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Research Article

Comparison of waste lithium-ion batteries recycling methods by different decision making techniques

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ABSTRACT

Today, to reduce fossil fuel consumption and to prevent gas emissions that are increasing day by day, vehicles working with electrical energy have started to be produced and developed. The environmental impact of the batteries of electric vehicles, which are increasing in number, is an undeniable fact and is predicted to be a major problem. In this study, three different alternative recycling processes were selected for waste lithium-ion batteries (LIBs), namely pyrometallurgical process, hydrometallurgical process and direct recycling. These processes were compared in terms of their technical, economic, environmental and social aspects using a Multi-Criteria Decision Making (MCDM) approach. From this point of view, in this study, entropy method which is an objective method was used to weight the criteria and Analytic Network Process (ANP) and TOPSIS methods were used to prioritise the alternatives in order to determine the best process for the recycling of waste LIBs. The alternatives were determined as being pyrometallurgical process, hydrometallurgical process and direct recycling, and these alternatives were evaluated in terms of environmental, economic, technical, and social dimensions. Afterwards, sensitivity analysis was performed. The ranking results showed that direct recycling is the best alternative (with values of 0.68 and 0.8101 for ANP and TOPSIS, respectively). In addition, sensitivity analysis was applied for the robustness of the results. As a result of the sensitivity analysis, direct recycling was found to be the best alternative.

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INTRODUCTION

The greenhouse gas emissions caused by the increasing fossil fuel consumption with the industrialization is the most important reason for the global climate change, which has been the problem of the whole world in the last century. Today, 20% of the CO_2 emissions in the European Union countries originate from road transport and a significant amount of fossil fuels are used in road transport [1]. Internal combustion engine vehicles are the cause of a non-negligible rate of fossil fuel consumption [2]. At the Paris Climate Summit held in 2016 with the participation of many countries, the importance of using hybrid and fully electric vehicles (EVs) in reducing global warming was emphasized [3]. In addition, countries committed to

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zero-emission transport at COP27. France and Spain, one of the largest vehicle manufacturers in Europe, have signed the Zero Emissions Vehicle Declaration, aiming for 100% zero emissions in light vehicle and pickup truck sales by 2035 at the latest in leading markets and by 2040 in others [4]. EVs are seen as having zero CO_2 emissions, but to ensure this is true, energy must come from non-fossil fuel sources such as nuclear and alternative energy [5]. EVs, which are increasing in number day by day, are seen as the technology of the future in the world and their use is encouraged in most countries. On the other hand, in parallel with the production and use of these electric vehicles, the production and use of electric vehicle batteries is also increasing. As a result of the studies, it is estimated that by 2030, 140 million EVs will be on the roads worldwide, and with this, 11 million tons of Li-ion battery waste will be generated. Annual waste streams of these batteries are expected to reach 340,000 metric tons by 2040 [6]. Therefore, while aiming to minimize the damage to the environment, these batteries bring with them another problem that causes environmental pollution [7].

Lithium-ion batteries (LIBs), developed in the late 20th century, have led to technological advances in the energy storage and portable electronics and electric vehicle sectors. Compared to other batteries, LIBs stand out thanks to their features such as almost zero memory effect, low self-discharge rate and large power storage capacity with a very high energy density/weight ratio [8]. LIBs generally consist of cathode, anode, separator, electrolyte and casing with sealing function. Lithium ions move along the electrolyte from the negative electrode anode to the positive electrode cathode, during discharge. During charging, the transport of lithium ions is reversed and the ions move from the cathode to the anode. Electrons leave the electrode active material from the current collector, which is a metal with high conductivity, to reach the external circuit. The separator, which allows the electrons to leave the cell and remains between the anode and the cathode, prevents the electrodes from short-circuiting, and also allows the exchange of lithium ions [9]. The schematic representation of the LIB is given in Figure 1.

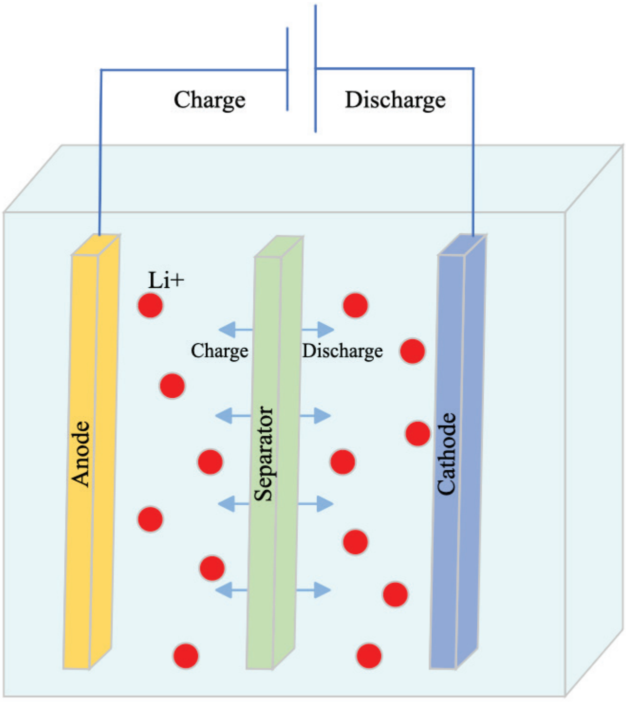

Figure 1. Schematic representation of a LIB [Adapted from 10].

LIBs generally contain transition metal oxides or phosphates, aluminum, copper, graphite, organic electrolytes containing harmful lithium salts, and other chemicals. Therefore, their reuse and recycling processes are very important. In addition, metals such as lithium, cobalt, nickel, copper and aluminum contained in LIBs are very valuable so these waste batteries must be collected and treated appropriately to prevent the disposal of potentially hazardous materials such as cobalt, nickel, manganese, cadmium, lead, etc. [10–12]. In the early 2010s, about 30% of lithium was used for the production of ceramics and glass. In the following years, batteries are thought to cause close to 60% of lithium consumption due to the use of lithium in small electronic devices such as smartphones and laptops and in larger systems such as electric vehicles and energy storage systems [13]. The increase in raw material demands from the EV market is projected to create short-term bottlenecks in lithium and battery-grade nickel supply and long-term excessive copper demand [14]. Lithium is limited as it is not a renewable resource. The supply of lithium has increased with the production of electric vehicles and, accordingly, the production and use of lithium-containing batteries. Only a small amount of used lithium is recycled, and it is thought that lithium shortages may occur if no solution is found to increase lithium recycling [15]. Looking at the production of lithium from raw materials, two hundred and fifty tons of ore spodumene (lithium aluminum silicate) or seven hundred and fifty tons of mineral-rich brine are required to produce just one ton of lithium. Processing raw materials of this scale can also have significant environmental impacts [16,17].

For these reasons above, recovery of metals is of great importance. In 2021, there were two hundred thousand metric tons of EV batteries suitable for recycling. It is predicted that this amount will reach seven million metric tons by 2035 [18]. The recycling of LIBs and the recovery of rare metals are also important for the transition to a circular economy. In the production, use and recycling of LIBs and their materials, circular economy principles are of great importance. Materials containing strategic rare earth elements such as lithium, cobalt and nickel are commonly used in lithium-ion batteries. Efficient use of these materials in a circular economy reduces the resources used and helps to sustain natural resources [19].

At this point, the use of Electrochemical Impedance Spectroscopy (EIS) technique is recommended before recycling process. This provides useful information to optimise the reuse or conversion of batteries for recycling and to minimise the environmental impact of waste batteries. EIS measurements can be used to obtain data on cell performance, to characterise the electrode material and the condition of the cell and to detect damage to the spent LIB. This increases LIBs' contribution to the circular economy and reduces the use of natural resources such as rare metals [20–22]. Thus, these precious metals can be recovered and reintroduced into production processes by recycling end-of-life LIBs [19]. The most commonly used methods for recycling LIBs are pyrometallurgical process, hydrometallurgical process and direct recycling [23–25]. In order to choose the best one among these three methods, it is necessary to consider different aspects such as environmental, economic, technical and social concepts. Taking these concepts into account, MCDM is a widely used and convenient tool for comparing methods.

According to our best knowledge, in the literature, two studies on the comparison of the processes used in the recycling of waste LIBs with Multi-Criteria Decision Making (MCDM) methods were found. The first of these were studied by Sangwan and Jindal (2012) and in this research werev developed an integrated MCDM model to compare different alternatives (disassembly, mechanical conditioning, pyrometallurgy, hydrometallurgy combination (A1), disassembly, mechanical conditioning, hydrometallurgy combination (A2), disassembly, pyrometallurgy, hydrometallurgy combination (A3), pyrometallurgy, hydrometallurgy combination (A4) and pyrometallurgy (A5)) used in the recycling of LIBs. A fuzzy Analytic Hierarchy Process (AHP) was used in the first step to calculate the weights for the different criteria. In the second step, the ranking of the different recycling processes has been calculated by means of fuzzy Technique for Order of Preference by Similarity to Ideal Solutions (TOPSIS). It was found that a combination of pyrometallurgy and hydrometallurgy was the best recycling process for LIBs [26]. In another study, Chakraborty and Saha (2022) compared 9 alternatives created by combining different recycling processes. For the comparison, fermatean fuzzy environment, entropy measure, and aggregation operators based MCDM models were developed and solved. As a result, it was seen that the Alternative 8 (blending of mechanical shredding, electrolyte extraction, electrode dissolution and cobalt electrochemical reduction) gave the best result and the Alternative 3 (blending of dismantling, acid leaching, chemical precipitation and solvent extraction) was the last in the ranking [27].

In the light of the literature studies, it is thought that there is a lack of literature on MCDM studies for the recycling of waste LIBs and more studies should be carried out with different methods. From this point of view, in this study, entropy method which is an objective method was used to weight the criteria and Analytic Network Process (ANP) and TOPSIS methods were used to prioritise the alternatives in order to determine the best process for the recycling of waste LIBs. The alternatives were determined as being pyrometallurgical process, hydrometallurgical process and direct recycling, and these alternatives were evaluated in terms of environmental, economic, technical, and social dimensions. Afterwards, sensitivity analysis was performed.

The rest of the paper is organised as follows: general information about MCDM is given in the materials and methods section and the two selected methods (ANP and TOPSIS) are described. In the same section, the selected recycling alternatives and criteria were examined in detail, and the decision matrix is constructed. Information about sensitivity analysis used to determine the stability of the results is given. The results obtained from two different MCDM methods and sensitivity analyses are presented in the results and discussion section. Finally, the conclusion summarizes the findings and indicates the future research direction.

MATERIALS AND METHODS

MCDM tools are generally used to reach the optimum decision when faced with multiple alternatives with conflicting and unmeasurable decision criteria. This method is widely used in decision making processes in science and engineering. MCDM is based on the decision-maker making a choice between at least two criteria. MCDM has many methods and when a decision maker wants to solve a problem, the first thing to do is to determine the method. Afterwards, it is necessary to create the criteria of the problem and determine the alternatives of the problem. The decision maker evaluates the alternatives based on the criteria and makes the right decision according to the best result among the alternatives [28].

Due to their different mathematical approaches, ANP and TOPSIS were preferred as MCDM methods in this study. Whereas TOPSIS is an objective method for measuring Euclidean distances, ANP is a subjective method based on pairwise comparison. This involved seeing how these different mathematical approaches might influence outcomes. In addition, ANP and TOPSIS are very active areas of MCDM research, and there are several studies that combine these two methods and compare their results [29–31].

The flowchart of the study is given in Figure 2. Firstly, the alternatives and the criteria were determined and decision matrix was performed. The criteria were weighted with the Entropy Method, and then the alternatives were evaluated with ANP and TOPSIS methods.

Determination of Alternatives

In this study, considering the literature studies, the three most commonly used methods, pyrometallurgical process,

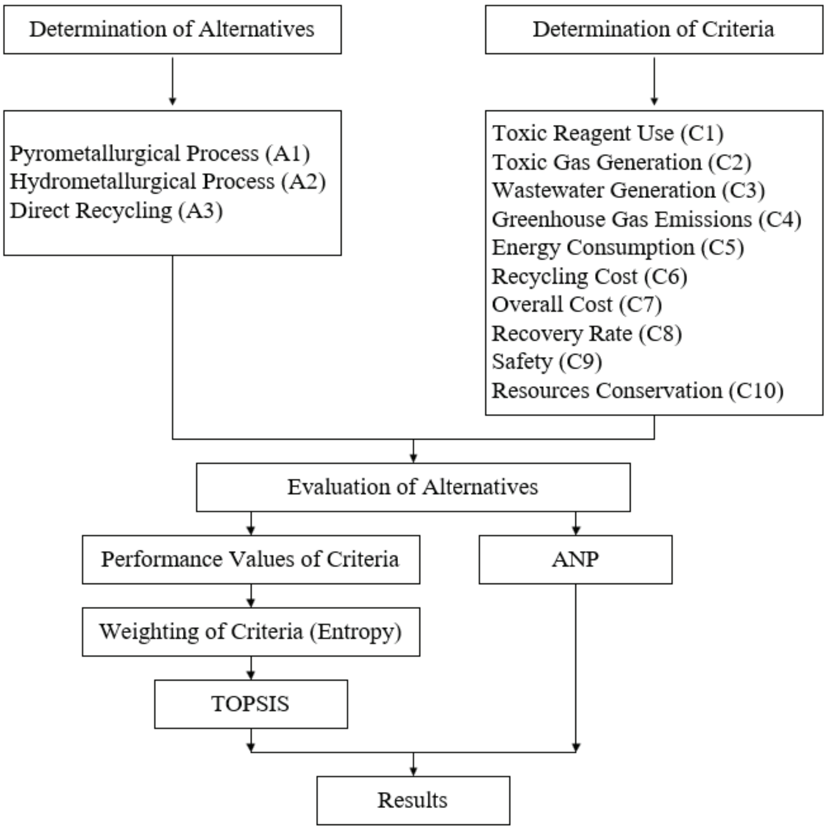

Figure 2. Flow chart.

hydrometallurgical process and direct recycling were selected as alternatives [23–25].

Pyrometallurgical Process (Alternative 1, A1)

The pyrometallurgical process is a high temperature melting process. In this process, LIBs are first incinerated in a foundry where compounds are decomposed and organic materials such as plastics and separators are incinerated. New alloys are then produced through carbon reduction. In the later stages, usually hydrometallurgical, the metal alloys are separated to recover the pure materials. In this process, only expensive metals such as cobalt, nickel and copper can be recovered with high efficiency, while the anode, electrolyte and plastics are oxidized and provide energy for the process. Lithium, aluminum, silicon, calcium and some iron are obtained in the slag phase. Slags can be processed by hydrometallurgical process to obtain pure metals or metal salts. Aluminum, on the other hand, acts as a reducer in the furnace, reducing the need for fuel [32,33]. The reason lithium cannot be recovered by pyrometallurgical processes is that organic materials such as paper, plastic and battery electrolyte burn and lithium remains in the slag. The metals contained in this slag have the potential to leak into the environment after going to storage [34].

Some of the thermal processes used during the pyrometallurgical process are; pyrolysis, melting, distillation and refining [35]. *Pyrolysis* is the decomposition of organic material by an intensive application of heat in the absence of oxygen. This process can be used to neutralize batteries and eliminate electrolytes as well as organic materials such as plastic and paper. In vacuum pyrolysis, the heating process is carried out in a vacuum in order to lower the boiling temperature and prevent secondary chemical reactions from occurring. *Melting* uses heat and chemical reduction to obtain metal, leaving slag and gases behind. *Distillation* can be used to thermally separate metals. Metals are evaporated at different temperatures and then condensed. *Distillation* can also be carried out using a vacuum. Since the decrease in pressure also reduces the evaporation temperature, there is no need for very high temperatures. In addition, thermal processes can be used to refine metals to high purity, eliminating unwanted materials [34].

During the pyrometallurgical process, a large amount of energy is consumed due to the operating temperature (~1500 °C) [36].

Figure 3 shows the flow chart of the pyrometallurgical recycling process.

The advantages of pyrometallurgical processes are as follows:

- It consists of simple operations.
- There is no need for operations such as separation and size reduction.

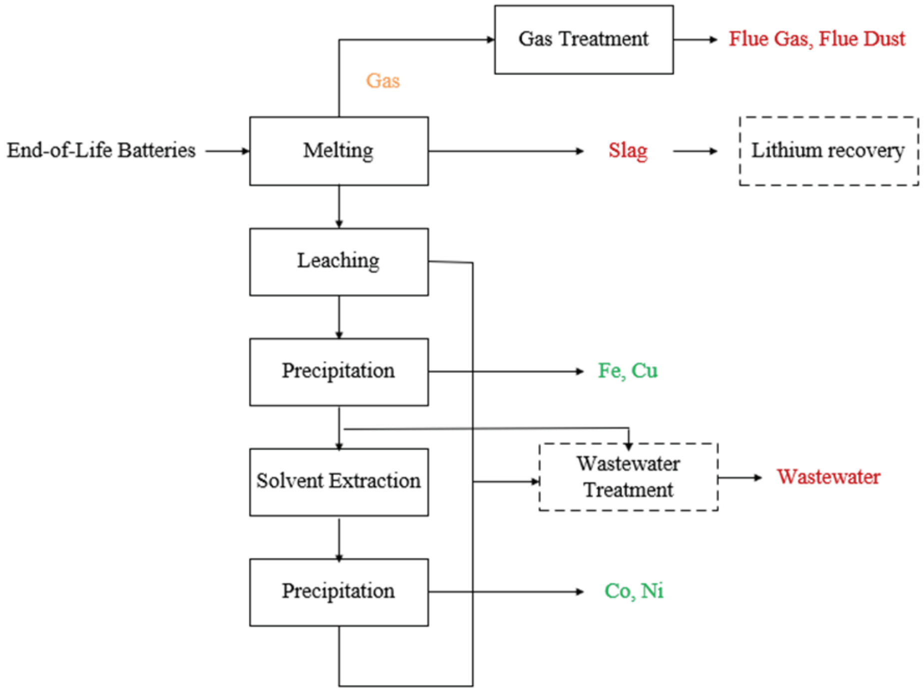

Figure 3. Flow chart of the pyrometallurgical recycling process [37].

(Green color represents products, orange color represents by-products and red color represents wastes and the dashed areas represent optional processes).

- Outputs that can be used in the production of new cathode materials are formed as a result of the process.
- The disadvantages of pyrometallurgical processes are as follows:
- CO_2 is produced during the melting process and a high amount of energy is consumed.
- The alloy requires extensive processing and is therefore costly.
- Most materials such as plastic, graphite and aluminum cannot be recovered [32].

After this process, harmful emissions may occur, including carbon dioxide, carbon monoxide, sulfur dioxide, volatile organic compounds and dust from scrap metals [34].

Hydrometallurgical Process (Alternative 2, A2)

Aqueous chemistry is used in the hydrometallurgical process. This process is carried out by leaching in acids or bases and then by concentrating and purifying [32]. This process is performed to recover LIBs after a pre-treatment [38]. Mechanical processes applied to batteries, such as shredding and dismantling, are part of the hydrometallurgical process. After these processes applied to the batteries, an acid solution is used to separate the elements. Even if the liquid solution can be used almost directly for the production of Ni-Co sulfates, elements such as lithium or copper may be lost. Shredding and disassembly can also cause material loss, and safety problems may arise depending on the amount of charge present in the battery [39]. For LIBs, ions in solution are separated by processes such as ion exchange, solvent extraction, chemical precipitation, electrolysis and precipitated as different compounds [32].

Figure 4 shows the flow chart of the hydrometallurgical recycling process.

The advantages of hydrometallurgical processes are:

- High purity materials may occur.
- Many of the LIB components are recoverable.
- The process is carried out at low temperature.
- Compared to the pyrometallurgical process, lower CO_2 emissions occur.
- The disadvantages of hydrometallurgical processes are:
- Cost increases as separation is required in this process.
- Since elements such as cobalt, nickel, manganese, iron, copper and aluminum in solution have similar properties, it is difficult to separate them and requires high cost.
- The cost is increasing to treat the resulting wastewater [32].

In hydrometallurgy, cobalt, lithium, manganese, nickel and, if present, graphite can be recovered [40]. Hydrometallurgical and pyrometallurgical processes can also be used together. With the pyrometallurgical process, the safety problems that may arise from the different chemical compositions, structures and charge states of the batteries are eliminated. In the hydrometallurgical process, the

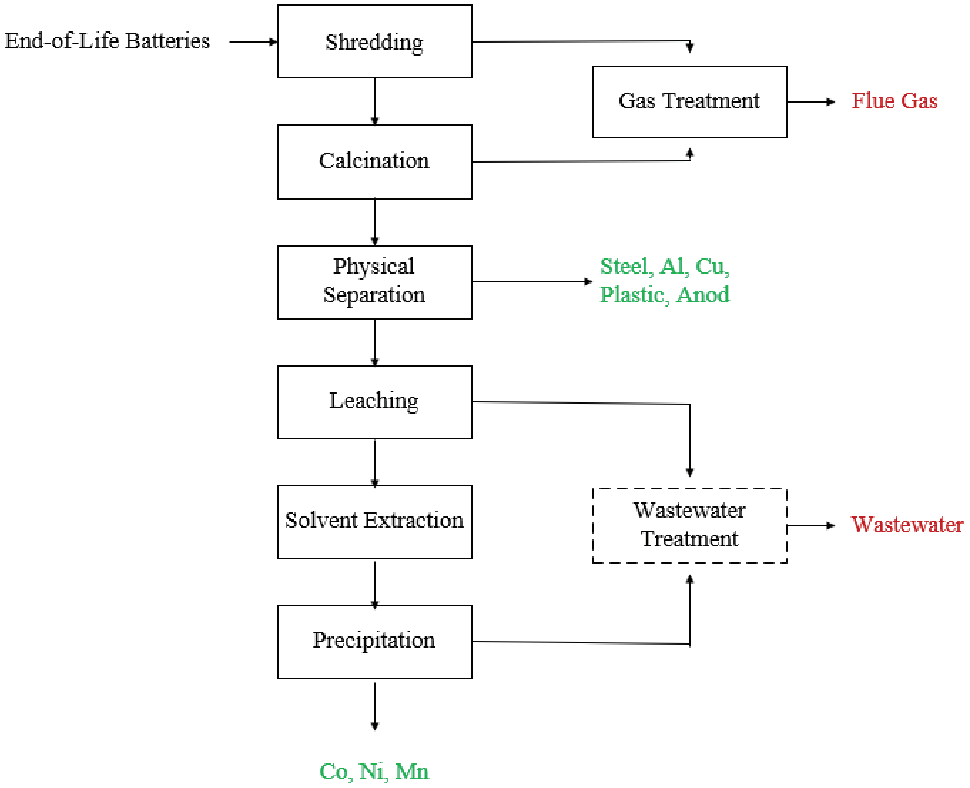

Figure 4. Flow chart of the hydrometallurgical recycling process [37].

(Green color represents products, red color represents wastes and the dashed areas represent optional processes).

separation and processing of the materials recovered in the slag is carried out with the help of different types of acids and chemicals [39].

Direct Recycling (Alternative 3, A3)

Direct recycling aims to recover the cathode material without any chemical change in the structure of the recovered material and to produce new batteries by renewing them. It is a physical recovery method in which processes such as separation by using gravity and magnetic separation are in question [9].

Significant energy inputs are needed for extensive processing of material recovered in pyrometallurgical and hydrometallurgical processes [41]. With the direct recycling method, in principle, the wastes produced during recycling are minimized and the cathode materials are recovered as reusable cathode mixtures instead of individual metals [42].

The direct cathode recycling process, similar to hydrometallurgical processes, begins with an evacuation and disassembly step, in which the external cell equipment can be individually removed and recycled. Direct cathode recycling involves removing the electrolyte using liquid or supercritical CO_2, then reducing the size of the recovered components and separating the cathode materials [41].

In this process, first of all, end-of-life LIBs are discharged and disassembled until they reach the cell level. They are then treated with supercritical CO_2, which can extract the electrolyte. After reducing the temperature and pressure, the CO_2 is removed from the electrolyte and this electrolyte can be reused for the manufacture of new batteries. Cells that do not contain electrolytes are separated and broken down. The cell components are then separated using physical methods, and the cathode materials can be brought together and reused in new batteries. Direct cathode recycling, which saves and regenerates powder cathode material for use in subsequent batteries, draws attention due to its low energy consumption and high recovery rate [43].

Figure 5 shows the flow chart of the direct recycling process.

High temperature, strong acid leaching and extensive gas purification are required for pyrometallurgical and hydrometallurgical recycling processes. This results in high costs, high energy consumption, water pollution and recycled materials with low resale value. Since cathode materials account for 30-40% of the total cost, a cost-effective and environmentally friendly direct recycling process providing reusable cathode materials can reduce energy consumption and cost of battery materials [44], [45]. Considering

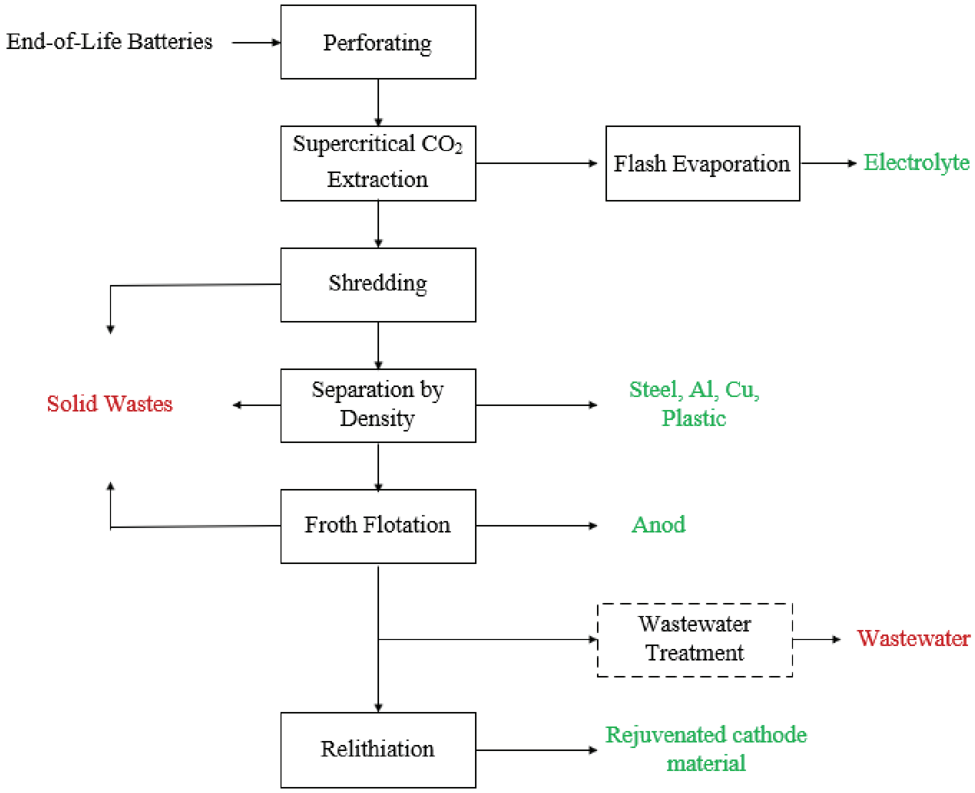

Figure 5. Flow chart of the direct recycling process [37].

(Green color represents products, red color represents wastes and the dashed areas represent optional processes).

all recycling processes, no recycling process can recover all materials and a small amount of waste has to be sent to landfill [34].

A comparison of the methods is given in Table 1.

Determination of Criteria

As a result of the literature survey, ten criteria have been defined in environmental, economic, technical, and social dimensions. These criteria and their explanations are given in Table 2.

Performance Values of Criteria

Considering the following explanations for the criteria, the decision matrix was created and then the mentioned methods were applied.

(C1) Toxic reagent use: In this criterion, the scoring was determined as 2 for pyrometallurgy, 8 for hydrometallurgy, and 4 for the direct recycling method. Although the hydrometallurgical process promises high efficiency for recycling, strong, dangerous and environmentally harmful acids are used in this process. In this method, preference of organic acids instead of dangerous inorganic acids is one of the important focus points [46]. In the direct recycling method, toxic solvents and acids are rarely used. In direct recycling, most methods that focus on re-functionalizing the active material are based on high-temperature treatments [9].

(C2) Toxic gas generation: Scoring made in this criterion was determined as 8 for pyrometallurgy, 4 for hydrometallurgy, and 2 for direct recycling [34].

(C3) Wastewater generation: In this criterion, the scoring was determined as 3 for pyrometallurgy, 8 for hydrometallurgy, and 2 for the direct recycling method. Wastes resulting from hydrometallurgical processes; the leaching stage is water and chemicals from co-precipitation and washing. In order to reduce or eliminate the generated wastewater and related costs, research on wastewater treatment, water reuse or reducing the amount of water in the process is still ongoing [32]. In the hydrometallurgical process, difficulties in recovery of low pH leaching and leaching of metals such as Al, Cu and Fe, formation of harmful by-products such as Cl_2, SO_x and NO_x and the resulting wastewater treatment are considered as important problems [6]. In the last stage of this process, wastewater is formed in the solvent extraction method used for separation [46].

Table 1. A comparison of the methods [9,37,38]

	Pyrometallurgical Process	**Hydrometallurgical Process**	**Direct Recycling**
Type of the Method	**Chemical**	**Chemical**	**Physically**
Recovered Materials	Copper compounds Iron compounds Co^{2+} in output Ni^{2+} in output Lithium compounds Aggregate (from slag)	Copper Steel Aluminum Graphite Plastics Lithium carbonate Co^{2+} in output Ni^{2+} in output Mn^{2+} in output Electrolyte solvents Electrolyte salts	Copper Steel Aluminum Graphite Plastics LCO NMC (111) NMC (622) NMC (811) NCA LMO LFP Electrolyte solvents Electrolyte salts
Advantages	High recycling rates Solvent free Simple operation	High recycling rates High purity product formation A wide variety of metals are recovered Low energy consumption Less waste gas	Environmentally friendly High specificity Low energy consumption High recovery rate Reduction in recovery costs
Disadvantages	High temperatures are needed May need other operations to effectively recover materials Li and Mn are not recovered More toxic gas generation and toxic gas treatment costs High energy consumption	Complex process Usage of toxic reagents Costly operation Excess wastewater generation Long processes	Not specific Does not allow simultaneous processing of different cathode materials High operational and equipment requirements are needed

LCO: Lithium cobalt oxide, NMC: Lithium nickel manganese cobalt oxide, NCA: Lithium nickel cobalt aluminum oxide, LMO: Lithium manganese oxide, LFP: Lithium iron phosphate

Table 2. Used criteria

Criteria No	Criteria Group	Criteria Name	Unit	Preference
C1	Environmental	Toxic Reagent Use	1-9	Decreasing
C2		Toxic Gas Generation	1-9	Decreasing
C3		Wastewater Generation	1-9	Decreasing
C4		Greenhouse Gas Emissions	kg/kg	Decreasing
C5		Energy Consumption	MJ/kg	Decreasing
C6	Economic	Recycling Cost	\$/kg	Decreasing
C7		Overall Cost	\$/kg	Decreasing
C8	Technical	Recovery Rate	1-9	Increasing
C9		Safety	1-9	Increasing
C10	Social	Resources Conservation	1-9	Increasing

(C4) Greenhouse gas emissions: Greenhouse gases originate from the smelting process in the pyrometallurgy process. In the hydrometallurgical process, the upstream production of chemicals contributes significantly to total greenhouse gas emissions. Based on 1 kg of end-of-life LIB, 2.21 kg of greenhouse gas is emitted for the pyrometallurgy process and 2.27 kg for the hydrometallurgy process. For the direct recycling method, this value is only 0.5 kg, and since this value is significantly lower than other methods, the direct recycling method has the potential to reduce emissions and be economically competitive [46]. According to a study, the direct recycling method has the potential to reduce greenhouse gas emissions from cathode material recovery by 81-98% and SOx emissions by 72%-100% [9].

(C5) Energy consumption: For end-of-life LIBs (NMC), the energy consumption per kg for the direct recycling method is about 4.2 MJ, which is only 25% and 13.8% of the pyrometallurgical and hydrometallurgical processes, respectively. Lighter processing conditions are a big factor in the direct recycling method [46]. According to a study, energy consumption per kg of $LiCoO_2$ is 108 MJ for pyrometallurgy, 89 MJ for hydrometallurgy and 91 MJ for direct recycling. The energy consumption for $LiCoO_2$ production per kg without any recycling process is approximately 151 MJ [25]. The average values were calculated as 108 MJ and 16.8 MJ for the pyrometallurgy process, 30.4 MJ and 89 MJ for the hydrometallurgy process, and 4.2 MJ and 91 MJ for the direct recycling method. The energy consumption values for pyrometallurgy, hydrometallurgy and direct recycling were determined as 62.4 MJ, 59.7 MJ, 47.6 MJ, respectively.

(C6) Recycling cost: Recycling costs are set at \$2.9, \$2.2 and \$1.6 per kg for pyrometallurgy, hydrometallurgy and direct recycling, respectively. The difference in recycling costs between the three methods is small because the costs include not only energy and material input, but also operating labor, maintenance and repair, and laboratory costs [46]. The final step of the hydrometallurgical process involves the separation and purification of the separated and filtered metallic components of the LIBs. Common processes such as solvent extraction, chemical precipitation and electrochemical deposition are used for separation. The solvent extraction process is widely used because of its ion selectivity advantages and the high extraction efficiency (>95%) offered by the many available extractants. The disadvantage of this process is the high upfront cost of extractors and the waste treatment cost, given the large volume processes [47].

(C7) Overall cost: Overall costs are \$0.5, \$0.3 and \$0.4 for pyrometallurgy, hydrometallurgy and direct recycling, respectively. Overall costs include administrative costs, distribution and selling costs, and R&D costs, and there is no significant difference between these three recycling methods. Although the costs of the three recycling methods are close to each other, the direct recycling method generates the highest profit compared to the hydrometallurgical and pyrometallurgical process because new LIBs can be produced directly with this method [46].

(C8) Recovery rate: Scoring was made based on the rate of material that pyrometallurgy, hydrometallurgy and direct recycling methods could recover, and this scoring was determined as 3 for pyrometallurgy, 5 for hydrometallurgy, and 8 for direct recycling. Dai et al. [37] showed material recovery efficiency for different recycling methods in their study. Looking at the values given in their study for plastics and electrolyte solvents, a material recovery efficiency of 50% is assumed as there may be less incentives for recycling compared to higher values of cobalt and nickel or metals with generally more stable demands. Besides, due to the lack of data on the direct recycling method, it is assumed that this method has the same material recovery efficiency (excluding the cathode materials) as the other two recycling methods. The recovery efficiency for cathode materials is assumed to be 90%, given the difficulties associated with separating the cathode material from the rest of the battery components.

(C9) Safety: In this criterion, the scoring was determined as 4 for pyrometallurgy, 2 for hydrometallurgy, and 7 for the direct recycling method. In the pyrometallurgical process, there is little safety risk in this process, as the cells and modules are all exposed to extreme temperatures with a reductant for metal reclamation [48]. In the pyrometallurgical process, there are different heating stages of the furnace during combustion. Thanks to the slow heating of waste batteries, the risk of explosion is reduced. In the hydrometallurgical process, there may be material loss due to shredding and dismantling, and safety problems may arise depending on the amount of charge present in the battery. Explosions may occur if batteries are not discharged before they rupture [39].

(C10) Resources conservation: In this criterion, the rate of recycled material and the amount of energy consumed were taken as a basis and scoring was made accordingly. This scoring was determined as 3 for pyrometallurgy, 5 for hydrometallurgy, and 8 for direct recycling.

In the application of the entropy and TOPSIS methods, the decision matrix was first created (Table 3).

Weighting of Criteria (Entropy Method)

The Entropy Method, also known as Shannon's Entropy method, is one of the objective weighting methods that is based on completely unbiased data and can overcome the shortcomings of subjective weighting methods. The Entropy Method, which is versatile and efficient, eliminates human-induced problems and gives results that are more in line with the facts [49]. One of the reasons why this method is suitable for use in MCDM problems is that it allows calculating the importance weights of the criteria without resorting to personal judgments and considerations [50]. Since the entropy weight indicates the degree of useful information, it can be concluded that the criterion with the larger entropy weight is more important in terms of decision making/evaluation [51].

Table 3. Decision matrix

	C1	C2	C3	C4	C5	C6	C7	C8	C9	C10
A1	2	8	3	2.21	62.4	2.9	0.5	3	4	3
A2	8	4	8	2.27	59.7	2.2	0.3	5	2	5
A3	4	2	2	0.5	47.6	1.6	0.4	8	7	8

The Entropy Method consists of the following steps:

Step 1: A decision matrix (Equation 1) is made, where the rows consist of alternatives and the columns consist of criteria. This situation is as follows for the decision matrix K:

$$K = \begin{bmatrix} b_{11} & \cdots & b_{1n} \\ \vdots & \ddots & \vdots \\ b_{m1} & \cdots & b_{mn} \end{bmatrix} m \text{ x } n \qquad (1)$$

Step 2: In order to get rid of the differentiation in units, the decision matrix is normalized and the Equation 2 is used to normalize the criteria:

$$S_{ij} = \frac{b_{ij}}{\sum_{j=1}^{m} b_{ij}} \quad j = 1, 2, \ldots, m; \; i = 1, 2, \ldots, n \qquad (2)$$

Step 3: Entropy values (ed_j) are calculated according to Equation 3:

$$ed_j = -k \sum_{j=1}^{m} S_{ij} \ln S_{ij} \qquad (3)$$

k = entropy constant = $(lnm)^{-1}$

$0 \leq ed_j \leq 1$ and $s_{ij} lns_{ij} = 0$ if $s_{ij} = 0$

However, ed_j values increase as the information transmitted by the j_{th} criterion decreases.

Step 4: The degree of differentiation (Equation 4) is calculated for each criterion:

$$fd_j = 1 - ed_j \qquad (4)$$

Step 5: The weight value (Equation 5), which is the degree of importance, is found for the i_{th} criterion:

$$ad_j = \frac{fd_j}{\sum_{i=1}^{n} fd_i} \quad j=1, 2, \ldots, m \qquad (5)$$

Looking at the above equation, it can be stated that criteria with large entropy weights are more important [52].

Evaluating of Alternatives (Analytical Network Process (ANP))

ANP is one of the widely used multi-criteria decision-making methods to solve various real-world problems due to its ability to consider the interrelated and complex relationships between decision elements and its ability to simultaneously apply qualitative and quantitative attributes. ANP provides a network of relationships between criteria that leads to more reliable results. Calculates complex relationships between decision elements by replacing a hierarchical structure with a network structure [53]. A decision problem analyzed with ANP is usually examined through a hierarchy or network of controls for benefits, costs, opportunities and risks. ANP uses the same basic comparison scale (1-9) (Table 4). This comparison scale enables the decision maker to intuitively combine experience and knowledge and show how many times an item dominates another item according to the criteria [54].

Step 1: In this step, the problem is defined and a decision model is created. The purpose, criteria, sub-criteria and alternatives related to the problem are clearly stated.

Step 2: The relationships between the criteria and sub-criteria of the problem are determined.

Step 3: Priority vectors are calculated from pairwise comparisons between criteria.

Step 4: Consistency analyzes of the comparison matrices are performed. To determine whether the comparisons are consistent, the Consistency Ratio (CR) must be calculated for each matrix after the comparison matrices have been created. The CR is obtained by dividing the Consistency Index by the Randomized Consistency Index. If the CR value is less than 0.10, it can be said that the pairwise comparisons are consistent. If values are greater than 0.10, there is inconsistency in the comparison. In this case, the comparisons should be repeated.

Step 5: The supermatrix is created. A new matrix is created by multiplying all the values in the unweighted supermatrix and the weights of the set. This matrix can be expressed as a weighted super matrix. All columns of the matrix are the same, and each gives the relative priorities of the elements in each set, with the priorities of the elements

Table 4. Comparison scales used in ANP [55]

Importance Level	Definition	Explanation
1	Equally Important	It contributes equally
3	Moderately Important	One option is slightly preferable than the other
5	Strongly Important	One option is strongly favored over the other
7	Very Strongly Important	One option is strongly favored over the other, and its superiority is clearly evident
9	Extremely Important	Evidence in favoring one option over another has the highest possible degree of validation
2, 4, 6, 8	For compromise between the above values	They are used when there is no good word to define for compromise. They represent the average values that can be given.

normalized to one. The supermatrix is taken to a power large enough to equalize the priorities at some point. The resulting matrix is called the limit supermatrix.

Step 6: The best alternative is chosen. With the resulting limit supermatrix, the importance weights for each criterion are determined. The best alternative in the problem of choice is the one that is the most important alternative and has the highest importance in the decision process.

The Super Decision program was used and "benefit-opportunity-risk-cost" analysis and "benefit-cost-risk (BCR)" analysis was performed to evaluate the recycling methods of LIBs. In order to achieve this, benefit and opportunity clusters were combined [56].

The following formulation is used in the program:

Formula: bB + oO + c (1/C) + r (1/R)

Here, firstly, evaluations were made within each cluster and the weights of each cluster were used as r = 1/2, c = 1/3, b = 1/6 and o = 0 to reach the result.

In Figure 6, BCR model for ANP are shown.

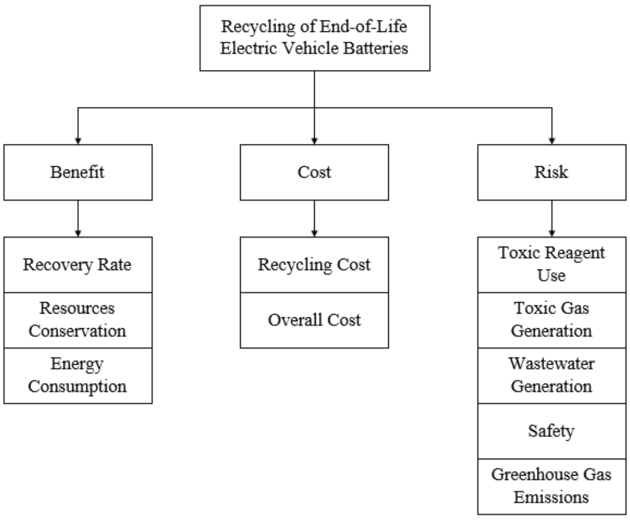

Figure 6. Benefit-cost-risk cluster criteria.

Evaluating of Alternatives (TOPSIS)

TOPSIS, which was first developed by Hwang and Yoon (1981), was developed based on the idea that the chosen alternative should have the shortest distance from the positive ideal solution and the farthest distance from the negative ideal solution to solve the multi-criteria decision-making problem. However, the best alternative should not only have the shortest distance to the positive ideal solution, but also the longest distance to the negative ideal solution. In short, the positive ideal solution consists of all the best achievable values of the criteria, while the negative ideal solution consists of all the worst achievable values of the criteria [57].

The steps used to implement TOPSIS are as follows:

Step 1: A normalized decision matrix (Equation 6) of useful and non-useful criteria is created.

$$r_{ij} = \frac{xij}{\sqrt{\sum_{j=1}^{J} xij^2}}, \; j = 1, 2, 3, \ldots., J; \; i = 1, 2, 3, \ldots., n \qquad (6)$$

x_{ij} = original score of the decision matrix
r_{ij} = normalized score of the decision matrix

Step 2: A weighted normalized decision matrix is created by multiplying the w_i weights of the evaluation criteria with the normalized decision matrix r_{ij} (Equation 7).

$$v_{ij} = w_{ij} * r_{ij}, \; j = 1, 2, 3, \ldots\ldots, J, \; i = 1, 2, 3, \ldots\ldots, n \qquad (7)$$

Step 3: The positive ideal solution (Equation 8) and the negative ideal solution (Equation 9) are determined.

$$A^+ = \{v^*_1, v^*_2, \ldots\ldots, v^*_n\} \text{ Maximum values} \qquad (8)$$

$$v^+_i = \{\max (v_{ij}) \text{ if; } j \in J; \min (v_{ij}) \text{ if; } j \in J^-\}$$
$$A^- = \{v^-_1, v^-_2, \ldots, v^-_n\} \text{ Minimum values}$$

$$v^- = \{\min (v_{ij}) \text{ if; } j \in J; \max (v_{ij}) \text{ if; } j \in J^-\} \qquad (9)$$

Step 4: The separation measures of each alternative from the positive ideal solution (Equation 10) and the negative ideal solution (Equation 11) are calculated.

$$s_i^+ = \sqrt{\sum_{j=1}^{n} \left(v_{ij} - v_j^*\right)}, \; j=1, 2, \ldots, J \qquad (10)$$

$$s_i^- = \sqrt{\sum_{j=1}^{n} \left(v_{ij} - v_j^-\right)}, \; i=1, 2, \ldots\ldots, J \qquad (11)$$

Step 5: The closeness coefficient (Equation 12) is calculated according to the ideal solution of each alternative.

$$CC_i = \frac{d_i^-}{d_i^* + d_i^-}, \; i=1, 2, \ldots, J \qquad (12)$$

Step 6: Alternatives are ranked from the most valuable to the least valuable according to the decreasing values of the closeness coefficient. The alternative with the highest closeness coefficient (CC_i) is selected [58].

Microsoft Office Excel program was used to evaluate with TOPSIS method. The entropy values (w_j) of the criteria weighted by the Entropy Method were used in the implementation of TOPSIS.

Sensitivity Analysis

Sensitivity analysis examines the effect of changing the coefficient values determined in a MCDM on the optimal solution of the problem. The decision maker can make better decisions if they can determine how critical each criterion is, in other words, how sensitive the current ranking of alternatives is to changes in the weights of the criteria [59,60].

In this study, a sensitivity analysis was conducted to determine the stability of the results. It is done to examine whether changing the weight of a criterion causes a change in the priority order of alternatives. Sensitivity analysis was performed with a decision matrix created with modified importance level values (Table 5).

RESULTS AND DISCUSSION

The sum of the weights of the criteria determined for all three alternatives was taken and the calculation was made by dividing each criterion by this weight sum, and thus a normalized decision matrix was obtained. Then, the entropy values of the criteria were found and the degree of differentiation of the information (d_j) was calculated. Finally, the entropy criterion weights (w_j) were calculated (Table 6).

According to the weighting processes made by using the Entropy Method, it has been seen that the most important evaluation criterion is wastewater generation. Greenhouse gas emissions (C4), toxic reagent use (C1) and toxic gas generation (C2), safety (C9), resource conservation (C10) and recovery rate (C8), recycling cost (C6), overall cost (C7) and energy consumption (C5) followed the wastewater generation criteria, respectively.

The criteria weights obtained by using the Entropy Method were transferred to the BCR model by normalizing for each cluster. Synthesis command in the main menu is used in ANP, and general results are obtained by performing synthesis in the highest-level network. The results obtained from ANP are shown in Table 7.

It is seen that the most useful criterion for the benefit cluster is the “Direct Recycling” Method, which has a very high difference compared to the other two alternatives. Looking at the cost cluster, “Pyrometallurgical Process” is the costlier alternative. In the results of the risk cluster, “Hydrometallurgical Process” is determined as the most risky alternative. According to the ANP synthesis results, the best alternative was “Direct Recycling (A3)”.

Table 8 shows the positive ideal and negative ideal separation measures, the relative closeness to the ideal solution, and the ranking results for TOPSIS. According to Table 8, the “Direct Recycling (A3)” alternative is in the first place in order of priority. It is followed by “Pyrometallurgical Process (A1)” and “Hydrometallurgical Process (A2)”, respectively.

As mentioned before a sensitivity analysis study was conducted to determine the stability of the results. Sensitivity analyses were performed for six linguistic criteria (1-9 scaling). These criteria are toxic reagent use (C1), toxic gas generation (C2), wastewater generation (C3), recovery rate (C8) safety (C9), and resource conservation (C10). The weight values used for the sensitivity analysis are given in Table 9. From Tables 10 and 11 it can be observed that direct recycling is the most favourable alternative for

Table 5. Decision matrix for sensitivity analysis

	C1	C2	C3	C4	C5	C6	C7	C8	C9	C10
A1	**3**	**6**	**4**	2.21	62.4	2.9	0.5	**2**	**6**	**2**
A2	**5**	**5**	**6**	2.27	59.7	2.2	0.3	**4**	**3**	**6**
A3	**7**	**3**	**3**	0.5	47.6	1.6	0.4	**7**	**8**	**7**

Changed values are shown in bold characters

Table 6. Entropy criterion weights (w_j) values

	C1	C2	C3	C4	C5	C6	C7	C8	C9	C10
wj	0.1539	0.1539	0.1868	0.1606	0.0072	0.0307	0.0227	0.0804	0.1234	0.0804

Table 7. ANP results

	Benefit	Cost	Risk	Overall	Ranking
A1	0.15	0.43	0.34	0.22	2
A2	0.35	0.30	0.40	0.10	3
A3	0.5	0.27	0.26	0.68	1

Table 8. TOPSIS results

	Si+	Si-	Ci+	Ranking
A1	0.1483	0.1511	0.5047	**2**
A2	0.2039	0.0790	0.2792	**3**
A3	0.0503	0.2147	0.8101	**1**

(Si$^+$ is the distance from the ideal solution, Si$^-$ is the distance from the negative ideal solution and Ci$^+$ is the relative closeness to the ideal solution.)

Table 9. Entropy criterion weights values for sensitivity analysis

C1	C2	C3	C4	C5	C6	C7	C8	C9	C10
0.0869	0.0596	0.0644	0.2363	0.0105	0.0451	0.0333	0.1816	0.1112	0.1707

Table 10. Sensitivity results of benefit, cost and risk clusters

	Benefit	Cost	Risk	Sensitivity Results	Sensitivity Ranking
A1	0.11	0.43	0.36	0.26	2
A2	0.25	0.31	0.53	0.32	3
A3	0.64	0.26	0.11	0.42	1

Table 11. Sensitivity results for TOPSIS

	Si+	Si-	Ci+	Ranking
A1	0,1854	0,0601	0,2448	**3**
A2	0,1593	0,1129	0,4148	**2**
A3	0,1084	0,1820	0,6267	**1**

the recycling of waste LIBs, even with the change of the weights of the criteria. As a result of the sensitivity analysis, a change in the overall ranking can be observed with the pyrometallurgical process ranked second and the hydrometallurgical process ranked third.

The results obtained for sensitivity analysis from ANP are shown in Table 10. The same rankings were obtained in the sensitivity analysis results as in the ANP results. “Direct Recycling” Method is the most useful criterion for the benefit cluster, “Pyrometallurgical Process” is the costlier alternative and “Hydrometallurgical Process” is determined as the most risky alternative.

When the general results are examined, it is seen that “Direct Recycling” is the best alternative. According to the sensitivity analysis results for TOPSIS, “Direct Recycling (A3)” is again the best alternative for recycling of LIBs (Table 11).

CONCLUSIONS

Most of the research studies on end-of-life LIBs focus on recycling methods that reduce environmental pollution and try to prevent natural resource consumption, thus contributing to environmental sustainability and circular economy. In this study, three different recycling methods were compared via MCDM. Firstly, the criteria were weighted with the Entropy Method, and then alternatives were evaluated with two different methods, ANP and TOPSIS. According to the weighting processes made by using the Entropy Method, it has been seen that the most important evaluation criteria are wastewater generation and greenhouse gas emissions, respectively. These criteria are followed by the use of toxic reagents with the same weight and the formation of toxic gas. According to the ANP overall results, it was seen that the best alternative was Direct Recycling. Direct Recycling Method is followed by Pyrometallurgical Process and Hydrometallurgical Process, respectively. In TOPSIS, positive ideal and negative ideal separation measures, relative closeness to the ideal solution and ranking results were obtained. In order of priority, the Direct Recycling alternative is in the first place followed by Pyrometallurgical Process and Hydrometallurgical Process, respectively. This is because the Direct Recycling alternative has more advantages in environmental management for end-of-life LIBs than other alternatives. As a result of the sensitivity analysis, Direct Recycling was found to be the best alternative for the recycling of LIBs in both ANP and TOPSIS methods. Direct recycling, designed to recover cathode material with morphological integrity. By reducing the number of processing steps required to re-synthesize cathode materials, this process has a comparatively low environmental impact. A main drawback of direct recycling is that the process is dependent on the input of specific cathode types for the recovery of high value materials. So, it is highly dependent on an efficient classification of battery types based on easy to understand labelling according to cell chemistry (Recycling of Lithium-Ion Batteries—Current State of the Art, Circular Economy, and Next Generation Recycling). For future studies, it is recommended that studies with criteria different from those used in this paper can be conducted in the future using different MCDM methods, such as Elimination and Choice Translating Reality (ELECTRE) and Preference Ranking Organisation Method for Enrichment Evaluations (PROMETHEE), among others. In addition, it is possible to develop the approach here with other methods such as Life Cycle Assessment and Life Cycle Cost Analysis. In this way, more accurate and more detailed information can be obtained to compensate for the limitations of this study.

CONFLICT OF INTEREST

The authors declare no potential conflicts of interest regarding the research, authorship and/or publication of this article.

DATA AVAILABILITY

The data used to support the findings of this study are included within the article.

AUTHOR'S CONTRIBUTIONS

All authors are contributed equally to bring out this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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