

N.E. Abdul LatifMetallurgy & Production
Eng. Dept., University of
Technology, Bagdad, Iraq.
a_muwafaq@yahoo.com**A.M. Ahmed**Metallurgy & Production
Eng. Dept., University of
Technology, Bagdad, Iraq.

Received on: 02/06/2016

Accepted on: 29/09/2016

Recovery of Cobalt and Lithium from Spent Lithium Ion Batteries

Abstract- In this research a combination of mechanical separation, reductive leaching and chemical precipitation are used to regain Li as Li_2CO_3 and Co as $\text{Co}(\text{OH})_2$ from waste lithium ion batteries. First batteries are dismantled and different component are separated. Anode and cathode foils are grinded and sieved to extract Al and Cu from spent battery powder. The separation efficiency for copper and aluminum are 97.68% and 84.41% respectively. The separated powder is leached with sulfuric acid and hydrogen peroxide solution to dissolve cobalt and lithium. The effects of sulfuric acid concentration, time, temperature, concentration of hydrogen peroxide and S/L on the leaching efficiency for cobalt and lithium are studied. Design of experiments by Taguchi method is used to determine experiments conditions, analysis the results, and identify the optimum leaching conditions. The optimum leaching conditions are 1.5 M acid concentration, 60 minute, 60 °C, 15% H_2O_2 , and 40 g/l solid to liquid ratio. Leaching efficiencies of 94.07% Co and 98.15% Li are achieved with these optimum conditions. Leaching solution of the optimum experiment is used to recover cobalt as cobalt hydroxide and lithium as lithium carbonate by chemical precipitation. An equivalent volume of 4M NaOH is used to precipitate cobalt as cobalt hydroxide. After 1 hour 99.98%, Co is precipitated and collected by filtration and dried. After collection of the $\text{Co}(\text{OH})_2$ product, the remaining solution is treated with equivalent volume of "saturated solution" of Na_2CO_3 at 100 °C to recover lithium as Li_2CO_3 . After 1 hour, 80 – 85 % Li is precipitated, filtrated and dried. Precipitation products are investigated with XRD, which confirmed that cobalt hydroxide and lithium carbonate are recovered from leaching solution.

Keywords- Chemical precipitation, Cobalt, lithium. Reductive leaching spent lithium ion batteries.

How to cite this article: N.E. Abdul Latif and A.M. Ahmed, "Recovery of Cobalt and Lithium from Spent Lithium Ion Batteries," *Engineering and Technology Journal*, Vol. 35, Part A, No. 2, pp. 139-148, 2017.

1. Introduction

The developed technology of electronic devices has been reached all aspect of people's life. These devices such as laptops, mobiles, digital cameras ...etc. are mainly powered by lithium ion batteries because of its better efficiency compared with other rechargeable batteries. The most valuable part of waste LIBs is electrodic powder, which mainly consists of C pasted on Cu foils and LiCoO_2 pasted on Al foils [1,2]. The cathode contain high valued nonferrous metals, 5–15 wt.% cobalt and 2–7 wt.% lithium, which are greater than that exist in ores or ore concentrates [3]. Therefore, it would be important to recover cobalt, lithium and other valuable metals from waste LIBs to conserve resources and protect the environment [2]. Nowadays, the main recovery methods for spent LIBs consist of, pyrometallurgical operation, hydrometallurgical operation or the combination of both and microbial metallurgy. The hydrometallurgical operation is most widely utilized to regain different metals from waste li-ion batteries by leaching followed by selective recovery processes for metals. High leaching efficiencies of lithium and cobalt have accomplished by leaching with inorganic acids, including H_2SO_4 [2-10] HCl, and

HNO_3 [6,7]. Various organic acids such as malic [1,12], citric [11,12,13], aspartic acid [12], and ascorbic [12, 14] were investigated as substitute leaching media to avoid the minor contamination inserted from vigorous inorganic acids. Most researchers reported experimental results of acidic leaching with H_2SO_4 in the existence of H_2O_2 as reducing agent [3,6] in order to enhance the dissolution of cobalt. The second step deals with the selective recovery of lithium and cobalt from the acid solution. According to literature, several main techniques have already been investigated to recover metals from acid solution, such as chemical precipitation and solvent extraction [15, 16,17]. In this work a combination of sieving and reductive leaching then chemical precipitation are utilized to obtain Li and Co from waste LIBs.

2. Experimental

I. Sample Collection and Preparation

Laptop batteries are chosen to recover the valuable metals. Laptop batteries are dismantled manually removing plastic shells, electric connection wires and pack protection printed circuit boards (PCB) and to obtain lithium ion batteries of cylindrical shape.

After that, batteries are discharged, cut, internal material recovered and grinded to powder. The batteries powder is characterized by atomic absorption spectroscopy, X-ray diffraction, scanning electron microscopy and energy-dispersive spectroscopy. Table 1,

shows the chemical composition of batteries powder analyses by AAS. Figure 1, shows the XRD pattern for batteries powder. Figures 2, shows SEM and EDX results.

Table 1: Chemical composition of main elements in batteries powder

| Element | Wt. % |
|-----------|--------|
| Cobalt | 6.432 |
| Lithium | 4.372 |
| Aluminium | 5.32 |
| Copper | 14.94 |
| Others | 68.936 |

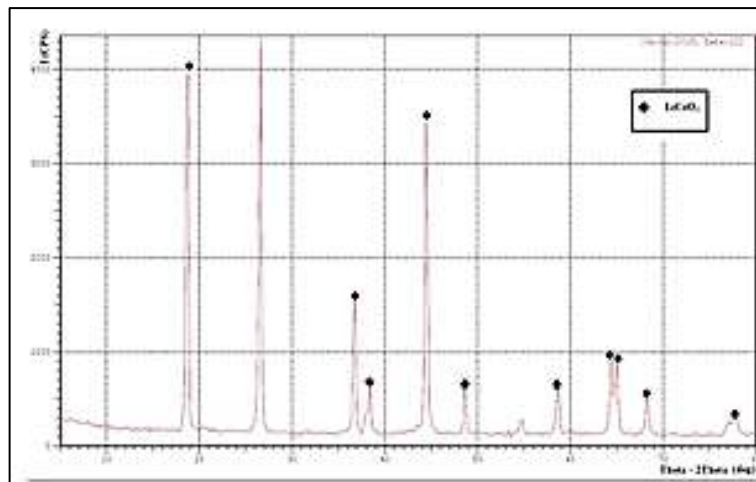


Figure 1: XRD pattern for batteries powder before leaching process

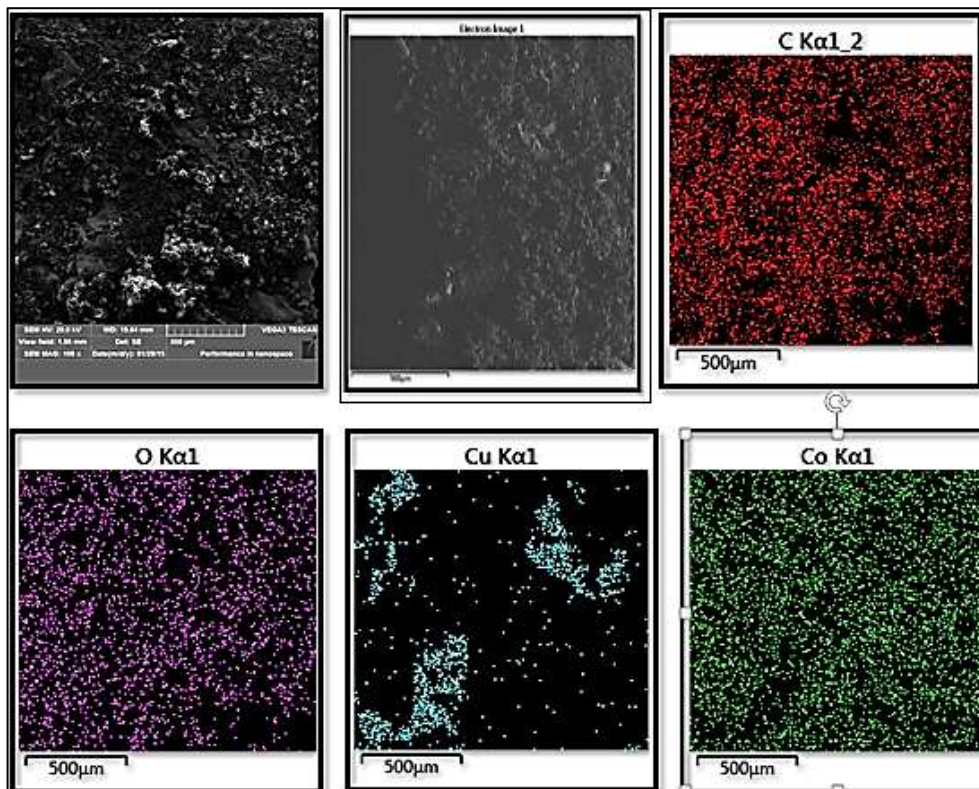


Figure 2: SEM and EDS mapping of spent batteries powder

II. Sieve Analysis or Mechanical Separation

This process is used to find out the possibility of separating copper and aluminum from the batteries powder on the bases of their particle size. Sieving process is carried out by using auto sieve shaker. Sieve sizes of [150 μm , 106 μm , 75 μm , 53 μm , -53 μm (pan)] are used for test using powder sample of 250 grams and sieving time of 10 minutes.

III. Leaching Process

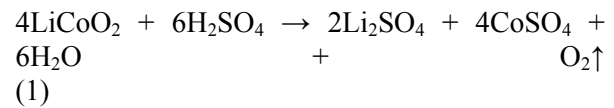
A. Design of Experiments

In order to determine the optimum leaching conditions during leaching experiments, design of experiments by Taguchi method is used. The studied parameters are sulfuric acid concentration, time, temperature, hydrogen peroxide concentration and solid/liquid ratio. Each of the previous parameters has four levels. Minitab program is chosen to select and create the proper Taguchi orthogonal arrays. Taguchi orthogonal arrays $L_{16} (5^4)$ which shown in Table 2, is selected according to the number of parameters and their level. Taguchi orthogonal arrays $L_{16} (5^4)$ is used to analysis leaching data and determine the optimum leaching condition.

B. Leaching Experiments

In this process batteries powder is leached to dissolve cobalt and lithium. In theory, only lithium cobalt oxide is dissoluble compound of batteries powder in H_2SO_4 with low concentration level. Therefore, to minimize volatilization contamination caused by reactants, and reducing the solubility of Cu in the leaching operation,

H_2SO_4 at appropriate concentration is selected as a more convenient leaching reagent in contrast to HCL and HNO_3 . Equation (1) illustrates this reaction [18].



Sulfuric acid is used as leaching agent along with peroxide hydrogen as reducing agent. The leaching cell consist of a 1000 ml three neck glass extractor, refluxing condenser, hot plate magnetic stirrer, holder and clamp, thermometer, impeller and rubber seals as shown in Figure 3. Leaching experiments are conducted in different H_2SO_4 concentration (0.5, 1, 1.5, 2M), temperature (25, 40, 60, 80 $^\circ\text{C}$), time (30, 60, 90, 120 Min.), solid-liquid ratio (40, 80, 120, 160 g/l), and concentration of hydrogen peroxide (0, 5, 10, 15%) to determine their effects on leaching efficiencies of cobalt and lithium. In the presence of hydrogen peroxide, Co is reduced to Co^{+2} from Co^{+3} to make it leachable with H_2SO_4 . Co^{+2} is more stable than Co^{+3} . In this case, Li and Co both are leached even at low concentration of H_2SO_4 [19]. The leaching process of LiCoO_2 in H_2SO_4 solution could be represented equation (2) [20].

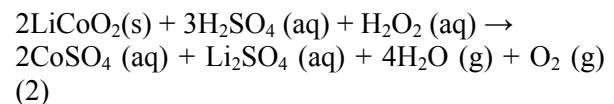


Table 2: Taguchi orthogonal arrays $L_{16} (5^4)$ for leaching condition

| No. | Acid Con. M. | time Min | Temp . $^\circ\text{C}$ | H_2O_2 % | S/L g/l |
|-----|-----------------|-------------|-------------------------------|-----------------------------|------------|
| 1 | 0.5 | 30 | 25 | 0 | 40 |
| 2 | 0.5 | 60 | 40 | 5 | 80 |
| 3 | 0.5 | 90 | 60 | 10 | 120 |
| 4 | 0.5 | 120 | 80 | 15 | 180 |
| 5 | 1 | 30 | 40 | 10 | 180 |
| 6 | 1 | 60 | 25 | 15 | 120 |
| 7 | 1 | 90 | 80 | 0 | 80 |
| 8 | 1 | 120 | 60 | 5 | 40 |
| 9 | 1.5 | 30 | 60 | 15 | 80 |
| 10 | 1.5 | 60 | 80 | 10 | 40 |
| 11 | 1.5 | 90 | 25 | 5 | 180 |
| 12 | 1.5 | 120 | 40 | 0 | 120 |
| 13 | 2 | 30 | 80 | 5 | 120 |
| 14 | 2 | 60 | 60 | 0 | 180 |
| 15 | 2 | 90 | 40 | 15 | 40 |
| 16 | 2 | 120 | 25 | 10 | 80 |

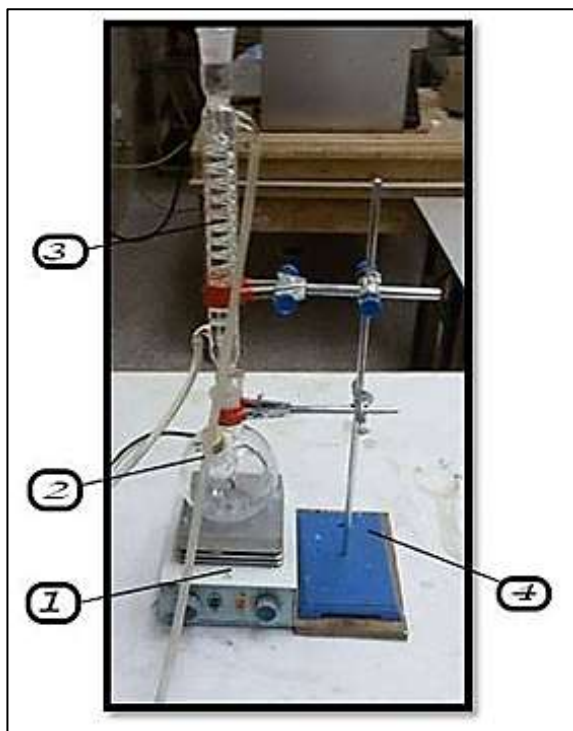


Figure 3: The leaching cell consists of: 1. hot plate magnetic stirrer, 2. three neck glass extractor, 3. refluxing condenser, and 4. holder and clamp

All leaching experiments are performed at a constant particle size of $-150\ \mu\text{m}$ and stirring speed of 100 rpm. After leaching, the leaching liquor is filtered and the solution is chemically analyzed by Atomic Absorption Spectroscopy type to determine ions concentration of lithium and cobalt. Leaching efficiencies for cobalt and lithium is calculated based on their concentration in the leaching solution.

IV. Precipitation of Cobalt and Lithium

The recovery of lithium and cobalt from filtered leaching liquor is performed by employing redox reaction. The pH of the solution had to be controlled by the addition of NaOH to precipitate cobalt [21]. Therefore, several concentrations of sodium hydroxide are tested to precipitate cobalt from leaching solution. After Cobalt is precipitated, the remaining liquor is separated by filtration, and treated with a saturated Na_2CO_3 liquor to precipitate Li [21]. The precipitation process is performed at a temperature of approximately 100°C . The remaining liquor after each precipitation step is chemically analyzed by Atomic Absorption Spectroscopy to determine concentration of the remaining lithium and cobalt and, to calculate the precipitation efficiency. The two precipitation products are dried in classic drying and heating chamber at 90°C for 30 Minute and analyzed by X-ray diffraction.

3. Results

I. Sieve Analysis or Mechanical Separation

In order to minimize the impurities (Cu and Al) an attempt is made to separate them by using sieve analysis to avoid the introduction of aluminum and copper ions in the precipitation steps. Because high concentration of aluminum and copper ions can go against the precipitation of Co if Cu and Al are leached in the leaching process. According to the sieve analysis results in Figure 4 and 5, 97.68% Cu and 84.41% Al are remained in $+150\ \mu\text{m}$ sieve. Therefore, the powder with particle size of $+150\ \mu\text{m}$ is removed and the powder with particle size of $-150\ \mu\text{m}$ is selected to be used in the following leaching experiments.

II. Leaching Process

A. Results Analysis and Identifying the Optimum Conditions

After finishing all leaching experiments, results are entered in the work sheet of Minitab program to analysis them. Tables 3 and 4 show Taguchi orthogonal arrays with its results, means and signal to noise ratio (SN) for cobalt and lithium, respectively. The optimum condition for each factor is determined by comparison between SN and mean of leaching efficiencies figures.

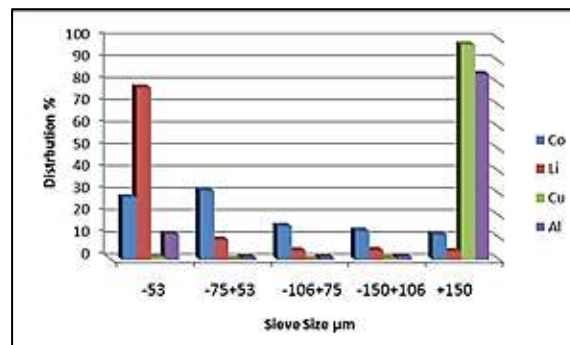


Figure 4: The distribution of batteries powder metals for different sieve size

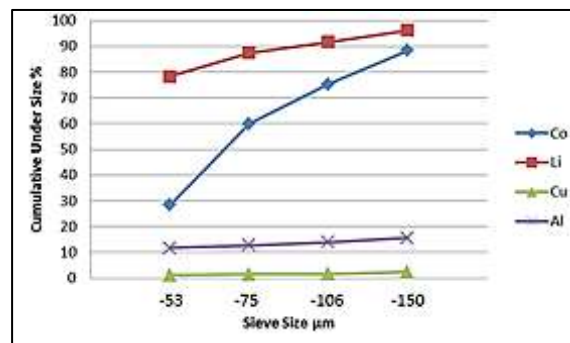


Figure 5: The size distribution analysis of batteries powder metals

Table 3: Taguchi orthogonal arrays $L_{16}(5^4)$ with its results for cobalt

| No. | Acid Con. M. | time Min. | Temp. °C | H ₂ O ₂ % | S/L g/l | Co ₍₁₎ % | Co ₍₂₎ % | Mean % | SNRA |
|-----|--------------|-----------|----------|---------------------------------|---------|---------------------|---------------------|---------|---------|
| 1 | 0.5 | 30 | 25 | 0 | 40 | 18.31 | 21.76 | 20.039 | 25.9408 |
| 2 | 0.5 | 60 | 40 | 5 | 80 | 40.53 | 42.59 | 41.5655 | 32.3667 |
| 3 | 0.5 | 90 | 60 | 10 | 120 | 28.64 | 36.07 | 32.3585 | 30.0277 |
| 4 | 0.5 | 120 | 80 | 15 | 160 | 21.82 | 25.07 | 23.4525 | 27.3412 |
| 5 | 1 | 30 | 40 | 10 | 160 | 53.38 | 44.67 | 49.026 | 33.7055 |
| 6 | 1 | 60 | 25 | 15 | 120 | 72.52 | 61.65 | 67.0885 | 36.4472 |
| 7 | 1 | 90 | 80 | 0 | 80 | 48.17 | 50.41 | 49.292 | 33.8487 |
| 8 | 1 | 120 | 60 | 5 | 40 | 85.01 | 84.95 | 84.985 | 38.5868 |
| 9 | 1.5 | 30 | 60 | 15 | 80 | 89.54 | 68.26 | 78.9045 | 37.7044 |
| 10 | 1.5 | 60 | 80 | 10 | 40 | 92.51 | 98.57 | 95.547 | 39.5912 |
| 11 | 1.5 | 90 | 25 | 5 | 160 | 44.69 | 54.58 | 49.6405 | 33.7874 |
| 12 | 1.5 | 120 | 40 | 0 | 120 | 40.46 | 35.10 | 37.7825 | 31.4802 |
| 13 | 2 | 30 | 80 | 5 | 120 | 71.11 | 50.31 | 60.7155 | 35.2815 |
| 14 | 2 | 60 | 60 | 0 | 160 | 41.09 | 35.62 | 38.3595 | 31.6113 |
| 15 | 2 | 90 | 40 | 15 | 40 | 83.57 | 81.47 | 82.527 | 38.3298 |
| 16 | 2 | 120 | 25 | 10 | 80 | 67.04 | 62.48 | 64.7665 | 36.2108 |
| RK | 1 | 4 | 5 | 2 | 3 | 5 | 8 | | 8 |

Table 4: Taguchi orthogonal arrays $L_{16}(5^4)$ with it results for lithium

| No. | Acid Con. M. | Time Min. | Temp. °C | H ₂ O ₂ % | S/L g/l | Li ₍₁₎ % | Li ₍₂₎ % | Mean % | SNRA |
|-----|--------------|-----------|----------|---------------------------------|---------|---------------------|---------------------|---------|----------|
| 1 | 0.5 | 30 | 25 | 0 | 40 | 42.128 | 38.985 | 40.5565 | 32.14164 |
| 2 | 0.5 | 60 | 40 | 5 | 80 | 53.264 | 60.835 | 57.0495 | 35.06763 |
| 3 | 0.5 | 90 | 60 | 10 | 120 | 35.126 | 44.467 | 39.7965 | 31.81702 |
| 4 | 0.5 | 120 | 80 | 15 | 160 | 44.131 | 32.488 | 38.3095 | 31.36406 |
| 5 | 1 | 30 | 40 | 10 | 160 | 58.363 | 58.985 | 58.674 | 35.36855 |
| 6 | 1 | 60 | 25 | 15 | 120 | 70.444 | 70.303 | 70.3735 | 36.94817 |
| 7 | 1 | 90 | 80 | 0 | 80 | 78.928 | 73.466 | 76.197 | 37.62202 |
| 8 | 1 | 120 | 60 | 5 | 40 | 89.202 | 79.503 | 84.3525 | 38.47887 |
| 9 | 1.5 | 30 | 60 | 15 | 80 | 85.694 | 73.485 | 79.5895 | 37.94039 |
| 10 | 1.5 | 60 | 80 | 10 | 40 | 94.07 | 85.445 | 89.7575 | 39.03133 |
| 11 | 1.5 | 90 | 25 | 5 | 160 | 68.76 | 77.193 | 72.9765 | 37.22014 |
| 12 | 1.5 | 120 | 40 | 0 | 120 | 66.968 | 51.967 | 59.4675 | 35.27776 |
| 13 | 2 | 30 | 80 | 5 | 120 | 86.442 | 54.484 | 70.463 | 36.28287 |
| 14 | 2 | 60 | 60 | 0 | 160 | 75.133 | 66.508 | 70.8205 | 36.95484 |
| 15 | 2 | 90 | 40 | 15 | 40 | 95.067 | 79.312 | 87.1895 | 38.70278 |
| 16 | 2 | 120 | 25 | 10 | 80 | 86.729 | 66.183 | 76.456 | 37.43228 |
| RK | 1 | 3 | 5 | 4 | 2 | | | | |

- *Effect of Acid Concentration*

To investigate the influence of H_2SO_4 concentration on leaching efficiency of the spent battery powder, leaching experiments are done in levels of 0.5 to 2 M. The results are presented in Figure 6.a. The results indicates that 29.35 % of Co and 43.93% of Li are leached out with 0.5 M. leaching efficiencies of Li and Co increased with the increase of acid concentration because of the excess reagent in the reactive system. At 1.5 M, leaching efficiencies for Li and Co are found to be 65.47% and 75.45% respectively. Further increase in acid concentration up to 2 M, didn't display any considerable effect in the reclaiming of Li and Co. This result can be demonstrated by equation (1) which points out that the increase in the reactive materials favors the forward reaction resulting in the increase in reaction efficiency. Meanwhile, the increase of $CoSO_4$ or Li_2SO_4 in the solution reversely can facilitate the backward reaction. Therefore, farther increase in (H_2SO_4) will make backward reaction turns to be in equilibrium with the forward reaction [3].

- *Effect of Time*

To study the influence of leaching time on leaching efficiency of the spent battery powder, leaching experiments are carried out in the range of 30 to 120 min. The results are presented in Figure 6.b. The results indicates that 52.17% of Co and 62.32% of Li are leached out within 30 min. Leaching efficiencies of the cobalt and lithium increased with the increase in leaching time. At 60 min, Leaching efficiencies of Li and Co are found to be 72% and 60.64% respectively. Further increase in time up to 120 min, didn't display any considerable effect in the reclaiming of Li and Co which mean that equilibrium is reached in 60 min reaction time. After that, the reverse reaction begins to take place.

- *Effect of Temperature*

To study the influence of leaching temperature on leaching efficiency of the spent battery powder, leaching experiments are carried out in the range of 25 to 80 °C. The results are presented in Figure 6.c. The results indicate that 50.38% of Co and 65.09% of Li are leached out at 25 °C. Leaching efficiencies of the cobalt and lithium increased with the increase in leaching temperature. At 60 °C, leaching efficiencies of Co and Li are found to be 58.65% and 68.64% respectively. Furthermore the increase in temperature up to 80

°C, didn't display any considerable effect in the reclaiming of Li and Co. The improved dissolution at a relatively low temperature of 60 °C is a result of the higher stability of hydrogen peroxide. At temperatures above, 60 °C H_2O_2 tends to rapidly decompose to water and oxygen.

- *Effect of Hydrogen Peroxide Concentration*

To study the influence of hydrogen peroxide concentration on leaching efficiency of the spent battery powder, leaching experiments are done in the range of 0 to 15 %. The results are presented in Figure 6.d. The results indicate that 36.37 % of Co and 61.76 % of Li are leached out at the absence of hydrogen peroxide. Leaching efficiencies of the Li and Co increased with the increase in hydrogen peroxide concentration. At 5%, the leaching efficiencies of Co and Li are found to be 59.22% and 71.21% respectively. Further the increase in hydrogen peroxide concentration up to 15%, shows an increase in the cobalt leaching efficiency to 62.99% while, the lithium leaching efficiency didn't show any considerable effect.

- *Effect of Solid Powder to Liquid Solution Ratio*

To study the influence of solid to liquid ratio on leaching efficiency of the spent battery powder, leaching experiments are carried out in the range of 40 to 160 g/l. The results are presented in Figure (6.e). The results indicate that 70.77% of Co and 75.46% of Li are leached out at 40 g/l. The leaching efficiencies of Li and Co decreased with the increase in solid to liquid ratio. At 160 g/l, the leaching efficiencies of Co and Li are found to be 40.12% and 60.2%, respectively. At low solid to liquid ratio the available surface area per unit volume of the solution is higher and therefor, Li and Co leaching is higher. However, the increase in the solid to liquid ratio decreases the available surface area per unit volume of the solution which causes decrease in leaching efficiencies of Li and Co.

B. *The Optimum Leaching Conditions*

In order to determine the optimum conditions for a leaching process, a comparison between the SN ratio and leaching efficiency figures for each factor was done. Figures 7, show the SN ratio of acid concentration, time, temperature, H_2O_2 concentration, and solid to liquid ratio.

The optimum leaching conditions are 1.5 M acid concentration, 60 Min., 60 °C, 15% H_2O_2 , and 40 g/l. The mean of leaching efficiencies predicted

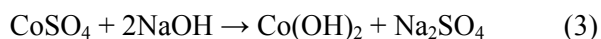
by Taguchi method for these conditions are 99.52% Co and 92.41% Li. Table 5, shows the results of optimum leaching experiments and their mean.

C. Analysis of Variance (ANOVA)

The effective percentage for each factor on the cobalt and lithium leaching efficiencies are calculated by Minitab program. The ANOVA calculation for cobalt and lithium are illustrated in Tables 6 & 7 respectively.

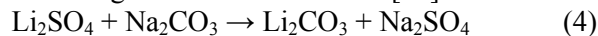
III. Recovery of Cobalt and Lithium

After determining the optimum conditions for the leaching process, leaching solution of the optimum experiment is used to recover cobalt by chemical precipitation as cobalt hydroxide $\text{Co}(\text{OH})_2$. One equivalent volume of 4 M NaOH solution is added to leaching solution. Precipitation of cobalt started at a pH of 6 and finished at a pH of 13. After 1 hour pink precipitate is filtered and removed from leaching solution and Co recovered. Cobalt precipitate according to equation [22].



According to the Atomic Absorption results 99.98% Co is precipitated. After that, the remaining solution is processed to recover lithium as Li_2CO_3 by adding saturated Na_2CO_3 solution. The precipitation operation is carried out at a temperature near 100°C due to the inverse proportional of Li_2CO_3 aqueous solubility to its temperature, e.g., $S_{\text{Li}_2\text{CO}_3}=1.52$ at 0°C and $S_{\text{Li}_2\text{CO}_3}=0.71$ (g/100 g H_2O) at 100°C , the precipitation operation is carried out at a

temperature near 100°C [23]. Equivalent volume of sodium carbonate reagent is added to the remaining solution. After 1 hour white precipitate is recovered after filtration. The Atomic Absorption results show that 80-85% Li is precipitated. Lithium precipitate following to the following the chemical reaction [22].



• Characterization of Recovered Products

According to the X-ray diffraction results, cobalt is precipitated as cobalt hydroxide as show in Figure 8 and, lithium is precipitated as lithium carbonate as show in Figure 9.

4. Conclusion

According to the results of this research, the following conclusions can be placed.

1- The mechanical separation can be used to remove copper and aluminum from waste LIBs powder with separation efficiencies of 97.68% and 84.41% respectively.

2- The optimum leaching conditions are 1.5 M acid concentration, 60 Min, 60°C , 15% H_2O_2 , and 40 g/l with leaching efficiencies of 94.07 % Co and 98.15 % Li.

3- Cobalt and lithium are recovered from leaching solution by chemical precipitation. Sodium hydroxide solution of 4M was added to leaching solution for 1 hour to recover 99.98 % Co as cobalt hydroxide. Saturated solution of sodium carbonate was added to the remaining solution at temperature close to 100°C for 1 hour to recover 80-85 % Li as lithium carbonate.

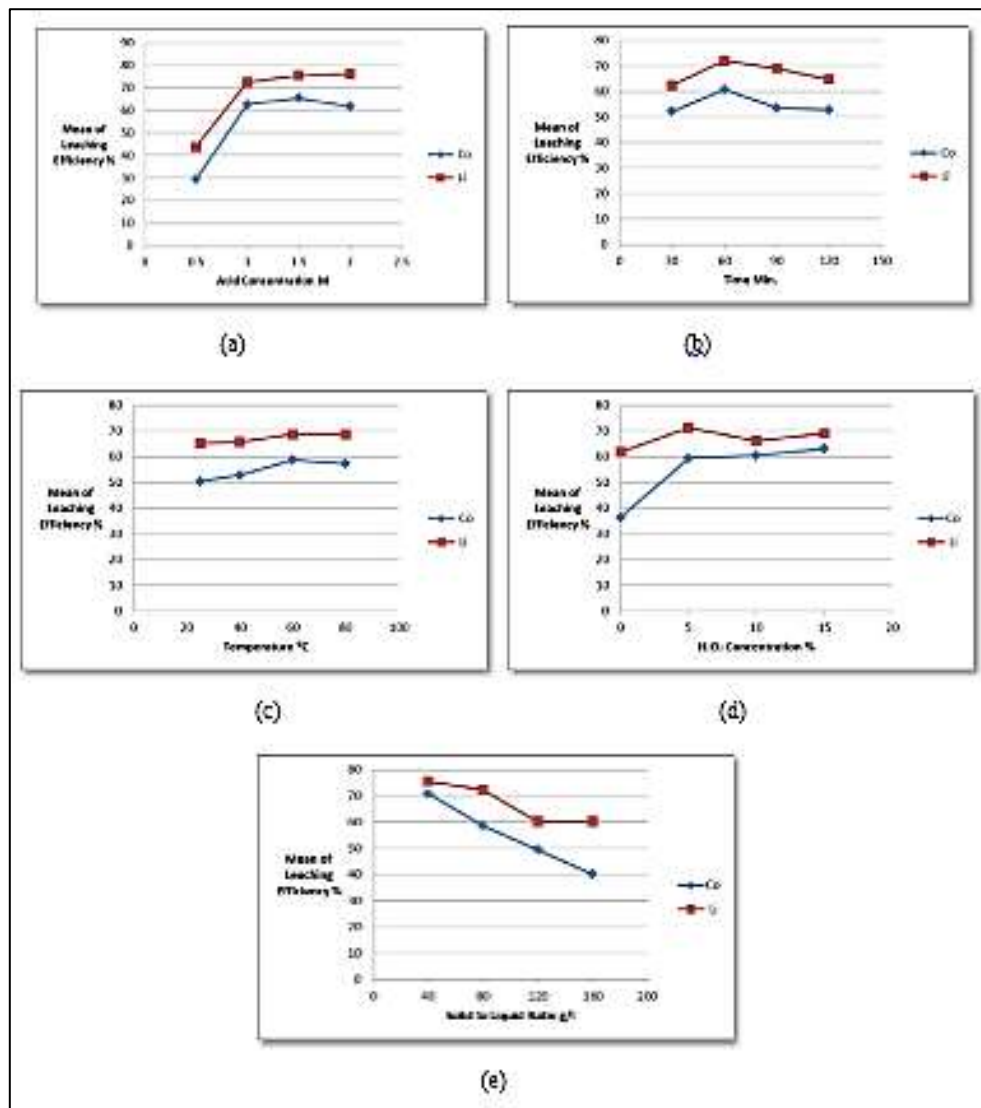


Figure 6: Effect of factors on leaching efficiency (a) acid concentration (b) time (c) temperature (d) H₂O₂ concentration (e) solid to liquid ratio

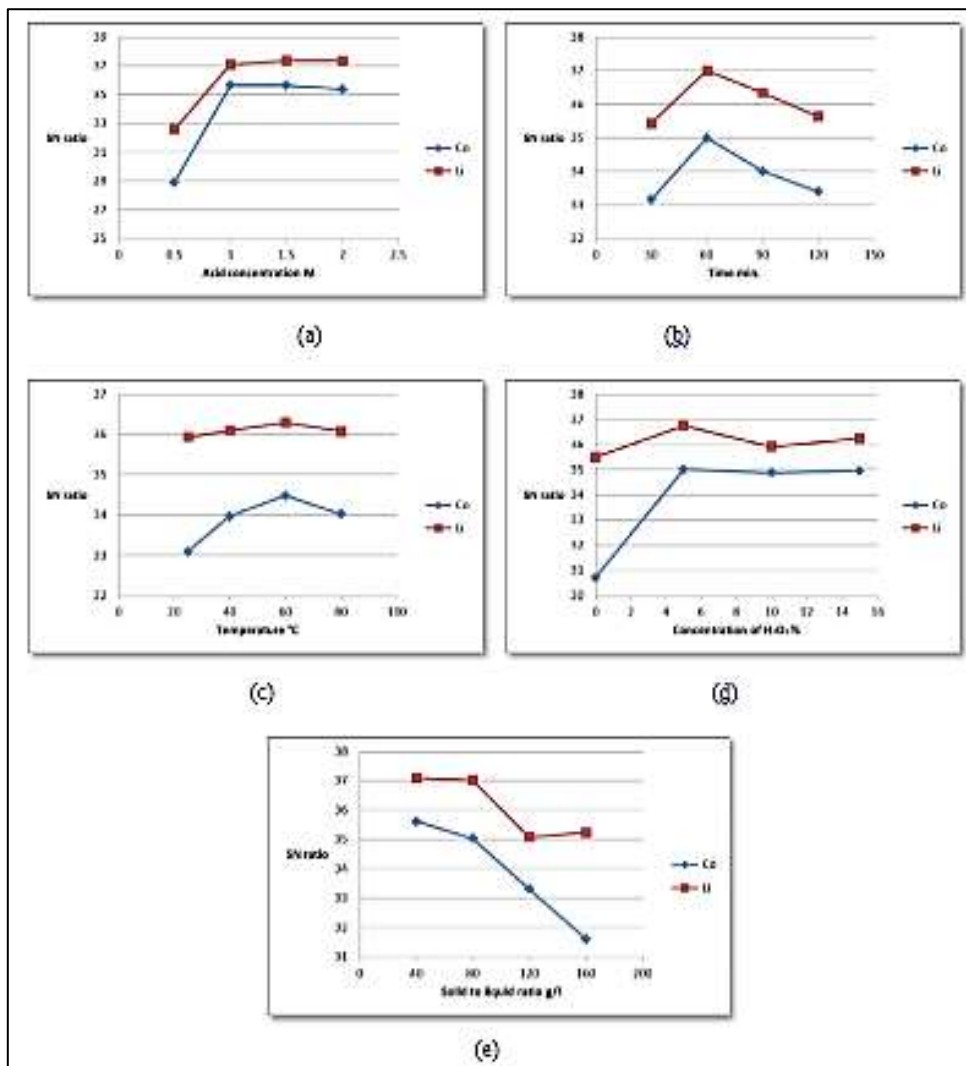


Figure 7: SN ratio of (a) acid concentration (b) time (c) temperature (d) H₂O₂ concentration (e) solid to liquid ratio

Table 5: the optimum leaching experiment results

| No. | Leaching efficiencies of Co % | Leaching efficiencies of Li % |
|------|-------------------------------|-------------------------------|
| 1 | 95.28 | 97.79 |
| 2 | 92.86 | 98.5 |
| Mean | 94.07 | 98.15 |

Table 6: ANOVA for leaching of cobalt

| Source of variance | Degree | Sum of squares | Variance | P % |
|---|--------|----------------|----------|-------|
| Acid concentration | 3 | 132.047 | 44.025 | 55.71 |
| Time | 3 | 8.096 | 2.699 | 3.42 |
| Temperature | 3 | 4.012 | 1.337 | 1.69 |
| H ₂ O ₂ concentration | 3 | 53.66 | 17.887 | 22.64 |
| Solid to liquid ratio | 3 | 39.205 | 13.068 | 16.54 |
| Error | 0 | --- | --- | --- |
| Total | 15 | 237.047 | --- | 100 |

Table 7: ANOVA for leaching of lithium

| Source of variance | Degree | Sum of squares | Variance | P % |
|---|--------|----------------|----------|-------|
| Acid concentration | 3 | 65.71 | 21.9033 | 73.04 |
| Time | 3 | 6.1052 | 2.0351 | 6.79 |
| Temperature | 3 | 0.267 | 0.089 | 0.3 |
| H ₂ O ₂ concentration | 3 | 3.4174 | 1.1391 | 3.8 |
| Solid to liquid ratio | 3 | 14.4617 | 4.8209 | 16.07 |
| Error | 0 | --- | --- | --- |
| Total | 15 | 89.9614 | --- | 100 |

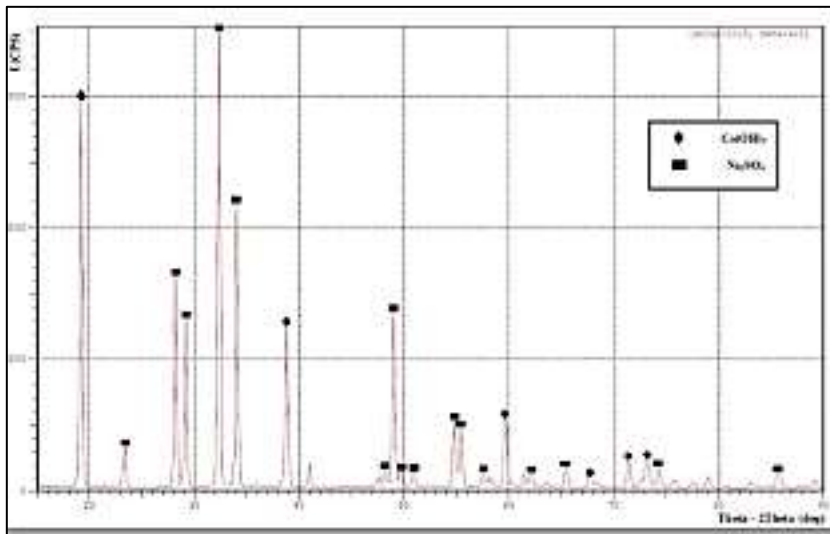


Figure 8: XRD pattern for cobalt precipitate

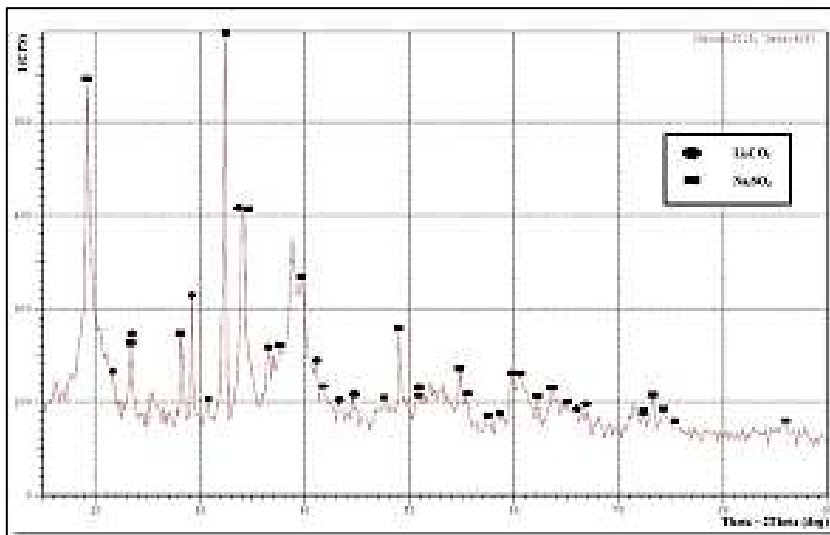


Figure 9: XRD pattern for lithium precipitate

References

[1] L. Li, J. Ge, R. Chen, F. Wu, S. Chen, X. Zhang, "Environmental friendly leaching reagent for cobalt and lithium recovery from spent lithium-ion batteries," *Waste Management*, Vol. 30, 2615–2621, 2010.

[2] L. Chen, X. Tang, Y. Zhang, L. Li, Z. Zeng, and Y. Zhang, "Process for the recovery of cobalt oxalate from spent lithium-ion batteries," *Hydrometallurgy*, Vol. 108, 80–86, 2011.

[3] L. Sun, and K. Qiu, "Vacuum pyrolysis and hydrometallurgical process for the recovery of valuable metals from spent lithium-ion batteries,"

- Journal of Hazardous Materials, Vol. 194, 378–384, 2011.
- [4] G. Jiana, J. Guoa, X. Wanga, C. Suna, Z. Zhoua, L. Yua, F. Kong, and J. Qiua, "Study on separation of cobalt and lithium salts from waste mobile-phone batteries", *Procedia Environmental Sciences*, Vol. 16, 495-499, 2012.
- [5] J. Wang, M. Chen, H. Chen, T. Luo, and Z. Xu, "Leaching study of spent Li-ion batteries", *Procedia Environmental Sciences*, Vol. 16, 443-450, 2012.
- [6] M. Joulié, R. Laucournet, and E. Billy, "Hydrometallurgical process for the recovery of high value metals from spent lithium nickel cobalt aluminum oxide based lithium-ion batteries," *Journal of Power Sources*, Vol. 247, 551-555, 2014.
- [7] F. Pagnanelli, E. Moscardini, G. Granata, S. Cerbelli, L. Agosta, A. Fieramosca, and L. Toro, "Acid reducing leaching of cathodic powder from spent lithium ion batteries: Glucose oxidative pathways and particle area evolution," *Journal of Industrial and Engineering Chemistry*, Vol. 20, 3201–3207, 2014.
- [8] P. Meshram, B.D. Pandey, and T.R. Mankhand, "Recovery of valuable metals from cathodic active material of spent lithium ion batteries: Leaching and kinetic aspects," *Waste Management*, Vol. 45, 306–313, 2015.
- [9] P. Meshram, B.D. Pandey, and T.R. Mankhand, "Hydrometallurgical processing of spent lithium ion batteries (LIBs) in the presence of a reducing agent with emphasis on kinetics of leaching," *Chemical Engineering Journal*, Vol. 281, 418–427, 2015.
- [10] L. Yu, B. Shu, and S. Yao, "Recycling of Cobalt by Liquid Leaching from Waste 18650-Type Lithium-Ion Batteries," *Advances in Chemical Engineering and Science*, Vol. 5, 425-429, 2015.
- [11] L. Li, L. Zhai, X. Zhang, J. Lu, R. Chen, F. Wu, and K. Amine, "Recovery of valuable metals from spent lithium-ion batteries by ultrasonic-assisted leaching process", *Journal of Power Sources*, Vol. 262, 2014, pages 380-385.
- [12] L. Li, J. B. Dunn, X. X. Zhang, L. Gaines, R. J. Chen, F. Wu, K. Amine, "Recovery of metals from spent lithium-ion batteries with organic acids as leaching reagents and environmental assessment," *Journal of Power Sources*, Vol. 233, 180-189, 2013.
- [13] L. Li, J. Ge, F. Wu, R. Chen, S. Chena, and B. Wu, "Recovery of cobalt and lithium from spent lithium ion batteries using organic citric acid as leachant," *Journal of Hazardous Materials* Vol. 176, 288–293, 2010.
- [14] L. Li, J. Lu, Y. Ren, X. X. Zhang, R. J. Chen, F. Wu, and K. Amine, "Ascorbic-acid-assisted recovery of cobalt and lithium from spent Li-ion batteries," *Journal of Power Sources*, Vol. 218, 21-27, 2012.
- [15] K. Provazi, B. A. Campos, D. C. R. Espinosa, and J. A. S. Tenorio, "Metal separation from mixed types of batteries using selective precipitation and liquid-liquid extraction techniques," *Waste Management*, Vol. 31, 59–64. 2011.
- [16] T. Suzuki, T. Nakamura, Y. Inoue, M. Niinae, and J. Shibata, "A hydrometallurgical process for the separation of aluminum, cobalt, copper and lithium in acidic sulfate media," *Separation and Purification Technology*, Vol. 98, 396–401, 2012.
- [17] X. Chen, Y. Chen, T. Zhou, D. Liu, H. Hu, and S. Fan, "Hydrometallurgical recovery of metal values from sulfuric acid leaching liquor of spent lithium-ion batteries," *Waste Management*, Vol. 38, 349–356, 2015.
- [18] J. Nan, D. Han, and X. Zuo, "Recovery of metal values from spent lithium-ion batteries with chemical deposition and solvent extraction," *Journal of Power Sources*, Vol. 152, 278–284, 2005.
- [19] A. K. Jha, M. K. Jha, A. Kumari, S. K. Sahu, V. Kumar, and B. D. Pandey, "Selective separation and recovery of cobalt from leach liquor of discarded Li-ion batteries using thiophosphinic extractant," *Separation and Purification Technology*, Vol. 104, 160-166, 2013.
- [20] L. Sun, and K. Qiu, "Organic oxalate as leachant and precipitant for the recovery of valuable metals from spent lithium-ion batteries," *Waste Management*, Vol. 32, 1575–158, 2012.
- [21] A.A. Nayl, R.A. Elkhatab, S. M. Badawy, and M.A. El-Khateeb, "Acid leaching of mixed spent Li-ion batteries," *Arabian Journal of Chemistry*, 2014. [Online], Available <http://www.sciencedirect.com/science/article/pii/S1878535214000689>
- [22] P. Meshram, B.D. Pandey, T.R. Mankhand, "Extraction of lithium from primary and secondary sources by pre-treatment, leaching and separation: A comprehensive review," *Hydrometallurgy*, Vol. 150, 192–208, 2014.
- [23] J. Li, P. Shi, Z. Wang, Y. Chen, and C. C. Chang, "A combined recovery process of metals in spent lithium-ion batteries," *Chemosphere*, Vol. 77, 1132–1136, 2009.

Author's biography



Nawal Ezzat Abdul Latif, Ph.D. in Metallurgy Engineering, M.Sc. in Metallurgy Engineering, B.Sc. in Production Engineering and Metallurgy, Professor since 2009, Dept. of Production Engineering & Metallurgy, University of Technology Iraq. She is studied Metallurgy engineering in the previous publications and extractive metallurgy in this research. She is also a member in the Iraqi Engineers Union.



Ali Muwafaq Ahmed, M.Sc. in Extractive Metallurgy, B.Sc. in Metallurgy Engineering, Bagdad, Iraq. He is studied extractive metallurgy in this research. He is also a member in the Iraqi Engineers Union

