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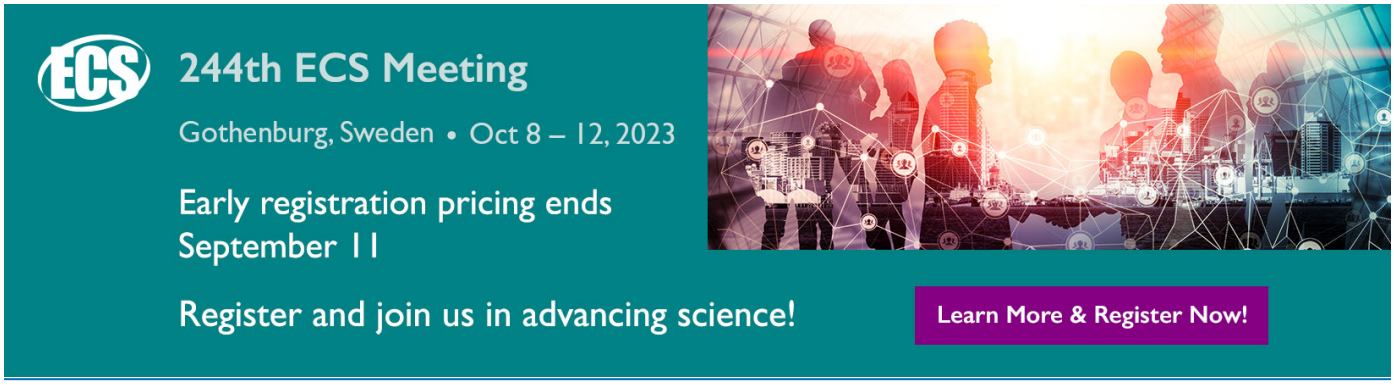
Direct Recycling of Lithium-Ion Cathode: A Green Solution (Applied To Laptop Batteries)

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Direct Recycling of Lithium-Ion Cathode: A Green Solution (Applied To Laptop Batteries)

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A green and automatic recycling separation process to recycle $LiCoO_2$ cathodes from laptop batteries was designed. Prior to that, an exhaustive study of the main chemistry used for laptop batteries was undertaken, analyzing the chemical composition of the cathodes of 160 spent batteries from laptops. After concluding that around 70% of the spent batteries analyzed presented $LiCoO_2$ as the cathode active material, regeneration experiments were undertaken to recover the $LiCoO_2$ to be able to be used in the manufacture of new laptop batteries. The influence of the cathode separation process was addressed, studying the difference between using a separation method that involves the use of thermal processes and using a separation method that implies the use of an organic solvent. After that, different regeneration methods were compared in order to reach the optimum process to obtain recovered cathodes which chemical, and microscopic structures that lead to the best electrochemical performance.
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The demand of lithium-ion batteries (LIBs) is continuously increasing due to their application in consumer electronics, electric vehicles (EVs), and grid storage, and it has been projected that global LIB market will reach $92.2 billion by 2024. Two huge challenges are posed in this scenario: the first one is the shortage of raw materials (Li, Co, Ni, etc.) and the increase of their cost and the second one is the environmental problem that the enormous amounts of spent batteries generate as some components are toxic and carcinogenic in nature.[1,2] Proper disposal of the accumulating spent LIBs would be not only friendly to the environment and human heath but also helpful for the sustainable supply of strategic resources.[3] Great efforts have been made to recycle cathode materials of LIBs because they account for almost 50% cost in the whole battery.[4] However, owing to the confusion of battery lifecycle management and the absence of consolidated production criteria, the recycling market of waste LIBs is not yet mature.[5]

The use of LIBs is widely spread, from smartwatches to electric buses by way of laptops. Depending on the type of electronic device, the battery must present different performance attributes. For example, smartwatches need a stored energy capacity between 0.1 to 0.5 Wh, mobile phones between 2 and 4 Wh, laptops between 30 and 100 Wh, electric cars between 5–10 kWh, and electric buses need a battery with a stored energy capacity of a few 100 kWh.[6] These different characteristics make batteries have different weights and thus, different amount of active cathode material.

Batteries are complex systems, with the active cathode material adhered to an aluminum foil which is together with the separator and the anode, all embedded in an electrolyte and wrapped with a plastic material. Related to the active cathode materials, different chemistries can be found, each of them with different properties: $LiCoO_2$ (LCO), with high energy density and high performance but poor safety and low power density and cycle lifespan; $LiFePO_4$ (LFP), with excellent safety and acceptable performance, power density and cycle lifespan, but poor energy density; $LiMn_2O_4$ (LMO), with good safety, power density and cost, but low energy density, cycle lifespan and performance; $Li(Ni,Co,Al)O_2$ (NCA), with excellent energy density and good power density, cycle lifespan and performance, with poor safety and high cost; and $LiNi_xMn_yCo_zO_2$ (NMC), with excellent energy density and good performance. About NMC cathodes, there are different chemistries, from NMC 111 ($LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$) to the most recent NMC 811 ($LiNi_{0.8}Mn_{0.1}Co_{01}O_2$) in an attempt to reach no-cobalt chemistries (due to its high cost and safety problems).[7] Figure 1 shows a schematic illustration of the battery assembly, from the material level to the battery pack level in order to show the complexity of a battery.

The complexity of batteries makes the separation of the active cathode material with minimum contamination of the rest components of the battery challenging.[8–10] In a previous study,[11] undertaken by this research team, the separation of the active cathode material from mobile phone batteries to be used as secondary raw material in the synthesis of ceramic pigments was addressed. Batteries from mobile phones have 9 g of active cathode material approximately while those from laptops present an average weight of 70 g, reason why the type of batteries was changed for the following studies.

With the commercial success of Li-ion batteries, it is natural that their mechanisms have been widely investigated. Processes leading to the capacity fade include electrolyte decomposition, Li deposition on the negative electrode, active material dissolution, structural changes in the active material, separator pore closure and increase in the cell stack pressure. While all the above-mentioned mechanisms do affect the cell capacity, the electrolyte decomposition through the formation of a solid electrolyte interphase (SEI) on the negative electrode can be considered as the most dominant. The SEI formation consumes lithium, which lead to the loss of lithium on the positive electrode.[12] During the charging and discharging of Li-ion batteries, the lithium ions continuously intercalate and de-intercalate between the cathode and the anode. This phenomenon generates an alteration in the layered structure due to multiple shrinkage and expansion.[13] The formation of the SEI prevents lithium ion from moving between the anode and the cathode, generating the cell failure.

There are three recycling technologies for end-of-life Li-ion batteries: pyrometallurgical, hydrometallurgical, and direct recycling. Most of the recovery processes of metals such as Co or Ni present in a lithium-ion battery are based on pyrometallurgical or hydrometallurgical methods that generate a huge impact on the environment.[14–23] With this recycling technologies, the embedded energy in the desired cathode particles is lost during such a destructive process.[24] For these reasons, direct recycling technology emerged, which retains the structure and morphology of the original cathode material. Xu et al. performed a life cycle analysis of the direct recycling and compared it with pyrometallurgical and hydrometallurgical techniques. The results showed that hydro and pyrometallurgical recycling methods required 30.6 and 18.4 MJ kg^{-1} of cells recycled, respectively, while direct recycling used only 3.5 MJ kg^{-1}. Besides green house gas emissions per kg of cells released during the recycling were 28% less in the case of direct recycling compared to the other two methods.[6,25]

Direct recycling is based on the relithiation of the material with the aim of restoring the Li composition and recovering the lithium

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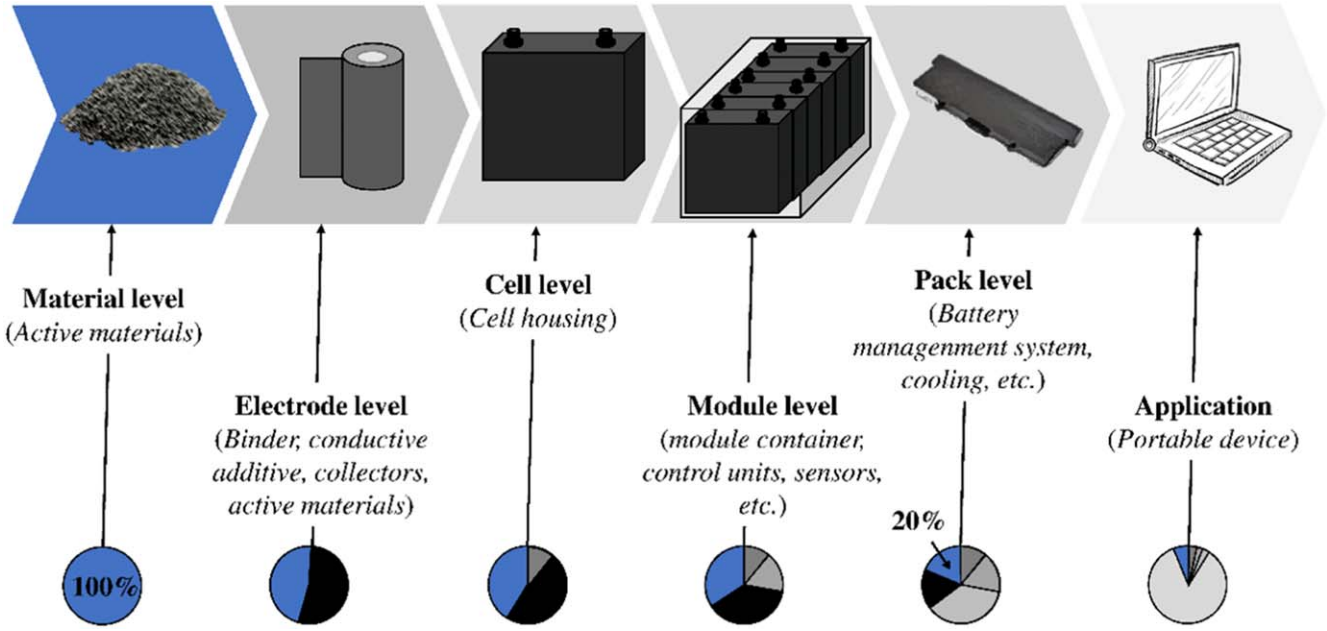


Figure 1. Schematic illustration of a battery assembly.

stoichiometry, as well as the original structure. Different studies for cathode regeneration can be found, in which different conditions for regeneration are described, although no one show a relithiation process that can be used at industrial scale, and no one has developed their processes for end-of-life laptop batteries.[26–36]

The present work is focused on the cathode regeneration from laptop lithium-ion batteries to be used in new ones. Around 165 different spent laptop batteries were chemically analyzed and those that presented LCO as active cathode material were selected for this study for being the main chemistry found in the disposed batteries analyzed. The influence of the cathode separation process over the performance of regenerated cathodes, together with the regeneration process itself, was studied. Degraded cathodes as well as regenerated were physicochemically characterized by wavelength-dispersive X-ray spectrometry (WD-XRF), inductively coupled plasma optical emission spectrometry (ICP-OES), combustion and infrared detection, potentiometry, X-ray diffraction (XRD), and scanning electron microscopy with energy dispersive X-ray microanalysis (SEM-EDX); followed by a series of electrochemical tests to check the effectiveness of the regeneration process applied.

The proposed method is a scalable and ecofriendly close-loop strategy for recycling spent cathodes of LIBs, showing a good application prospect.

Experimental Part

Materials.—Around 160 spent LIBs used in laptops from different manufacturers and with different sizes were chosen for this study. The regeneration tests were carried out over spent and unused $LiCoO_2$ (LCO) LIBs, as well as pristine LCO purchased from Xiamen AOT Electronics Technology Co., LTD., that has been used as standard in the electrochemical tests. Unused batteries were purchased from Dell Computer, S.A.

Discharging pretreatment step.—Before any manipulation, spent LIBs must be discharged as they always retain some residual energy which may cause safety issues such as short-circuiting and self-ignition. One of the commonest methods of discharging is to immerse batteries in a salt solution, so spent LIBs were discharged in 5%(wt) sodium chloride solution for 48 h and air-dried afterwards to make the dismantling process safe.[21,37]

Study of the actual chemistries of the disposed laptop lithium-ion batteries.—Before carrying out the regeneration experiments, the study of the cathode chemistries that are being used in current batteries is needed. As battery composition is not informed by the suppliers, the spent ones were first manually dismantled to perfectly know the composition of the active cathode materials. Lithium was analyzed by ICP-OES using an Agilent model 5100 SVDV spectrometer and the rest of the major components was determined by WD-XRF using a PANalytical model AXIOS spectrometer, preparing the sample as fused beads.

Cathode separation process.—The separation method can significantly affect the electrochemical performances of the regenerated cathodes. In this study, different separation processes of the cathode were studied prior to regeneration.

Mechanical and thermal separation process.—Active cathode material was separated using a semiautomatic process design by this work group which is under patent, that involved a first cutting step of the shell by means of blades, followed by several calcination processes at different temperatures and stirring processes in an aqueous medium to obtain the material with little contamination from the foils, anode material, separator and electrolyte.[11] A prior study stated that the application of heat to the cathode electrode (formed by the aluminum foil, PVDF, carbon black, and active cathode material powder) to remove the binder (PVDF) and let the cathode powder release from the foil led to the decomposition of the cathode structure when its chemistry was NMC 111.[38] In order to study the influence of the separation process in the final product obtained, the same brand and model of the spent battery used in this study was bought and it was also submitted to the separation process.

Organic solvent separation method.—The second separation method applied implied the use of an organic solvent. Aluminum foil with the active cathode material adhered to it was separated from the rest of the battery components manually. Then, the foil was scrapped and added into a baker with 150 ml of N,N-Dimethylformamide (DMF), heating the system for 2 h at a temperature of 70 °C while stirring. After that, the powder released was collected and dried at a temperature of 100 °C.[38]

Regeneration process.—Two different relithiation processes were studied: direct solid-state sintering treatment and hydrothermal treatment followed by a short thermal annealing step.

Solid-state sintering.—For solid-state sintering, experiments were undertaken based on the regeneration process stablished by Shi et al.[23] The degraded cathode material powder was carefully mixed with Li_2CO_3 and sintered at different temperatures. The optimum regeneration conditions (amount of Li_2CO_3 and sintering temperature) were those that resulted in a regenerated cathode that showed a sharp and intense diffraction peak.

Hydrothermal treatment and thermal annealing step.—Hydrothermal regeneration process was undertaken following the procedure stablished by Shi et al.,[23] mixing the degraded cathode with a LiOH solution in an autoclave. After the hydrothermal process took place, the powder was submitted to an annealing process at 850 °C for 4 h in oxygen atmosphere, adding 5% Li_2CO_3 in excess.

Spent and regenerated active cathode material characterization.—A detailed chemical, phase, and microscopic understanding of the phase transformation characteristics in these cathode structures will lead to tailoring of the material structure for better electrochemical performance.

The chemical composition of the spent and regenerated materials was determined by WD-XRF, ICP-OES, and potentiometry for the determination of fluorine using a Crison model micropH 2002 pH-meter, fitted with a fluorine ion-selective electrode and a reference electrode. Carbon was determined by combustion followed by IR detection using a Carbon elemental analyzer model CS-200 from LECO. The crystal structure was characterized by X-ray powder diffraction (XRD) using a BRUKER Theta-Theta model D8 Advance. The morphology of the particles was studied by SEM-EDX using a FEG-ESEM Quattro S from Thermo Fisher.

Electrochemical characterization.—Battery research is tackling the complex electrochemical problems so, establishing a standardized protocol for the data is very beneficial for machine learning from the data in the open-access literature,[39] which means that the electrochemical characterization of batteries should follow the same procedure in order to be able to compare different studies.

To carry out the electrochemical tests, working electrodes were prepared using a typical slurry-coating method. The slurry was prepared by mixing the corresponding active cathode material, conductive carbon black (Super P), and polyvinyl fluoride (PVDF) in a mass ratio 90:5:5 in N-methyl-2-pyrrolidone (NMP) as solvent. It was then coated onto aluminum foil by Dr Blade and dried at 100 °C for 12 h in a vacuum oven. Working electrodes for electrochemical analysis had a diameter of 10 mm and were made up using Swagelok-type cells. Li metal foil was used as the counter and reference electrode, and an electrolyte-soaked monolayer membrane as separator. The electrolyte was a 1 M hexafluorophosphate lithium salt ($LiPF_6$) dissolved in ethylene carbonate and dimethyl carbonate. Cell assembly was carried out in an inert atmosphere glovebox. Electrochemical tests were carried out in a BSC-8010 potentiostat from BioLogic, conducting galvanostatic discharging tests at a speed of 0.2C.

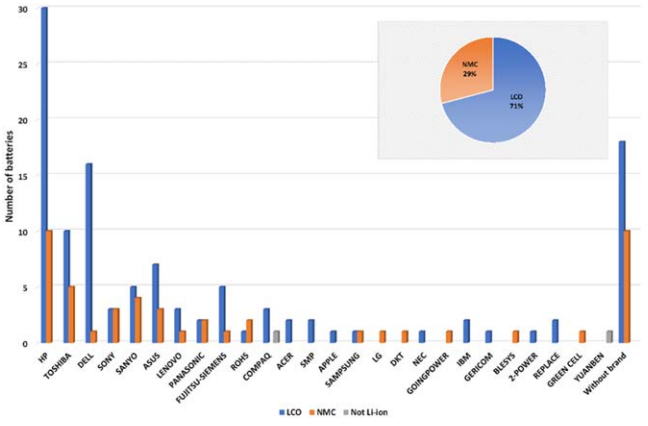


Figure 2. Chemistry of the active cathode material of the spent batteries from laptops analyzed.

Results and Discussion

Study of the currentchemistries of the disposed laptop lithium-ion batteries.—A total of 165 different spent batteries from laptops were analyzed in this study. This preliminary study had two different objectives. On the one hand, the study permitted to know the type of cathodes present in the spent batteries at this moment and, on the other hand, it allowed for stablishing an automatic battery separation process, that could be industrially implemented, according to their cathode composition that led to the recovery of cathodes of different chemistries separately and thus a specific recovery system for each chemistry, resulting in an improvement in the quality of the recovered product.

Figure 2 shows the chemistries found for each battery grouped by brands.

The results obtained showed that 71% of the spent LIBs analyzed presented LCO as active cathode material so, the regeneration study was undertaken with LCO cathodes as it is the major chemistry found in the disposed laptop LIBs at this moment. Among all the spent batteries with LCO cathodes, regeneration experiments were undertaken with a DELL battery model 60wh TYPE PW23Y because it was commercially available and so, experiments could be carried out over the spent and the unused one.

Table I shows the chemical composition of the pristine, spent, and unused cathode materials of the selected laptop battery.

From the analysis obtained, it can be seen that while the concentration of the transition metal remains almost invariable (although it is higher in the spent material due to the loss of lithium that provokes the concentration of the rest of elements present), the loss of lithium in the degraded material is quite significant.

Influence of the cathode separation process.—Different cells of the unused battery (new) were separated using the mechanical and thermal separation process (referenced as MTU-LCO) and the organic separation process (referenced as OU-LCO), and they were chemically characterized, the results being shown in Table II.

No significant differences were observed in the composition of the separated material apart from the carbon content present in the sample separated with the organic solvent, as it is not eliminated in this process.

Both separated active cathode materials were submitted to the electrochemical test. The electrochemical tests at low cycles, together with the pristine (standard), are shown in Fig. 3.

The maximum discharge capacity of the pristine is around 150 $mAh{\cdot}g^{-1}$, which is closed to the theoretical (153 $mAh{\cdot}g^{-1}$). MTU-LCO and OU-LCO showed similar behavior to that of the pristine.

Song et al.[38] studied the influence of the use of heating in the separation of NMC 111 from the aluminium foil and concluded that when the temperature applied was above 400 °C, the material was easily separated but the discharge capacity decreased considerably. They related this decrease in capacity to the decomposition of the NMC 111. However, contrary to what they stated, no differences in capacity were found when separating the LCO active material using a temperature of 400 °C compared to those obtained in the pristine and those obtained in the material separated with organic solvent.

So, as both separation processes led to same results, the preferred separation process was that developed was this research group and under patent, that implies the use of mechanical and thermal separation processes, as it is greener because no organic solvents are used.

Table I. Co and Li content of the cathode materials used for the study.

	Pristine LCO (Pristine)	Spent material (S-LCO)	Unused material (U-LCO)
Co (wt%)	60.2	63.3	59.5
Li (wt%)	7.1	3.3	6.9

Table II. Chemical composition of the cathodes separated using different processes.

	MTU-LCO	OU-LCO
Co (wt%)	59.7	58.5
Li (wt%)	6.7	6.3
Al (wt%)	0.2	0.4
P (wt%)	0.14	0.05
F (wt%)	1.0	0.9
C (wt%)	<0.1	2.0

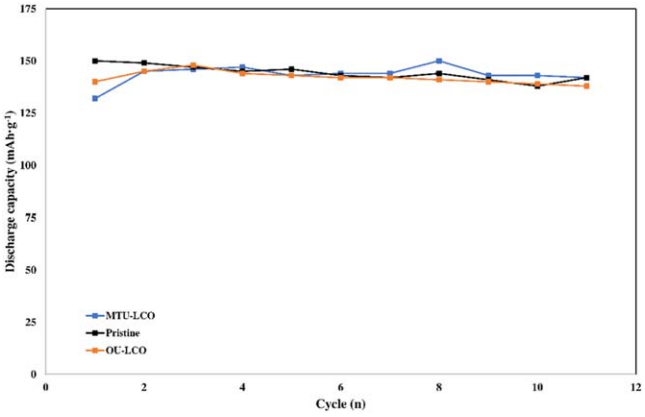


Figure 3. Cycle life of the unused cathode separated with different separation processes.

Regeneration of cathode materials.—Solid-state sintering regeneration experiments were undertaken modifying the sintering temperature and the amount of Li_2CO_3 added to the cathode for relithiation. The sintering process was carried out for 12 h with a ramping rate of 5 °C min^{-1} in air atmosphere, modifying the sintering temperature: 750 °C and 850 °C. The other variable studied was the amount of Li_2CO_3 added for the relithiation. The amount of Li_2CO_3 was calculated to make the mixture cathode:Li_2CO_3 with a Li/Co molar ratio of 1.05 and 2.05, respectively.

Figure 4 shows the XRD patterns of the cathodes regenerated at different temperatures and with different amount of Li_2CO_3.

The intensity of the main peak increased with the relithiation process and, from the results obtained, it could be seen that maximum intensity of the peak was obtained at a temperature of 850 °C and adding the amount of Li_2CO_3 necessary to have a Li/Co molar ratio of 2.05 which differs from the results obtained in the literature, where good results were obtained with a Li/Co molar ratio of 1.05.[16,35,40] No impurity diffraction peaks appeared in the samples after relithiation. When the cathode was degraded, the diffraction peak of Li_xCoO_2 at (003) plane was at $2\Theta = 18.61°$, when $x = 0.5$. After relithiation, when $x = 1$, the corresponding diffraction peak at (003) clearly shifted to a higher angle, reflecting a decrease in the c lattice constant, appearing at $2\Theta > 18.9°$, which had been already observed in previous studies published by Zhao et al.[13]

After optimizing the solid-state sintering process, this was compared with hydrothermal regeneration. The chemical composition of regenerated cathodes is presented in Table III, where SS-LCO refers to the cathode regenerated by solid-state sintering and H-LCO refers to the cathode regenerated by the hydrothermal process, together with the chemical composition of the spent cathode (S-LCO).

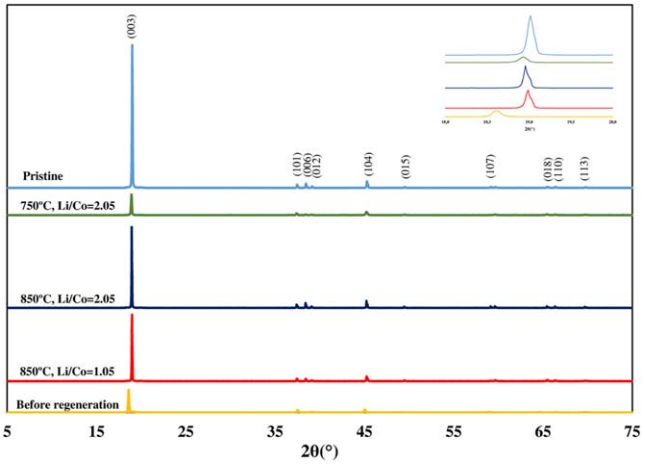


Figure 4. XRD patterns of the regenerated materials sintered at different temperatures and with different amount of Li_2CO_3.

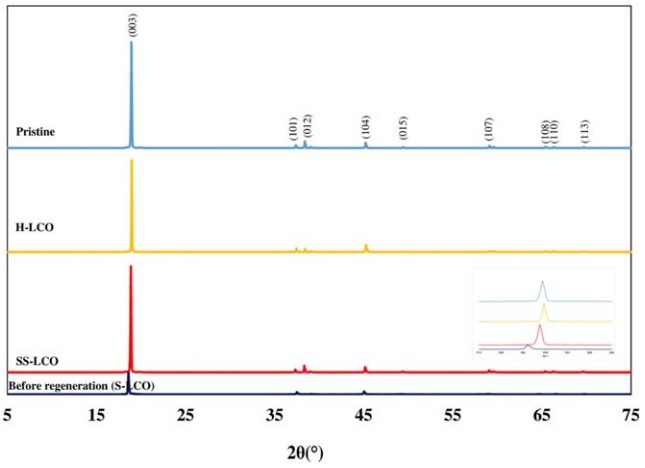


Figure 5. XRD patterns of the regenerated cathodes.

Table III. Co and Li content of the spent and regenerated cathodes.

	S-LCO	SS-LCO	H-LCO
Co (wt%)	63.3	60.0	59.4
Li (wt%)	3.3	6.3	6.8

As it can be seen, both regeneration processes were efficient from a chemical point of view as lithium and cobalt content of the regenerated cathodes was closed to the pristine one.

Figure 5 shows the comparison of the XRD patterns of the regenerated cathodes, together with the same cathode before regeneration.

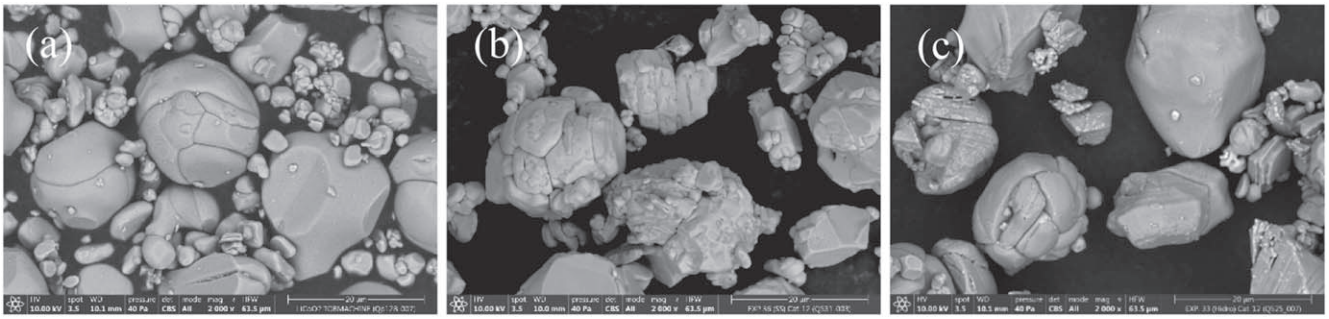


Figure 6. SEM of the (a) pristine, (b) SS-LCO, and (c) H-LCO.

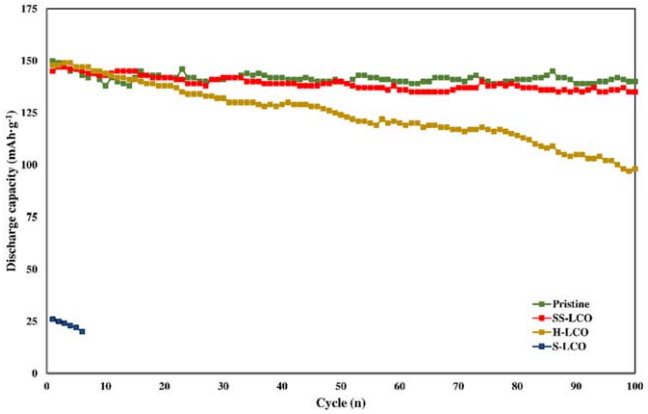


Figure 7. Cycle life of the spent and regenerated cathodes with different regeneration processes.

The intensity of the (003) peak of the regenerated cathodes considerably increased using both regeneration processes, although the cathode regenerated using the solid-state sintering method presented a higher peak intensity. Besides, the shifting to a higher angle when $x = 1$ in Li_xCoO_2 was also observed with both processes. The SEM images of the regenerated powders are presented in Fig. 6, together with the pristine, to visualize difference in morphology due to the relithiation processes.

The morphology and particle size of the regenerated powders remain nearly the same after regeneration under different conditions, with good similarity with the pristine. Both regenerated powders showed similar morphology and particle size, suggesting that the regeneration approach does not affect their morphology and sizes, as detected by other researchers.[41,42]

Finally, regenerated cathodes were electrochemically tested together with the pristine and the spent, the cycle life of all of them being displayed in Fig. 7.

As expected, spent cathode showed deficient performance at low C-rate, and it was not able to retain the charge. The discharge specific capacity of the cathode regenerated by the solid-state sintering process was about 145 $mAh{\cdot}g^{-1}$, which is close to that of the pristine (around 150 $mAh{\cdot}g^{-1}$), and the cycle life was also excellent as after 100 cycles it still had more than 90% of its initial capacity. The pleasant electrochemical performance could be explained because under the relithiation process, the lithium ions from the lithium source were promoted to enter the degraded cathode, occupying the voids in $LiCoO_2$ crystal, and rebuilding the $LiCoO_2$ layer structure.

The discharge capacity of the regenerated cathode by the hydrothermal method was about 148 $mAh{\cdot}g^{-1}$, which is also good, but the cycle life was poor as after 100 charge/discharge cycles it had lost 33% of its initial capacity. It can be seen that the solid-state sintering regenerated cathode displayed the best electrochemical performances. The results are consistent with the XRD results shown in Fig. 5, where higher peak intensity of the (003) peak in SS-LCO was observed.

So, the optimal regeneration process would consist in the separation of the degraded cathode by the developed process using mechanical and thermal processes and the subsequent regeneration using a solid-state sintering process at 850 °C for 12 h with a ramping rate of 5 °C min^{-1} in air atmosphere and adding the necessary amount of Li_2CO_3 to make the mixture cathode:Li_2CO_3 with a Li/Co molar ratio of 2.05.

It is a solution for the recovery of cathodes more environmentally friendly, as neither the use of acids is needed (compared to hydrometallurgy) nor the use of very high temperatures (compared to pyrometallurgy) and a subsequent synthesis process to obtain the cathode again. The separation of the cathode and its regeneration is only needed.

Conclusions

From the study of 160 disposed laptop batteries, it was concluded that LCO chemistry is the preferred for this application. Therefore, this study was undertaken over LCO cathodes although it is interesting to widen it to NMC cathodes as it can be the major chemistry in the near future.

The implementation of a regeneration process at industrial level will be possible thanks to the automatic cathode separation process, permitting the recycling of lithium-ion batteries be a reality.

This study revealed that separation process did not play an important role in the subsequent regeneration step when working with LCO.

Chemical and phase identification results showed that the composition and structure was recovered after the relithiation process. Electrochemical characterization showed excellent efficiency for the regenerated material by the solid-state sintering method after 100 cycles of charge/discharge at low C rates (0.2C), while cathode regenerated by the hydrothermal method showed poor cycle life.

The study should be enlarged to at least 500 cycles if the regenerated cathode is expected to be used in laptop batteries. Finally, the extension of this research to electric vehicle batteries and the possibility of recoveringtheir cathodes would make the electric vehicle a reality.

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