

Effective Parameters on High-Purity Lithium Carbonate Production from Spent Lithium-Ion Batteries

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ABSTRACT: *The use of lithium-ion batteries in electronic devices is growing rapidly. As a result, the demand for the consumption of lithium metal has increased. Although spent lithium-ion batteries contain sources of precious metals, they seriously threaten human health and the environment. Therefore, the recovery of lithium-ion batteries may prevent environmental pollution. The hydrometallurgy method was applied as the recovery process due to its high recovery efficiency, low energy consumption, and high reaction rate. It is widely used in the recycling process of spent lithium-ion batteries. In this research, instead of all reports concerning synthetic wastewater, industrial wastewater containing lithium was used as feed. Effective parameters on lithium recovery in the form of lithium carbonate and its purity were the initial mass of solution to final mass of solution or concentration ratio, the mole ratio of sodium carbonate to lithium sulfate, raffinate usage, and the cooling effects. Results showed that the optimum condition to achieve maximum purity and recovery of lithium carbonate was obtained at a concentration ratio of 15-20. At different tests with the mole ratio of sodium carbonate to lithium sulfate as 1, 1.5, and 2, the highest recovery efficiency was obtained at the ratio of 1.5. The use of sediment-free raffinate in the last stage also played a big role in lithium recovery. To use the raffinate solution, the raffinate must first be removed from the saturated state of sodium sulfate. Then sodium carbonate becomes saturated in raffinate and is added to the original solution. Under the above conditions, lithium carbonate was obtained with a purity of approximately 99% and a recovery of 65%. The combined process of evaporation with cooling was also a proper process for producing lithium carbonate. In this state, the purity and recovery of the final product were approximately 97% and 75%, respectively.*

KEYWORDS: *Spent lithium-ion batteries; Lithium carbonate; Recovery; Purity.*

INTRODUCTION

Lithium carbonate is used in manufacturing cathodes in lithium-ion batteries, as an additive in ceramic and glass industries, and to treat manias associated with bipolar

disorder [1-6]. It does not exist in nature. Thus, it should be prepared from lithium-ion sources. These sources are classified as primary or natural (such as minerals containing lithium,

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1021-9986/2023/9/2933-2940

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Table 1: The wastewater components in this research

| Element | Quantity (mg/kg or ppm) |
|---------|-------------------------|
| Na | 39768 |
| Li | 645 |
| S | 23646 |
| Al | 86 |
| Ca | 43 |

brines, and seawater) and secondary sources (such as spent lithium-ion batteries). Due to the hazards of lithium-ion batteries to the environment, lithium should be recovered from the batteries [7-13].

Generally, a battery consists of three main parts: cathode, anode, and electrolyte. In a lithium-ion battery, the cathode is a lithium metal oxide compound (such as lithium cobalt oxide (LiCoO₂ or LCO), lithium nickel oxide (LiNiO₂), lithium vanadium oxide (LiV₂O₃), lithium manganese oxides (LiMn₂O₄ or LiMnO), lithium ferrous phosphate (LiFePO₄ or LFP) and a complex compound like LiNi_{0.33}Mn_{0.33}Co_{0.33}O₂). The most common carbon anode is graphite. A separating layer is between the anode and cathode. The electrolyte is made of lithium salt in an organic solvent [14-18].

Pyrometallurgy, biometallurgy, and hydrometallurgy processes are general methods applied to recover lithium from spent lithium-ion batteries [7,9,19,20]. The pyrometallurgy process suffers from the emission of toxic gases. The processing time in the biometallurgy method is high. Therefore, these two processes are not recommended for lithium recovery at industrial scales. Due to the high recovery and purity of the metals, low energy consumption, and low air pollution, the hydrometallurgical process is a good method [7,9,19,21].

Lithium recovery from spent lithium-ion batteries has been studied by various researchers. During the recovery process, the elements such as cobalt, nickel, manganese, and lithium are separated through different steps. For this purpose, the spent batteries are electrically discharged and dismantled. Then they are leached. The obtained wastewater from each stage is used as the feed for another step [19, 21-23].

The wastewater used in this research contains nickel, cobalt, manganese, and lithium. Several methods have been applied to separate the metals [8, 24,2 5]. Due to the use of sulfuric acid, oxalic acid, and sodium hydroxide in the separation of three former metals, the obtained feed

contains high levels of sodium sulfate and sodium oxalate. They cause some difficulties in recovering the low level of lithium sulfate in the wastewater. One of these problems is obtaining an acceptable level of lithium concentration in the wastewater. More concentrations of lithium lower the maximum yield of the lithium carbonate. The precipitation of lithium carbonate needs solution evaporation. However, due to the high concentrations of sodium sulfate, sodium oxalate, and sodium carbonate in the solution, the solution evaporation causes the precipitation of these salts and consequently a reduction in lithium recovery. On the other hand, in the presence of sodium carbonate in the solution, with a gradual increase in lithium sulfate concentration, a percentage of lithium sulfate is converted to lithium carbonate and removed from the process flow before the final precipitation of lithium carbonate [24, 25].

In all published research articles, spent lithium-ion battery wastewaters with high levels of lithium and low levels of other elements have been used. In other words, all researchers used synthetic wastewater. In this research, the industrial spent lithium-ion battery wastewater was used. This wastewater contains high levels of sodium sulfate, sodium oxalate, and sodium carbonate. Lithium will be recovered as lithium carbonate from the wastewater. The effective parameters will be studied to obtain high purity and recovery of lithium carbonate.

EXPERIMENTAL SECTION

Materials

The lithium-ion battery wastewater was prepared by Iran Energy Resources Development Organization. The wastewater composition is given in Table 1.

In order to finally precipitate and convert lithium sulfate to lithium carbonate, sodium carbonate (purity of 99.9 wt%, Merck Company) was used. Sulfuric acid (purity of 95-97 wt%, Merck Company) was consumed. In all stages, deionized water was used for washing.

Experimental procedure

The initial feed was evaporated in several stages to increase the lithium concentration. During the evaporation, the wastewater was stirred by a mechanical stirrer so that the precipitates are not settled. The evaporation was performed in three stages. After each stage, precipitates were washed. The concentrated solution obtained from the three-stage evaporation was heated to a temperature of 90 °C. Based on the examined parameters, a prepared solution

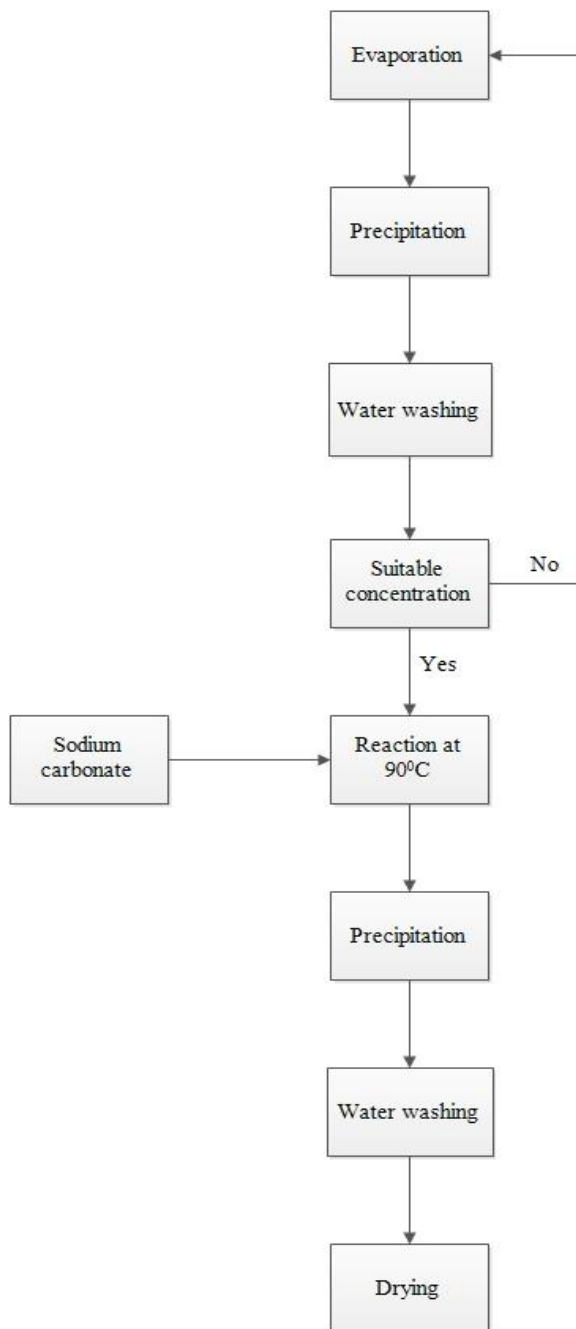


Fig.1: Flow diagram of lithium carbonate production process

of sodium carbonate at the temperature of 90 °C was added to the concentrated solution obtained from the three-step evaporation operation. According to the experiment type, a certain time was considered for the reaction. During the reaction, the solution was stirred at 500 rpm. By adding sodium carbonate solution to a concentrated solution containing lithium, white precipitates of lithium carbonate were observed. The precipitates were separated from

the liquid solution by passing them through a filter paper and washed *via* a hot lithium carbonate solution. The obtained precipitates were heated in an oven at 100 °C for 12 hours and then weighed. The solution passed through the filter was used as a sodium carbonate solvent for use in subsequent experiments. The obtained dried precipitates were analyzed by inductively coupled plasma and X-ray diffraction, and the percentage of salts was calculated. Fig. 1 shows the flow diagram of the lithium carbonate production process.

Analysis

Inductively coupled plasma or ICP (Perkin-Elmer Optima model 5300 DV) was used in the experiments. In ICP, Ar, N₂, and air were used as plasma, purge, and shear gases, respectively. The sample flow rate to ICP was 1.5 cm³/min. The crystal structures of the solid products were also characterized by X-ray diffraction (model RigakuD/Max-RBL) using Cu K α radiation ($\gamma= 1.54060 \text{ \AA}$) with 40 kV of voltage and 30 mA of current.

RESULTS AND DISCUSSION

Preliminary tests showed that the main effective parameters on lithium recovery and its purity are the initial mass of solution to final mass of solution or concentration ratio, the mole ratio of sodium carbonate to lithium sulfate, raffinate usage, and the cooling effects.

Effect of concentration ratio

In the overall process of lithium carbonate production, the initial solution should be significantly concentrated. If the concentration of other salts is insignificant compared to lithium salt, the concentration process is performed by continuous evaporation of the primary solution. Otherwise, with the evaporation of the solution, the salts start to precipitate, and the concentration of lithium salt is still small. In this situation, it is necessary to remove the formed precipitates from the solution in several steps because the solution stirring will be difficult. On the other hand, lithium particles are trapped between the precipitates. Thus, the lithium recovery will decrease. Multi-stage filtration of salts solves this problem to a great extent, but some lithium is removed with precipitates in each stage of filtration. For this reason, there will be an optimal point for the concentration ratio (final mass of the solution containing lithium to the initial mass of the lithium

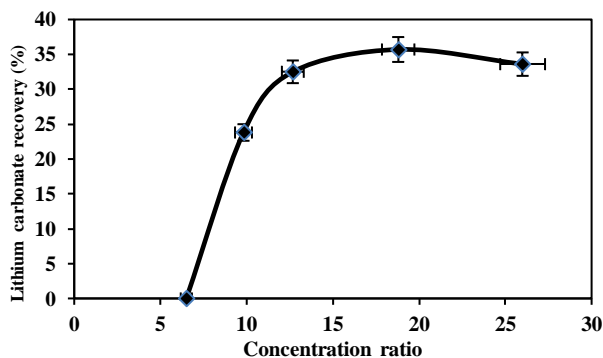


Fig. 2: Lithium carbonate recovery versus concentration ratio

solution). Fig. 2 shows lithium recovery versus the concentration ratio.

As shown, lithium carbonate does not precipitate up to the concentration ratio of 7. By increasing the concentration ratio to nearly 20, the lithium recovery reaches its maximum level and then decreases. The increase in recovery up to a concentration ratio of 20 is due to the formation of more lithium carbonate in the last step (the step of adding sodium carbonate to the lithium sulfate solution). At higher ratios, more precipitates may be separated from the solution during the evaporation stages. Therefore, the solution trapped within the precipitates will increase, and therefore, more lithium will be wasted.

Fig. 3 shows the purity of lithium carbonate obtained at various concentration ratios. As seen, by increasing the concentration ratio up to 10, the purity reaches its maximum and then decreases and remains at a constant value. It should be mentioned that for the tests performed, lithium carbonate precipitates were not washed at the last stage. Thus, purity may be lower than the levels reported in the following sections. At a lower concentration ratio, since the obtained mass of lithium carbonate sediments is small, the amount of solution trapped in the sediments is small. Therefore, the purity has increased. On the other hand, the trapped dissolved sodium sulfate is not saturated. At higher concentration ratios, the mass of lithium carbonate precipitates increases, and the trapped solution is also saturated with sodium sulfate.

It can be concluded that in solutions containing significant amounts of non-lithium salts, the maximum recovery occurs at lower concentration ratios (and lower lithium concentrations). To study other parameters, the concentration ratio between 16 and 20 was selected.

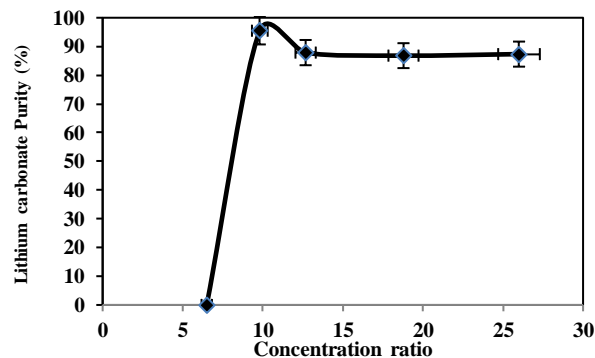


Fig. 3: Lithium carbonate purity versus concentration ratio

Effect of mole ratio of sodium carbonate to lithium sulfate

By adding sodium carbonate to a solution containing lithium sulfate, lithium carbonate precipitates due to its lower solubility in the solution. One of the parameters affecting this phenomenon is the amount of sodium carbonate added to the concentrated solution [25]. Theoretically, the amount of mole of sodium carbonate should be equal to the mole of lithium sulfate in the solution, but in practice, there is no possibility of interaction between all carbonate and lithium ions in the reaction medium. Therefore, it is necessary to add more sodium carbonate than the theoretical amount. On the other hand, adding too much sodium carbonate reduces the purity of the product, which is due to sodium carbonate being trapped within the lithium carbonate crystals. This will increase the operation cost. In this research, the molar ratio of sodium carbonate to lithium sulfate was investigated as 1, 1.5, and 2. Table 2 presents the results obtained for different mole ratios of sodium carbonate to lithium sulfate.

As shown, increasing the ratio from 1 to 1.5 increases the probability of interaction between carbonate and lithium ions and significantly improves recovery. The purity of lithium carbonate has not changed significantly. With a further increase in the ratio, lithium recovery, and lithium carbonate purity have reached 52.3% and 99.07%, respectively. At this ratio, the recovery has slightly increased and the purity also improved slightly, but the increased values are not enough to justify the use of the ratio of 2 for the production of lithium carbonate at an industrial scale due to the related costs. Therefore, using the ratio of 1.5 is suggested both in terms of product quality and reducing costs at the industrial scale.

Table 2: Recovery and purity of lithium carbonate at different molar ratios of sodium carbonate to lithium sulfate

| The molar ratio of sodium carbonate to lithium sulfate | Lithium recovery (%) | Purity of lithium carbonate (%) |
|--|----------------------|---------------------------------|
| 1 | 45.68 | 98.06 |
| 1.5 | 51.70 | 98.85 |
| 2 | 52.30 | 99.07 |

Table 3: Effect of raffinate use on lithium recovery

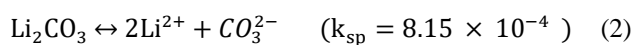
| Use of raffinates | Lithium recovery (%) | Lithium carbonate purity (%) |
|--|----------------------|------------------------------|
| No using the raffinate solution | 49 | 99< |
| Using raffinate as a sodium carbonate solvent | 65 | 99< |
| Adding raffinate to the primary wastewater and evaporation steps | 25 | 99< |

In sub-saturated conditions, the solvated salt exists in its constituent ions. For example, lithium sulfate exists in the form of Li^+ and SO_4^{2-} up to 35 g in 100 g water (the solubility of lithium sulfate at 25 °C) [26]. If the concentration of lithium sulfate exceeds 35 g in 100 g water, the lithium sulfate salt will start precipitating. When sodium carbonate is added to the solution containing lithium sulfate and other components such as sodium sulfate, sodium oxalate, etc., each salt in its sub-saturated conditions exists in its ionic form such as Na^+ , SO_4^{2-} , $\text{C}_2\text{O}_4^{2-}$, and also CO_3^{2-} ions in solution because of sodium carbonate addition.

In these conditions, if the combination of an anion and a cation forms a salt with solubility lower than the other salts, the mentioned salt at its super-saturated conditions starts precipitating. In the case of lithium sulfate, the Li^+ and SO_4^{2-} will form in the wastewater as follows:



By adding sodium carbonate to the wastewater, the Na^+ and CO_3^{2-} will form in solution:



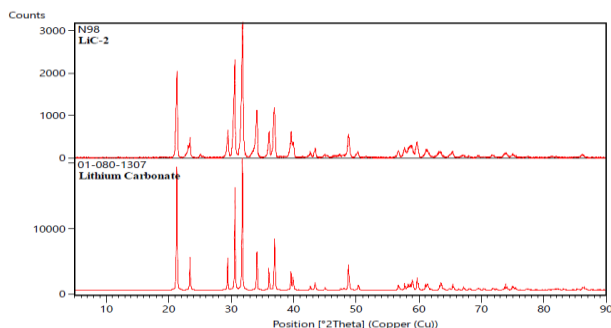
Interaction of Li^+ and CO_3^{2-} will result in the formation of lithium carbonate with lower solubility in water ($k_{\text{sp}} = 8.15 \times 10^{-4}$ and solubility of 1.3 g lithium carbonate in 100 g water at 25 °C [28]) compared to sodium sulfate (solubility of 40 g sodium sulfate in 100 g water at 25 °C [28]) and lithium sulfate (solubility of 35 g lithium sulfate in 100 g water at 25 °C). The lithium carbonate is in super-saturation conditions and starts precipitating. Moreover, other components may affect on the solubility of lithium carbonate or other salts in water.

Effect of raffinate usage

As it was mentioned, in order to produce lithium carbonate from a solution containing lithium sulfate, sodium carbonate was added to the concentrated solution. Because all the lithium sulfate in the solution is usually not converted into lithium carbonate during the precipitation stage, a significant amount of lithium ions enters the solution phase passing through the filter. If the solution containing lithium is not returned to the lithium carbonate production cycle, the efficiency of the process will decrease significantly. Also, if the solution contains significant amounts of non-lithium salts (such as sodium sulfate, sodium oxalate, and sodium carbonate), returning this solution to the production cycle will cause problems. For example, if this solution is returned to the beginning of the process, due to the presence of sodium carbonate, it will gradually produce lithium carbonate during the concentration steps, and the produced lithium carbonate along with sodium sulfate precipitates will leave the process, and the recovery is very difficult. In order to return the lithium ions to the production line, this raffinate solution can be used in the step of adding sodium carbonate to the final concentrated solution to produce lithium carbonate. The presence of sodium carbonate in the raffinate solution causes to decrease in the amount of required fresh sodium carbonate. Also, the available lithium is returned to the process and reduces the need to concentrate the primary solution. On the other hand, by adding fresh sodium carbonate to this solution, tiny crystals of lithium carbonate are formed in small amount, which remains suspended in the solution in colloidal form. These crystals play the role of nucleation when adding the saturated sodium carbonate solution to the concentrated solution obtained from the wastewater and significantly enhance the

Table 4: Effect of cooling on purity and recovery of lithium carbonate

| Process | Lithium recovery in the form of Li_2CO_3 (%) | Purity of Li_2CO_3 (%) |
|-------------------------------------|--|--|
| 3 stage evaporation | 63 | > 99 |
| 2-stage evaporation+1-stage cooling | 73.43 | 97 |

**Fig. 4: XRD patterns of lithium carbonate obtained by using raffinate and then adding sodium carbonate**

Production efficiency of lithium carbonate at this stage. In order to use the raffinate solution, the raffinate must first be removed from the saturated state of sodium sulfate. In other words, firstly, the raffinate enters the cooling system, and the temperature of the solution is reduced to 5 °C. Then, sodium carbonate becomes saturated in the raffinate and is added to the original solution. Table 3 reveals the lithium recovery in different methods of using the raffinate solution.

As shown, if the raffinate is not used, lithium recovery was measured at about 49% due to various ions in the initial solution and the low concentration of lithium-ion. Moreover, if the raffinate is used to prepare saturated sodium carbonate solution, the recovery of lithium increases by 65% due to an increase in the concentration of lithium ions and also the effect of nucleating crystals. Finally, if the raffinate solution is added to the primary wastewater and goes through the evaporation process, a large percentage of lithium will leave the main process as lithium carbonate during the evaporation, and lithium recovery will decrease by 25%.

Fig. 4 illustrates the XRD patterns of lithium carbonate obtained by adding sodium carbonate to the raffinate and then adding it to the concentrated wastewater. As seen, using the method not only increases the yield significantly but also enhances the purity of the produced lithium carbonate.

The cooling effects

Concentrating the initial wastewater (containing

significant sodium sulfate) to obtain the desired lithium level could be performed via a cooling process along with evaporation. In this process, the solubility of sodium sulfate reduces below 30 °C and it precipitates as sodium sulfate decahydrate. In the wastewaters containing high sodium sulfate and sodium oxalate contents, the concentration process leads to achieving the solution saturated with sodium sulfate and sodium oxalate. The precipitates of sodium sulfate and sodium oxalate should be filtered and separated. Lithium is trapped within the crystals in each filtration stage, in turn, lowers the process efficiency. Lowering the temperature causes the removing a portion of sodium sulfate as a precipitate. By increasing the temperature in the evaporation process, the system gets away from saturation, and the lithium concentration will increase through evaporation. It should be considered that in the cooling process, instead of the evaporation process, the obtained solution is not saturated with sodium sulfate. This causes to occur the reaction between lithium sulfate and sodium carbonate in a non-saturated sodium sulfate solution, leading to a significant increase in lithium recovery.

After the cooling stage, some precipitates were observed during the evaporation. It seems that they are lithium carbonate. Sodium carbonate may be added in the previous stages, or sodium oxalate decomposed under high temperatures and forms carbonate salts. To prevent this, by adding sulfuric acid before the evaporation stage, pH of the system was reduced from 11 to 6. The purity and recovery of lithium carbonate are compared between the cooling process and evaporation method (Table 4).

As shown in Table 3, using of cooling step increases recovery by 10% more than evaporation alone. Instead, the purity of the product decreases by 2%.

Table 5 represents the values of lithium carbonate purity and recovery in different research compared to the present study. In spite of the high levels of sodium sulfate and sodium oxalate in wastewater used in the present study, the value of purity is competitive compared to similar studies. In addition, the value of purity may be increased up to 99.9 % by the carbonation of obtained powder with CO_2 gas. On the other side, lithium recovery due to

Table 5: Comparing the results of this work with similar studies

| Process | Purity (%) | Lithium recovery (%) | Ref. |
|---------------------------------|------------|----------------------|------|
| Current study | 99.1 | 65 | - |
| Evaporation and crystallization | 99.8 | 70-72 | [27] |
| Chemical precipitation | 99.6 | 84.5 | [25] |
| Evaporation and crystallization | 99 | 90.1 | [28] |
| Chemical precipitation | 99 | 81 | [29] |
| Evaporation and crystallization | 98 | 80 | [21] |

high levels of sodium sulfate and sodium oxalate is lower than the results obtained by other researchers. In order to increase recovery, alternative methods such as refrigeration can be used which increases the complexity of the process.

CONCLUSIONS

Two targets in the production of lithium carbonate from lithium-ion spent batteries are the purity and recovery of lithium carbonate. The results showed that the main effective parameters on lithium recovery and its purity are the initial mass of solution to final mass of solution or concentration ratio, the mole ratio of sodium carbonate to lithium sulfate, raffinate usage, and the cooling effects. Optimization of the process indicated that for obtaining high purity and recovery of lithium carbonate, the concentration ratio should be between 15 and 20. Washing the sodium sulfate precipitates obtained in each evaporation step with distilled water at 0°C plays a significant role in the lithium recovery efficiency and increases the efficiency. Also, washing the final lithium carbonate precipitates with saturated lithium carbonate solution increases the purity of the final lithium carbonate.

At different tests with the mole ratio of sodium carbonate to lithium sulfate of 1, 1.5, and 2, the highest recovery efficiency was achieved at the ratio of 1.5. Of course, the same yield was obtained at the ratio of 2, but due to the presence of sodium carbonate impurities in the product, the ratio of 1.5 was selected as the optimal ratio.

Since the solution obtained in the last step is saturated with sodium sulfate and the reaction temperature is 90 °C, it is difficult to control the precipitation of the solution at this temperature so that the solution does not evaporate and sodium sulfate precipitates are not observed in the product. The use of sediment-free raffinate in the last stage can also play a big role in lithium recovery because most of the lithium is lost in the last stage and does not react with sodium carbonate. Under

the above conditions, lithium carbonate was obtained with a purity of approximately 99% and a recovery of 65%.

In a combined process of evaporation along with cooling, the only difference is the cooling of the solution resulting from evaporation, which causes the solution to move away from the saturated state of sodium sulfate. In this case, the purity and recovery of the final product were approximately 97% and 75%, respectively.

Acknowledgment

The authors thank Iran Energy Resources Development Organization for financial support.

Received : Nov.20, 2022 ; Accepted : Feb.06, 2023

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