on pure laboratory bench level using 2L reactors (Lab Scale = TRL 3-4). Now the most recent results were obtained with 40L reactors (XL Scale = TRL 5) confirming the results. For the next step, a demonstration plant (TRL 6-8) is being built to allow process in larger scale to be processed on continuous approach. So far tests were carried on batches. Figures 6 and 7 illustrate the achieved main results in different scales as well the expected minimum targets on demonstration plant.

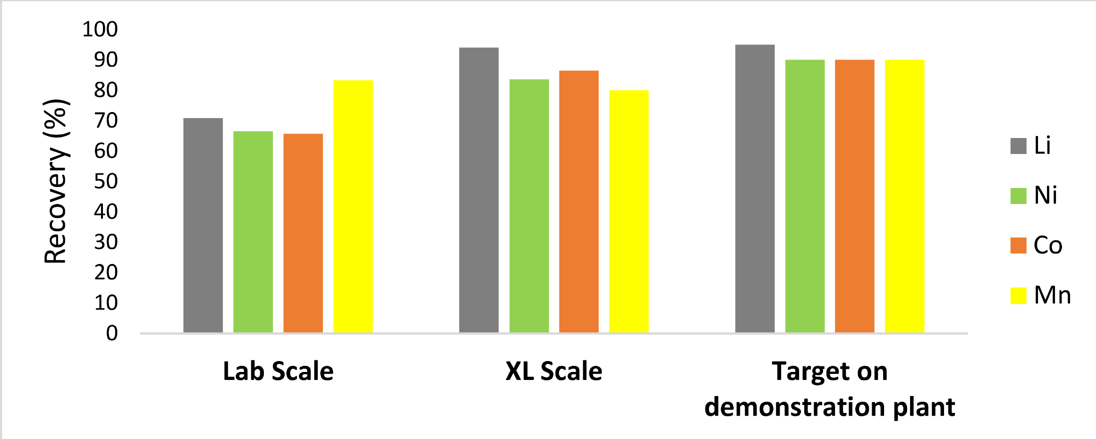

Fig. 6. Recovery rate on different scales for Tupy-LAREX process.

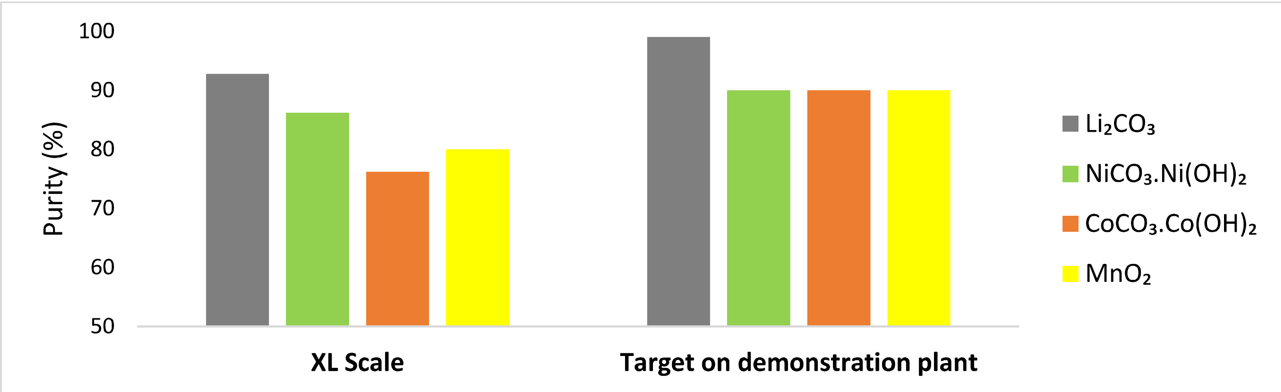

Fig. 7. Purity level for different scales for Tupy-LAREX process.

Figure 8 illustrates the demonstration plant being built to conclude process development before commercial applications. This plant is being installed in Sao Paulo, Brazil, inside IPT (Institute for Technological Research), that is experienced to support companies to scale up manufacturing processes. Demonstration campaigns are being organized with customers to validate process data on actual commercial batteries to deploy business models depending on the customer specifics.

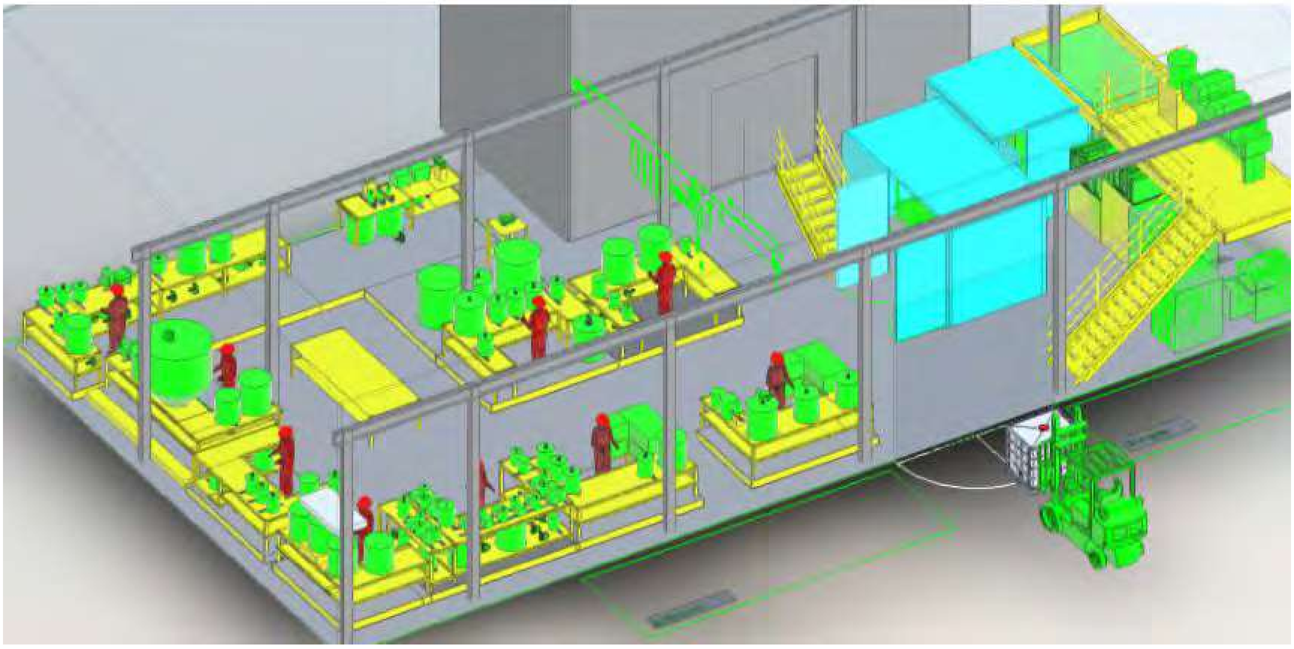

Fig. 8. Demonstration plant layout being installed in Sao Paulo, Brazil.

Conclusions

The Tupy-LAREX process is a recycling hydrometallurgical route with flexibility to treat all Lithium-ion battery configurations (cylindrical, pouch and prismatic), which has lower energy consumption if compared to pyrometallurgical processing, and lower GHG emission. It enables the recovery of battery metals (Ni, Co, Li, Mn, Al) with high purity (>90%) and high recovery efficiencies (>90%).

In addition, it may help the mitigation of environmental issues related to the manufacturing and life cycle of batteries. Evaluations of energy demand show reductions of 46% in current TRL level for hydrometallurgy compared to pyrometallurgy. It can be even improved up to 57% in the next TRL increase loop.

The GHG emission can be 3 times lower for Tupy-Larex compared to traditional pyrometallurgy when considering in addition to energy consumption the purity level obtained in each process. Such evaluation always considering the same country energy matrix.

The scale up activities up to now consolidated very good results on recovery rates and purity levels. The demonstration plant level to go further TRL 6-8 is now being built in Sao Paulo, Brazil, to enable campaigns with actual applications consolidating the innovative process through continuous flow compared to batch process done at lab scales up to now. It will also enable validation of commercial conditions to set specific plants depending on customer needs.

References

[1] Papadis E, Tsatsaronis G. Challenges in the decarbonization of the energy sector. Energy 2020; 205:118025.

[2] Martins LS, Guimarães LF, Botelho Junior AB, Tenório JAS, Espinosa DCR. Electric car battery: An overview on global demand, recycling and future approaches towards sustainability. J Environ Manage 2021; 295:113091.

[3] Harper G, Sommerville R, Kendrick E, Driscoll L, Slater P, Stolkin R, et al. Recycling lithium-ionVDI-Analyse der CO2äq-Emissionen von Pkw batteries from electric vehicles. Nature 2019;575:75–86.

[4] Girardi P, Gargiulo A, Brambilla PC. A comparative LCA of an electric vehicle and an internal combustion engine vehicle using the appropriate power mix: the Italian case study. Int J Life Cycle Assess 2015; 20:1127–42.

[5] Guilherme, R., Garbe, T., Cifoni, F., Kersten, T.; *Biofuels as a strategy for CO2e-Reduction in Brazil*; 10th International Engine Congress, Baden Baden, February 2022.

[6] Breiter, A., Schuldt, T., Linder, M., Siccardo, G. and Vekic, N.; *Battery recycling takes the driver's seat*; McKinsey's Advanced Industries Practice, March 2023.

[7] Volkening, M.; Sagner, K.; Battery recycling: The economics of a multi-billion Euro circular economy in the making; 32nd Aachen Colloquium Sustainable Mobility 2023, Aachen, October 2023. Paper No. 037.

[8] *Global Supply Chains of EV Batteries*; IEA - International Energy Agency, France, July 2022.

[9] Dai, Q.; Spangenberger, J.; Ahmed, S.; Gaines, L.; Kelly, J. C.; Wang, M.; *EverBatt: A closed-loop battery recycling cost and environmental impacts model*. United States, 2019.

[10] Gobo, L. A., Ferrarese, A., Kumoto, E.A., Obara, R., Botelho Junior, A. B., Tenório, J. A. S., Espinosa, D. C. R.; *Analysis of energy demand for recycling electric vehicle batteries through a flexible hydrometallurgical process*. SIMEA 2023, Sao Paulo, Agosto, 2023.

[11] Ferrarese, A., Kumoto, E.A., Gobo, L. A., Botelho Junior, A. B., Tenório, J. A. S., Espinosa, D. C. R.; *Flexible hydrometallurgy process for electric vehicle battery recycling*. SAE Brasil 2022 - 2022-36-0072, Sao Paulo, November 2022.

[12] Xu, P.; Yang, Z.; Yu, X.; Holoubek, J.; Gao, H.; Li, M.; Cai, G.; Bloom, I.; Liu, H.; Chen, Y.; An, K.; Pupek, K. Z.; Liu, P.; Chen, Z; *ACS Sustainable Chemistry & Engineering*; 2021, 9 (12), 4543-4553.