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Reclamation of Lithium Cobalt Oxide from Waste Lithium Ion Batteries to Be Used as Recycled Active Cathode Materials

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Abstract

Waste laptop batteries (Type-Lithium ion) have been collected and manually dismantled in the current work. Active electrode materials were scraped off from the copper current collector and polyethylene separators. The aluminum current collectors were found to be severely damaged and attached with the electrode material. It was treated with NaOH later to be recovered as Al_2O_3. The leaching of $LiCoO_2$ was done by 3 M HCl aided by 5% H_2O_2 at 60˚C from the scraped active electrode materials ($LiCoO_2$ and graphite) leaving the graphite completely. Co was precipitated as hydroxide by the addition of NaOH and later converted to Co_3O_4. The remaining solution was treated with saturated Na_2CO_3 to acquire Li_2CO_3 as crystalline precipitate with high purity. The recovery of Co and Li was 99% and 30%, respectively. Co_3O_4 and Li_2CO_3were mixed in stoichiometric proportions and calcined around 950˚C with air supply to achieve $LiCoO_2$ successfully.

Keywords

$LiCoO_2$, Lithium Ion Battery (LIB), Reclamation of $LiCoO_2$, Leaching, Active Cathode Materials

1. Introduction

In the current world of technology, efficient connectivity greatly depends on mobility. Portable electronic devices which have evolved almost as our electronic organs thrive on rechargeable power sources—currently on lithium ion batteries (LIBs). The construction of such batteries utilizes Lithium compounds—Lithium Cobalt Oxide ($LiCoO_2$) being the most popular of them all—as active cathode materials. $LiCoO_2$ acts as storage of electricity while charging and as a source while discharging. This costly component turns into the major environmental danger after the service life of the battery if improperly disposed [1]. The cobalt in such materials posses threats to the ecology being a heavy metal. Lithium being explosive in nature in the elemental form carries the risk of accidental explosion and toxic gas emission while burnt off or ill-treated during informal recycling [2] [3].

LIBs find their use commonly in laptop computers, cellular phones, camera, rechargeable lights and numerous modern life appliances and very recently in electric vehicles. An estimated total production of 12.7 billion mobile phones, 94.4 million laptop computers, and 768.9 million digital cameras [4] was reported by the United Nations till 2010. Clearly equal number of LIBs is to be handled after their lifetime. For the year 2016 alone, 4.7 billion unique mobile phone subscribers were reported [5]. Estimated 135.98 million [6] domestic mobile phone subscribers were present in mid 2017, which forecasts the number of LIBs joining the waste stream very soon. Approximately 0.3 million [7] computers are being consumed each year in the country, of which a substantial amount is the laptop computers having LIBs.

The importance of extractable metals from LIBs for a country like Bangladesh with no primary metallic sources cannot possibly be over exaggerated, while sorted electronic waste stream has already found its way to efficient recycling facilities abroad. In addition, if the key ingredient—$LiCoO_2$—can be recycled locally, domestic manufacturing of LIB could be possible empowering national economy.

From the late ninety's, researchers have been working on the development of recycling opportunities from LIBs. Thermal [8] and Mechanical [9] treatments have been experimented successfully. Solvent extraction by PC-88A [10], Acorga M5640 [11] and Cyanex 272 [11] [12] have yielded great recovery rates, however, the availability and post treatment for such solvents along with the cost associated needs much consideration. Although dissolution by N-methylpyrrolidone (NMP) has been proven [2] [13] helpful, the authors found difficulties with the output (trials with DMSO were also unsuccessful). Chemical leaching with organic [14] [15] or inorganic [16] [17] acids and subsequent precipitation of metallic salts to be used as precursors for the formation of active cathode materials-deems to be the most attractive option till today due to the low cost associated and process simplicity. The current work utilizes HCl leaching of $LiCoO_2$ aided by H_2O_2 as reducing agent after laborious yet simple manual dismantling of lithium ion batteries from laptop computers. Cobalt and lithium were then precipitated prudentially by basic treatments as $Co(OH)_2$ (later turned to Co_3O_4) and $Li_2(CO_3)$. Finally they were mixed and calcined to produce $LiCoO_2$.

2. Materials and Methods

2.1. Materials

Waste laptop batteries (Brand: HP, Type: Li ion, 6 cell) were collected from the local scrap market (Elephant road, Dhanmondi, Dhaka). The chemical reagents

(HCl, NaOH, H_2O_2 and Na_2CO_3) used in this study were of analytical grades (Merck, Germany and Scherlue, Spain). For all purposes de-ionized water (pH 6.5 - 7.5) was used.

2.2. Methods

For the estimation of carbon and organic contents, a CHNS-O (Brand: Thermo Fisher, Model: Flash 2000, Origin: USA) was used. To identify the metallic elements in the active electrode materials, a WDXRF (Brand: Shimadzu, Model: LAB Center XRF-1800, Origin: Japan) was used. Phase identification data of several intermediate products and raw materials were obtained using XRD (Brand: Bruker, Model: D8 Advance, Origin: Germany). Concentration of ions in different intermediate solutions was characterized by an AAS (Brand: Shimadzu, Model: AA7000, Origin: Japan). A (Brand: Jeol, Model: 71031SE2A, Origin: Japan) FESEM was used to acquire micrographs.

3. Experimental

3.1. Mechanical Breaking

Waste laptop batteries were mechanically broken with pliers to separate the plastic casing, connectors, thermo-couples and additional materials from the 6 cell assembly. Each cell was discharged by dipping in a 5% NaCl solution for 2 hours. The cells were then broke mechanically under a fume hood with pliers and a screw driver to unravel the coiled PE separators and the electrodes to be kept on a steel tray inside the fume hood to naturally dry for 3 days.

3.2. Separation of Aluminum and Active Electrode Materials

The Cu electrodes and PE separators were then manually scraped to remove active electrode materials (AEM, consisting graphite and $LiCoO_2$) as far as possible. Further scraping was done by the wet brushing of PE separators with DM water and a brush.

The Al electrodes and attached AEM were treated with 1M 500 ml NaOH. The Al foil reacted with the NaOH to form $Na_2Al_2O_4$ and the AEM got separated. This portion of AEM was treated again with 1 M 100 ml NaOH to assure any un-reacted Al got in the solution. The solution (total 600 ml) was then filtered to recover the AEM and washed subsequently with DM water before drying at 100˚C overnight. The filtrate was treated with dry CO_2 bubbling through it with rigorous stirring. Eventually the solution turned white and $Al(OH)_3$ precipitation formed. It was observed that this process was accelerated in warm (~50˚C) condition. It was then filtered and washed before drying overnight at 100˚C. The filtrate is again treated with dry CO_2 to recover additional $Al(OH)_3$ precipitates. The amount achieved from this second treatment was reasonably small. It was then filtered and dried the same way. A portion from the filtrate was taken for Atomic Absorption Spectrometer (AAS) to quantify any remaining amount of Al. The dried $Al(OH)_3$ was characterized with X-Ray Diffractometer to identify any possible impurities and confirm the $Al(OH)_3$ phase(s) formed. It was finally treated at 600˚C for 4 hours to get Al_2O_3. The separated AEM were ball milled to fines. The milled AEM was analyzed in an elemental analyzer (CHNS-O) to quantify the amount of graphite and organic materials remaining. Also X-Ray fluorescence was done to reconfirm the type of active cathode material.

3.3. Leaching

The leaching and simultaneous removal of graphite from the AEM was done by 3 M HCl with a solid to liquid ration of 1:20, aided by 5% H_2O_2. The solution was then filtered to acquire the graphite particles on the filter paper to be washed and dried subsequently. The dried graphite powder was then analyzed by XRD to confirm the absence of $LiCoO_2$.

3.4. Cobalt and Lithium Recovery

A portion of this solution was analyzed in the AAS to estimate the amount of leached Co and Li. Rest of the solution was treated with 65 ml 4 M NaOH (per 100 ml leach solution) to reclaim Co as cobalt hydroxide precipitates at near pH 11. The $Co(OH)_2$ obtained was then treated at 600˚C for 3 hour to get Co_3O_4. The remaining solution was condensed to half of the initial volume by heating. Later it was treated with saturated $Na_2(CO_3)$ and boiled for some time to get a precipitate of Li_2CO_3. The solutions–before and after the Li separation-were also taken for AAS analysis to measure the Co and Li recoveries respectively.

3.5. $LiCoO_2$ Formation

To synthesize $LiCoO_2$ stoichiometric proportions of Co_3O_4 and Li_2CO_3 was calculated to be 1:1.5 (molar) according to the following equation:

$$6Li_2CO_3(s) + 4Co_3O_4(s) + O_2(g) \xrightarrow{\Delta} 12LiCoO_2(s) + 6CO_2(g)$$

A set of samples of total mass 1 gm was prepared by mixing the weighted reactants in a mortar-pestle and pressing in a die up to 20 kpa to form green tablets. The tablets were then sintered in a tube furnace with and without air supply. The sintering cycles were similar to **Figure 1**.

The reaction suggests that, O_2 is required for the fulfillment of the reaction. Hence, later experiments were done with an additional air flow provided by a pump attached with the tube furnace. The tablets were characterized by XRD after each time period of sintering, to identify the phases formed.

4. Results

To evaluate the recovery process and possible electrode material loss, the physical dimensions of the electrode current collectors and the PP separators were measured (**Figure 2**).

Table 1 and **Table 2** summarize the nominal amounts of the overall constituents of LIB.

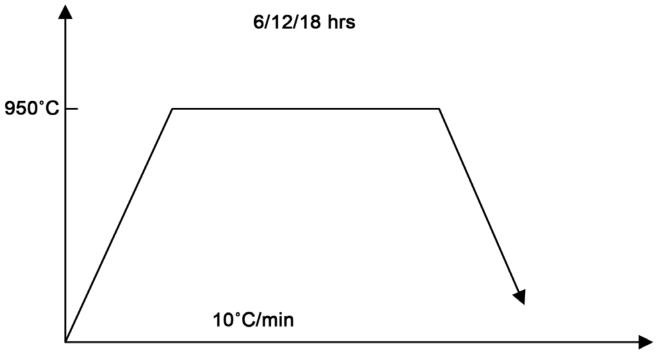

Figure 1. Sintering cycle for the synthesis of $LiCoO_2$.

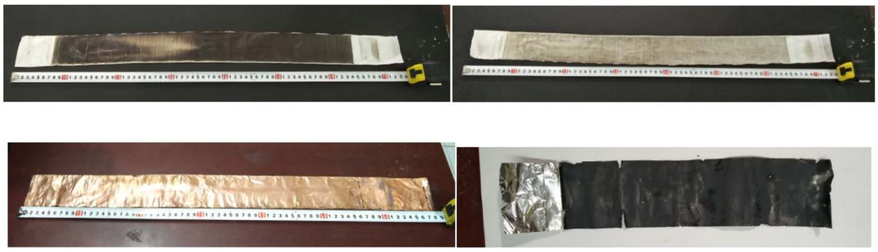

Figure 2. Separators (top) and Electrodes (bottom) of LIB.

Table 1. Measurements of the electrodes and separator.

Measurements	Length, mm	Width, mm	Thickness, μm	Calculated mass, gm
Cu	680	58	8	16.96236
Al	680	57	12	7.534944
PE Separator	740	58	14	6.82119

Table 2. Amount of different constituents of a LIB.

Constituent	gm	%
Cell Assembly	316.0	100
Plastic Casing, Connector, Thermocouple	50.0	15.8
6 Cell	266.0	84.2
Cell Casing, separators, parts	51.0	16.1
Cu Electrodes	17.2	5.4
PE Separator	6.8	2.2
Cathode Material	130.9	41.4
graphite	31.6	10.0
Al Electrode	7.5	2.4
Organics and Volatiles	20.9	6.6

4.1. Active Electrode Material Identification

The active electrode materials were identified by X-Ray Fluorescence (XRF) analysis (Table 3). It gave a definite indication that the active cathode material

Table 3. Amount of different elements in the AEM.

	XRF Data			CHNSO analyzer Data				
Element	**Co (%)**	**Cu (%)**	**Al (%)**	**C (%)**	**N (%)**	**H (%)**	**S (%)**	**O (%)**
Quantity	96.95	1.53	1.51	45.63	0.73	0.33	~0	~0

was rich in Co, later identified to be $LiCoO_2$. Lithium cannot be effectively identified by XRF, being a lighter (atomic no: 3) metal. There was an indication of the presence of small amounts of Cu and Al in the cathode material, possibly due to corrosion and the dismantling-scraping process.

Also the content of graphite was estimated with an Elemental (C, H, N, S, O) Analyzer. The presence of insignificant amounts of H, N, S and O indicated that almost all organic materials used were volatile and the remaining was mainly carbon (graphite). From this result, the amount of $LiCoO_2$ in the electrode material was estimated to be 54% (round figure) to be used in the leaching calculations.

4.2. Reclamation after Leaching

The Atomic Absorption Spectroscopy results are Table 4.

It depicts the concentration of metallic ions in solutions achieved in different steps. The phases of the reclamation products were identified by XRD later on.

4.3. XRD Analysis

The XRD analysis of active electrode materials (Figure 3) detected the presence of Graphite (JCPDS: 65-6212) and $LiCoO_2$. After the removal of Al current collector by NaOH, subsequent CO_2 treatment and drying; $Al(OH)_3$ (as Bayerite, JCPDS: 20-0011) was formed, which turned to Al_2O_3 (JCPDS: 50-0741) after calcination at 1000˚C for 1 hour (Figure 4). The residue after leaching was almost pure Graphite (JCPDS: 65-6212). The reclamation products were identified to be $Co(OH)_2$ (later calcined to Co_3O_4 (JCPDS: 73-1701) at 650˚C for 1 hour) and Li_2CO_3 (as Zabuyelite, JCPDS: 83-1454) as shown in Figure 5 and Figure 6. $Co(OH)_2$ was amorphous in nature, as a result no crystalline pattern was achieved in the XRD.

The formation of $LiCoO_2$ at 950˚C for different time durations (6, 12, 18 hours) showed different scenarios (Figure 7). Without air the 6 hour treatment showed the formation of less crystalline $LiCoO_2$ (JCPDS: 50-0653) and an impurity phase, possibly $LiAlO_2$ (JCPDS: 33-0776)–formed in contact with the porcelain boat. After the improvisation of additional Al_2O_3 layer below the green tablet, the 12 hour treatment showed much crystalline $LiCoO_2$ and Co_3O_4 (JCPDS: 73-1701) and some unidentifiable impurity phase-as an indication of Li_2CO_3 loss and incomplete reaction. The 18 hour treatment resulted in $LiCoO_2$, Co_3O_4 and CoO (JCPDS: 77-7548). At this point it seemed conclusive that further treatment will never reach a complete conversion to $LiCoO_2$, as there is a Li deficiency and a possible tendency of Co_3O_4 to convert to CoO with time under the treatment conditions.

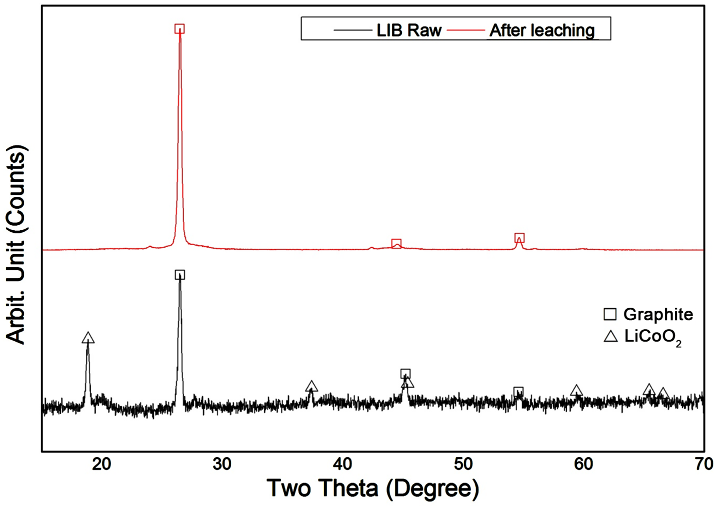

Figure 3. XRD spectra of LIB active cathode materials before and after leaching.

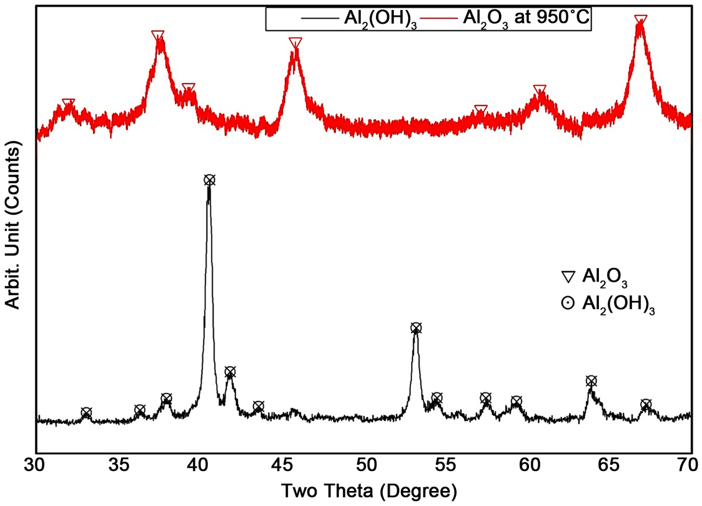

Figure 4. XRD spectra of aluminum extraction as alumina.

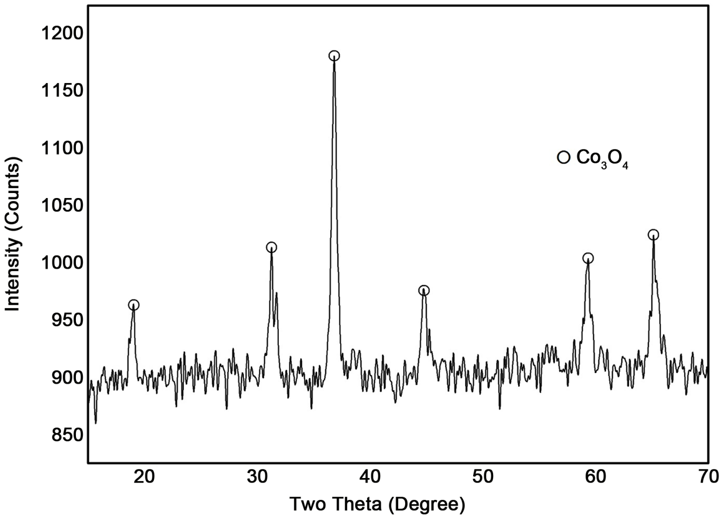

Figure 5. XRD spectra of cobalt oxide synthesis.

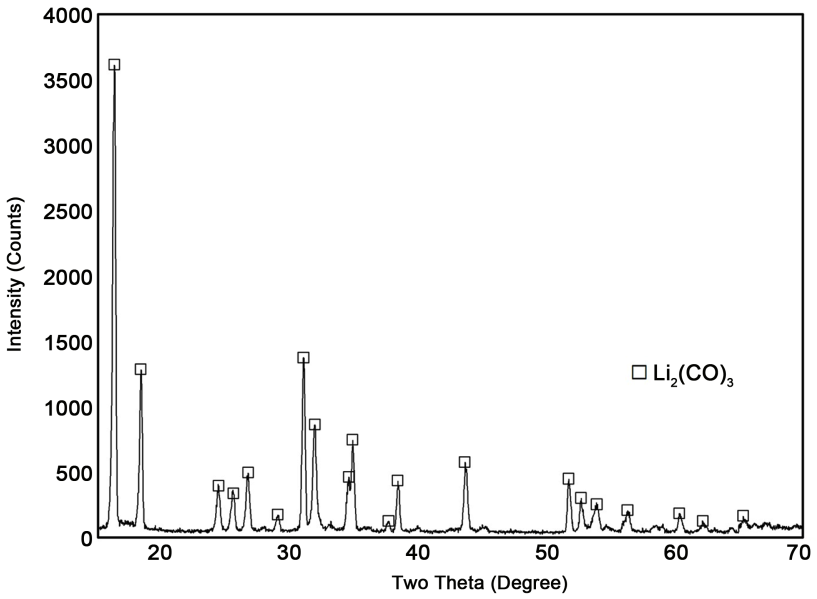

Figure 6. XRD spectra of lithium carbonate synthesis.

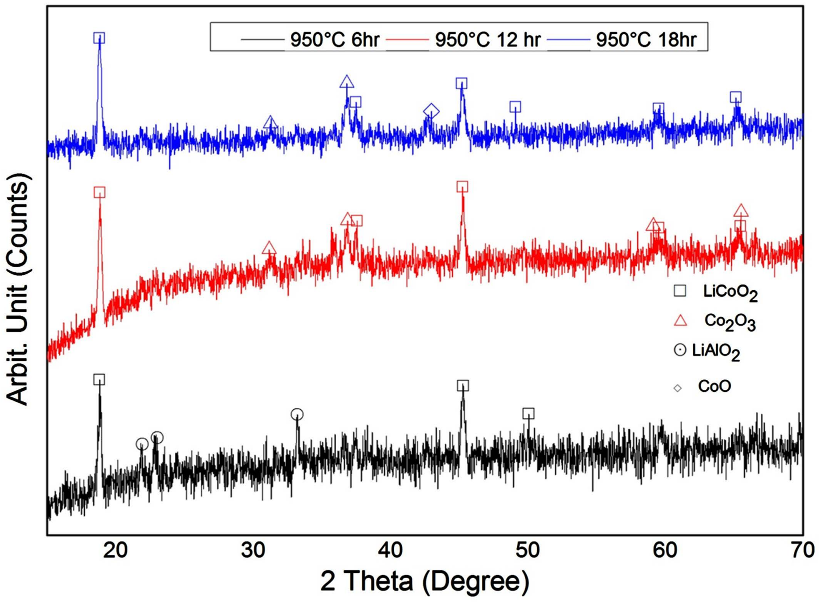

Figure 7. XRD spectra of $LiCoO_2$ synthesis without air.

Table 4. Normalized AAS data of solutions derived at different stages.

Elements	Concentration (ppm)				Recovery (%)
	After Al removal	Leach Solution	After Co removal	After Li removal	
Li	0	2424.1	2407.52	1741.5	30%
Cu	0	79.21	0	0	-
Al	248.72	198.7	105.5	108	-
Co	0	17404	1.48	0	99%

With air, the 6 hour treatment at 950˚C showed complete crystalline formation of $LiCoO_2$ (JCPDS: 50-0653) with no observable impurities (**Figure 8**). The product remained the same in the later experiments with prolonged time durations to 12 and 18 hours.

4.4. SEM Micrographs

The SEM micrographs shown in **Figure 9** depicted irregular shaped grains of synthesized $LiCoO_2$ particles throughout the sintered body.

5. Discussion

The dismantling process proved to be an effective one, regardless the labor intensive procedure requiring very little sophistication. Needless to mention, the safety requirements for the dismantling process-fume hood, safety goggles, gloves, etc. were maintained as required.

It was observed that the discharging step of the cells in brine solution initiated some leakage (through the cell vent) to the brine solution and turned the solution color to brown/orange. After some time, the solute precipitates itself and could be effectively filtered. It was assumed to be the commonly used electrolyte (LiPF6) dissolved in Ethylene/Dimethyl Carbonate (EC/DMC) solvent, but the assumption could not be verified as the amount of the filtrate was very small and it projected no crystalline peaks in XRD.

It was found that aqueous stirring or ultrasonication had poor effect on the separation of electrode materials from the metallic electrodes and the polymeric separators. Even when DMSO solvent used for PVDF binder removal, ultrasonication resulted in a separation of small metallic particles. As a result scraping was deemed to be the optimistic way.

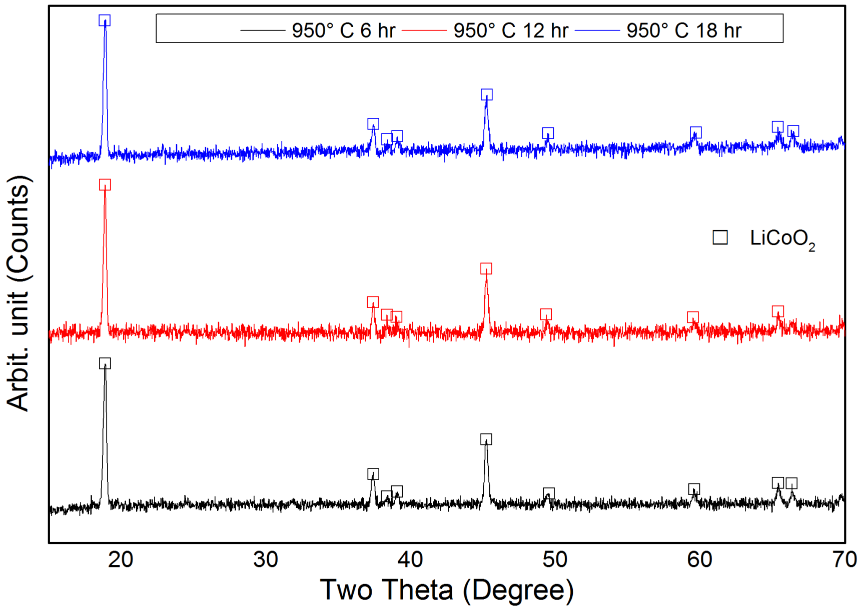

Figure 8. XRD spectra of $LiCoO_2$ Synthesis with air.

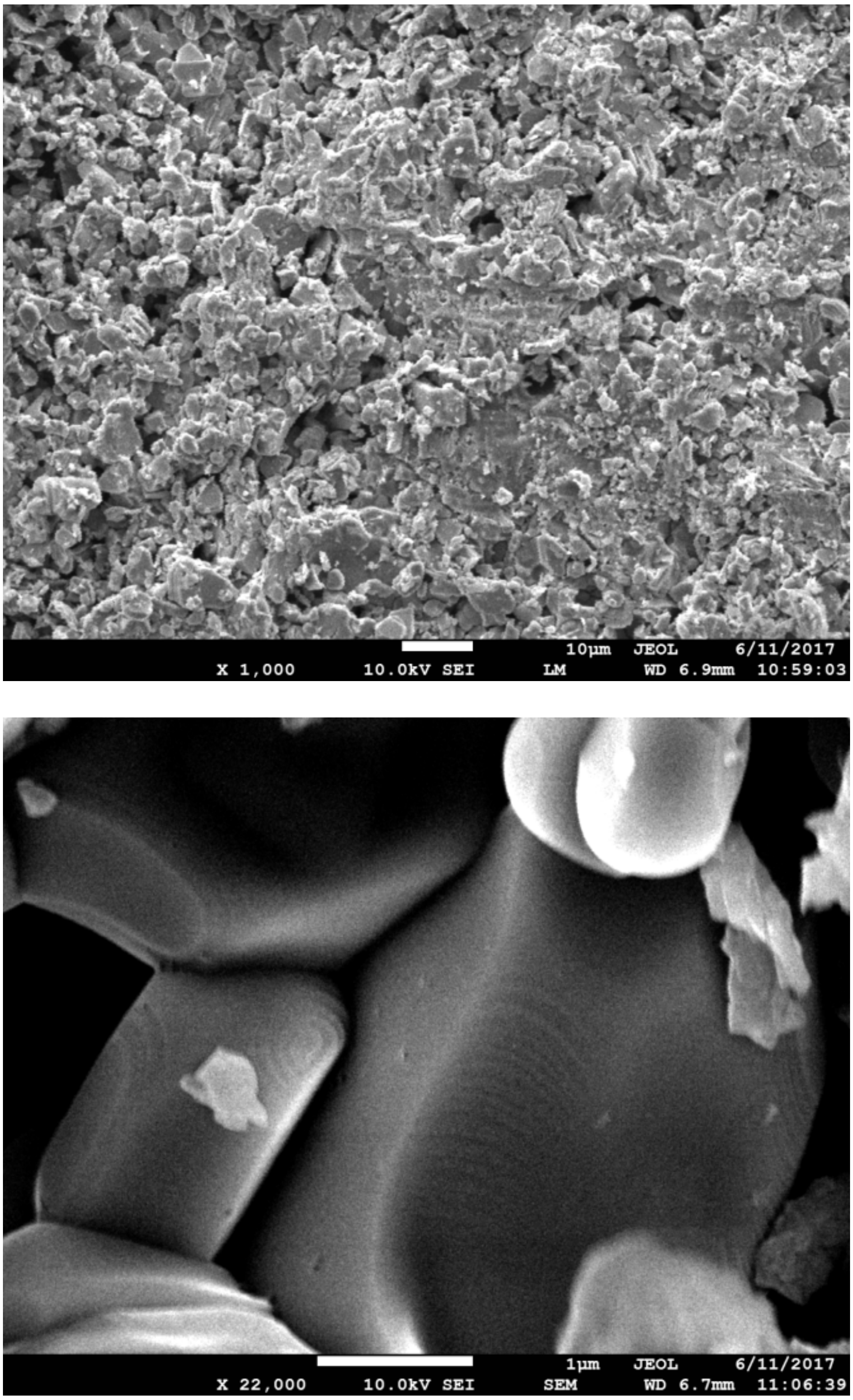

Figure 9. SEM micrographs of synthesized $LiCoO_2$.

The separation was very easy for the Cu electrodes because it contained mainly graphite on both sides, unlike the Al electrodes containing graphite as well as $LiCoO_2$ pasted with organic binder(s). As for the PE separators, the scraping process was intermediately successful leaving a good amount of graphite and $LiCoO_2$ still attached on both side. It was later wet scraped. Aluminum Electrodes—in most of the cases—got highly corroded and blistered; possibly due to rapid charge/discharge of the cell assembly in actual use. Also the PE separators got black shades in some cases, on the $LiCoO_2$ side due to the same reason. At some points Al electrodes and AEM got severely attached with the PE separators, possibly because of the severe heat generated due to charging/discharging rapidity. Hence brushing did not help the separation of AEM from Al electrodes. And negligible amounts of AEM were stuck with the PE separators even after brushing treatment.

The mechanical breakage of the cell assembly and later the cells, readily provided reusable or recyclable Cu foils, PE separators, steel casings and plastic (PE, PP, Bakelite, etc.) portions. After the leaching process, the almost pure graphite was achieved which also could find numerous applications. The NaOH treatment of the cathode dissolved the Al and was later recovered by the CO_2 bubbling. The calcined $Al(OH)_3$ turned to Al_2O_3 which has more commercial value.

During experiments it was found that without milling complete leaching of $LiCoO_2$ is tough as it gets trapped inside the graphite. The sheet like layered structure of graphite might also contribute to this effect by trapping Li^+ ions inside from both the electrolyte and the reaction product of the AEM from charging/discharging cycle. Hence the AEM was milled to fines.

The AAS data confirms that the aluminum current collector has been effectively removed from the cathode. For Cobalt and Lithium ions derived from the leaching of $LiCoO_2$, 99% Co and 30% Li could be successfully recovered by the leaching and reclamation process. The Li recovery was low due to some process factors. The solubility of Li_2CO_3 in water decreases with increasing temperature. There was no clear indication of completion of the formation of Li_2CO_3 at near 100˚C. Subsequent filtering also incorporated the solidification of some NaCl crystals on the filter paper alongside Li_2CO_3, as the solution was already very rich in Na^+ and Cl^- ions. The precipitates were washed at least 5 times with boiling DI water to remove the NaCl. Collaterally some Li_2CO_3 were lost in the process. Although the recovery of Li_2CO_3 was less, the purity (confirmed by XRD) was very high.

The formation of $LiCoO_2$ depends on various parameters like composition, environment and heating rate, etc.. Researchers [18] [19] have formed $LiCoO_2$ from various raw materials at above 800˚C with different time duration (close to 24 hrs). A simple sintering cycle was hence chosen to verify the outcomes.

The addition of air supply with the tube furnace provided tremendous results. The formation of $LiCoO_2$ was complete in all the samples. It could be suggested that under such conditions $LiCoO_2$ formation may be complete even in less time. The authors hope to perform electrochemical tests in order to measure the reversible capacity of the re-synthesized $LiCoO_2$ as future research. It was observed that porcelain boats (made of Kaolinite clay, a form of alumino-silicates) reacts with Lithium salts to produce aluminates or silicates if there is a contact with the green tablet. Hence, a layer of α-Al_2O_3powder was put on the porcelain boats bellow the green tablet(s) later on.

6. Conclusion

In this study, a lithium ion battery assembly has been manually broke into constituent electrodes, separators and other components. A proven reclamation process of Li and Co has been investigated by leaching with HCl and precipitating Li and Co compounds by basic treatments. The compounds ($Li_2(CO_3)$ and Co_3O_4) have been effectively used to synthesize $LiCoO_2$ by a simple thermal treatment. Different characterization techniques have proven the process to be efficient, simple and cost effective.

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