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Original Theoretical Research

Method for Recycling Lithium-Ion Batteries with the Extraction of Valuable Components

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Abstract

Introduction. Due to the increasing demand for lithium-ion batteries, it has become a pressing issue to find an environmentally friendly and safe way to dispose of old batteries. The life cycle of these batteries is shorter than that of the equipment they power, which leads to a growing amount of waste. This waste poses a serious problem for disposal and can have harmful effects on the environment. At the same time, recycling spent lithium-ion batteries offers a solution. By extracting valuable components we can return these components to the production process and create a closed-loop system. In this regard, the aim of this study is to investigate the methods of recycling lithium-ion batteries and to analyze the proposed method for their disposal, which involves extracting valuable components such as Li2CO3, while introducing the principles of a closed-loop economy into the production process.

Materials and Methods. The methods of systematizing scientific literature on lithium-ion battery recycling were used. The "Mpr_Dipl" software was used to select the most promising method, which includes direct decision-making, paired comparison, and weighted sum methods. A technological process for lithium-ion batteries processing was developed using the COMPASS-3D software.

Results. As a result of the analysis, the advantages and disadvantages of each lithium-ion recycling method were highlighted. A hydrochemical method was selected using the multi-criteria decision-making method. A five-stage process for lithium ion battery processing with lithium carbonate extraction was developed, including grinding, separation, filtration, precipitation, and wet Li2CO3 capture. The material balance for the developed method was calculated.

Discussion and Conclusion. The developed recycling system ensures safe recycling of used lithium-ion batteries with minimal negative environmental impact and maximum recovery of valuable components. These results can be used to optimize the recycling process and maximize the extraction of valuable materials from spent lithium-ion batteries for further sale as lithium carbonate, thereby generating additional revenue.

Keywords: lithium-ion battery, recycling, disposal, circular economy, hydrometallurgy

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Оригинальное теоретическое исследование

Способ утилизации литий-ионных аккумуляторов с извлечением ценных компонентов

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Аннотация

Введение. В связи с постоянно растущей потребностью в литий-ионных аккумуляторах (ЛИА) и увеличением количества уже используемых накопительных устройств актуальной темой на сегодняшний день является создание экологичного, безопасного и дешевого способа их утилизации. Жизненный цикл литий-ионных аккумуляторов меньше, чем оборудования, где они применяются, поэтому возрастает риск образования большого количества отходов, которые могут привести к серьезным проблемам с утилизацией и пагубному воздействию на окружающую среду. В то же время отработанные литий-ионные аккумуляторы можно использовать вторично, извлекая из них ценные компоненты для возвращения в производственный цикл. В связи с этим целью данной работы является исследование методов утилизации литий-ионных аккумуляторов и анализ предложенного авторами способа их утилизации с извлечением ценных компонентов (Li_2CO_3) при внедрении принципов экономики замкнутого цикла в производство.

Материалы и методы. Авторами использовались методы систематизации научной литературы по проблематике утилизации литий-ионных аккумуляторов. Для выбора наиболее перспективного из них была использована программа Mpr_Dipl. В ней заложены прямые методы принятия решений, метод парных сравнений и метод взвешенной суммы. Разработка технологической схемы процесса переработки ЛИА проводилась в программе «КОМПАС-3D».

Результаты исследования. В результате анализа были выделены достоинства и недостатки каждого метода утилизации литий-ионных аккумуляторов, а также выбран гидрохимический способ с использованием методики решения задач с многокритериальным выбором. Предложена технологическая схема процесса переработки литий-ионных аккумуляторов с извлечением карбоната лития, состоящая из пяти стадий: измельчение, разделение, фильтрация, осаждение и вылавливание влажного осадка Li_2CO_3. Рассчитан материальный баланс разработанного способа утилизации.

Обсуждение и заключение. Разработанная авторами система утилизации обеспечивает безопасную переработку отработавших литий-ионных аккумуляторов при минимальном негативном воздействии на окружающую среду и максимальном выделении ценных компонентов. Результаты исследования могут быть использованы для модернизации процесса утилизации литий-ионных аккумуляторов с целью извлечения дополнительной прибыли от продажи карбоната лития.

Ключевые слова: литий-ионный аккумулятор, переработка, утилизация, экономика замкнутого цикла, гидрометаллургия

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Introduction. Global energy consumption is increasing every year due to population growth, economic development, and technological progress. Scientists are currently researching the development and efficient use of renewable energy sources (RES), such as wind, solar, hydropower, and tidal power [1]. However, most of these renewable energy sources have an irregular nature, requiring the use of storage devices to ensure a consistent supply of energy from these sources [2]. One solution to this challenge is the use of lithium-ion batteries. Every year, the demand for lithium-ion batteries continues to grow and is expected to continue growing in the near future. This is due to the development of new materials and improvements in production processes. The growth in demand for LIBs is supported by expert estimates. According to Fortune business insight forecasts, the market for lithium-ion batteries will reach $193 billion in volume by 2028 (Fig. 1).

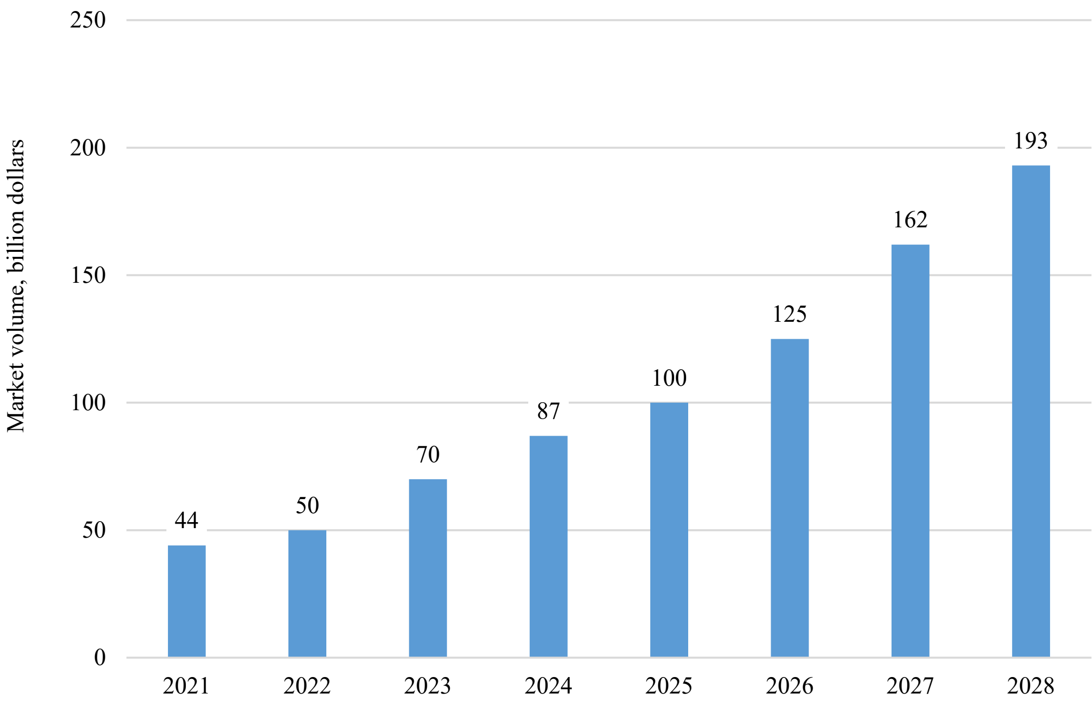

Fig. 1. Volume and forecast of the dynamics of the global lithium-ion battery market, billion US dollars

To date, there are a limited number of LIB production plants in Russia. The largest domestic enterprises producing lithium-ion batteries include:

– Aspil Energy (Pyatigorsk);

– Saturn (Krasnoyarsk);

– Uralelement (Chelyabinsk);

– Scientific Research Aeroinstitute Istochnik OAO (Saint Petersburg).

It is important to note that battery components are imported from China and Bolivia [3].

Due to the active use of LIBs, a pressing issue today is how to create an environmentally friendly, safe, and cost-effective method for disposing of used batteries. The life cycle of lithium-ion batteries depends on the linear model or linear economy in which batteries are produced, used, and then disposed of. According to predictions [4], approximately 95% of lithium-ion batteries produced worldwide remain unprocessed and end up in households, which is an inefficient approach that leads to a significant amount of waste and high material costs for disposal [5]. To address this issue, it is essential to implement closed-loop economic practices and utilize LIB with the extraction of valuable materials [6]. The aim of this research is to develop a recycling system for lithium-ion batteries that allows for the extraction of valuable components (lithium carbonate).

Materials and Methods. An analytical analysis of the methods of disposal of lithium-ion batteries has been conducted. The analysis was based on domestic and foreign research. To identify the most promising approach, we used the method of problem-solving with a multi-criteria selection using the Mpr_Dipl software.

We calculated the material balance of the system based on [7].

The process sheet was created using the COMPASS-3D program.

Research Results

1. Analysis of existing methods for LIB processing. Recycling methods are a potential solution for extending the lifespan of lithium-ion batteries in the economic cycle [8]. This is an integral part of a closed-loop economy, as it facilitates the internal circulation of materials and reduces the depletion of resources associated with primary production [9].

LIB recycling is crucial for minimizing the environmental impact of mining and waste disposal, while also conserving resources and reducing costs associated with manufacturing new batteries.

According to Figure 2, the most common recycling methods for lithium-ion batteries include physical, pyrometallurgical, and hydrochemical processes [10].

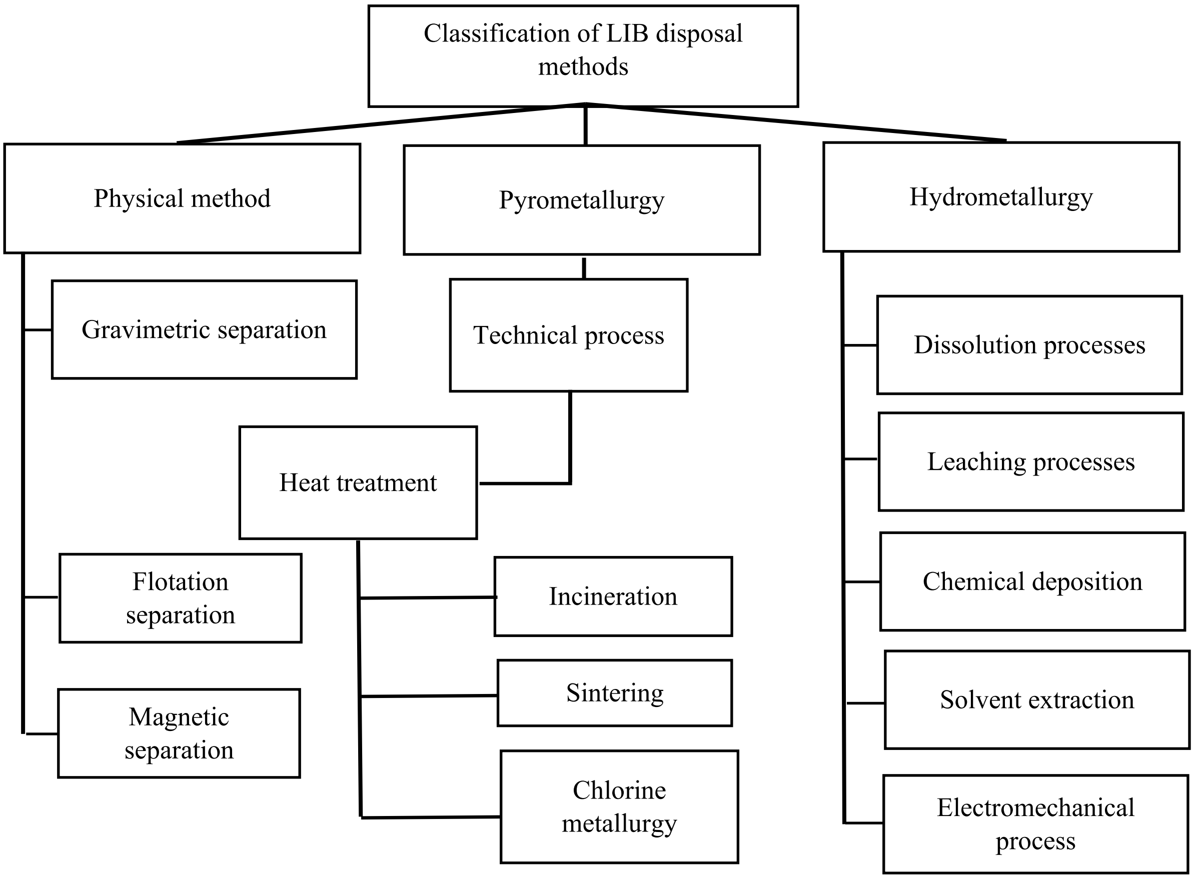

Fig. 2. Classification of LIB disposal methods

Physical method involves disassembling batteries using various techniques to separate the battery components based on their physical properties, such as density and magnetism. The main techniques used in this method include gravimetric separation, flotation separation, and magnetic separation. This method is one of the most popular ways of recycling lithium-ion batteries due to its low cost and environmentally friendly nature. However, it is important to note that physical processing techniques may not always be as effective at extracting all valuable components as other methods.

Pyrometallurgy is a well-known and reliable method that involves processing materials, often allowing for the extraction of a high percentage of valuable metals, such as cobalt and nickel, from battery waste. This process requires a significant amount of energy, but it is generally reliable and does not require specific settings for processing materials with a particular composition, which makes it suitable for materials with varying compositions, such as electronic and battery waste [11].

Pyrometallurgical process consists of three stages: metal reduction, pyrolysis, and gas combustion. During pyrolysis, the organic components of lithium-ion batteries are thermally degraded. As a result, toxic flue gases are produced, which can harm the technological process [12].

The main commercial companies involved in the pyrometallurgical processing of lithium-ion batteries are Sumitomo-Sony in Japan and Umicore AG & Co. KG in Belgium [13].

The hydrometallurgical process involves the use of aqueous solutions to extract the necessary metals from cathode material [14]. In the hydrometallurgical process, the metal extraction rate is high with lower energy costs and no toxic emissions into the atmosphere. The essence of the process is that the crushed material is treated with acid or alkali to dissolve metals. Then the resulting solution is purified and the metals are extracted. Due to the complex structure of the cells, the initial stage is grinding, followed by leaching and mechanical separation phases, which include ferromagnetism. The separation of carbon from metal oxide can also be carried out by foam flotation [15].

The main advantages of hydrometallurgy are the use of inexpensive reagents, low environmental impact, low operating costs, high occupational safety, and the possibility of industrial scale.

2. Selection of the most efficient method for LIB disposal. To select the most promising and logically sound technology for lithium-ion battery disposal, a task with a multi-criteria choice was developed. A solution was found that met the most important criteria for effective disposal using a special software program.

To solve this problem, we needed input data: alternatives — in our case, these were the disposal methods themselves. The evaluation criteria were ranked on a scale from 1 to 5: “Possibility of integration into the production process”, “Labor/energy intensity of the process”, “Level of recovery of materials”, “Process safety”, “Formation of by-products” where 1 was considered as the most favorable outcome, and 5 — the least favorable, respectively.

The main selection criterion for this task was the “Possibility of integration into the production process”, since the fundamental solution would be the use of the resources of the main production line, and assigning weight coefficients to the criteria was as follows:

“Possibility of integration into the production process” — 0.4;

“Labor/energy intensity of the process” — 0.15;

“Level of recovery of materials” — 0.2;

“Process safety” — 0.1;

“Formation of by-products” — 0.15;

Table 1 presents the ranking criteria for all three disposal methods.

Table 1

Data for the multi-criteria selection task

Criteria	Processing method		
	Physical	Pyrometallurgical	Hydrometallurgical
Labor/energy intensity of the process	2	3	4
Possibility of integration into the production process	3	5	2
Level of recovery of materials	4	5	2
Process safety	4	2	3
Formation of by-products	2	5	3

To determine the most suitable alternative, we used the Mpr_Dipl program. This program contains methods of decision-making in multi-criteria tasks: direct methods (characterized by the characteristic dependence of the usefulness of an alternative on its ratings according to some special criteria), methods of paired comparisons (criteria are ordered by importance, after which the best alternative is considered to have a higher score according to a more important criterion, regardless of the ratings according to other criteria.), the weighted sum method (methods of decision-making under conditions of certainty and under conditions of uncertainty). This program was used by the authors of article [16] to select a method for recycling sunflower husks. Figure 3 provides the calculation results.

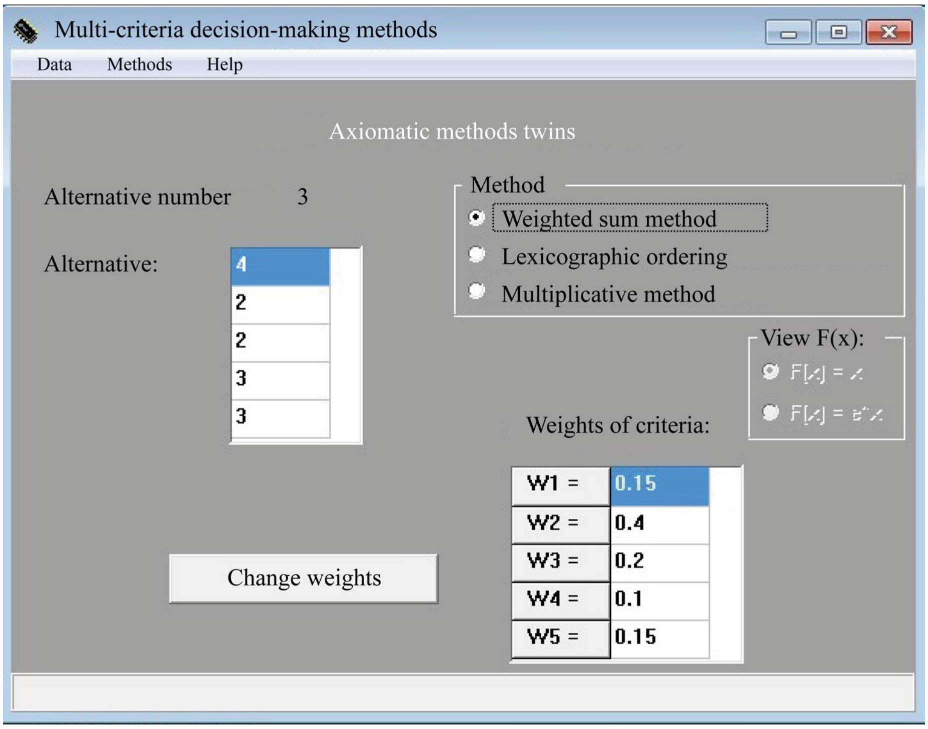

Fig. 3. Calculation results of the Mpr_Dipl program

Thus, the most suitable method of lithium-ion batteries recycling in this case was the "Hydrochemical method" (Alternative No. 3).

3. Technology for spent LIB recycling using a hydrochemical method. Based on the results of analyzing various technologies for lithium-ion battery processing, it has been decided to use a hydrochemical method involving the extraction of lithium carbonate in several stages. Lithium carbonate is a valuable product with wide application in metallurgy. It is used for steel desulfurization, as well as in pyrotechnics, the production of glasses and plastics, electrical insulating porcelain, sitals, and in agriculture as a fertilizer and feed additive. Figure 4 shows the technological scheme for the LIB processing process.

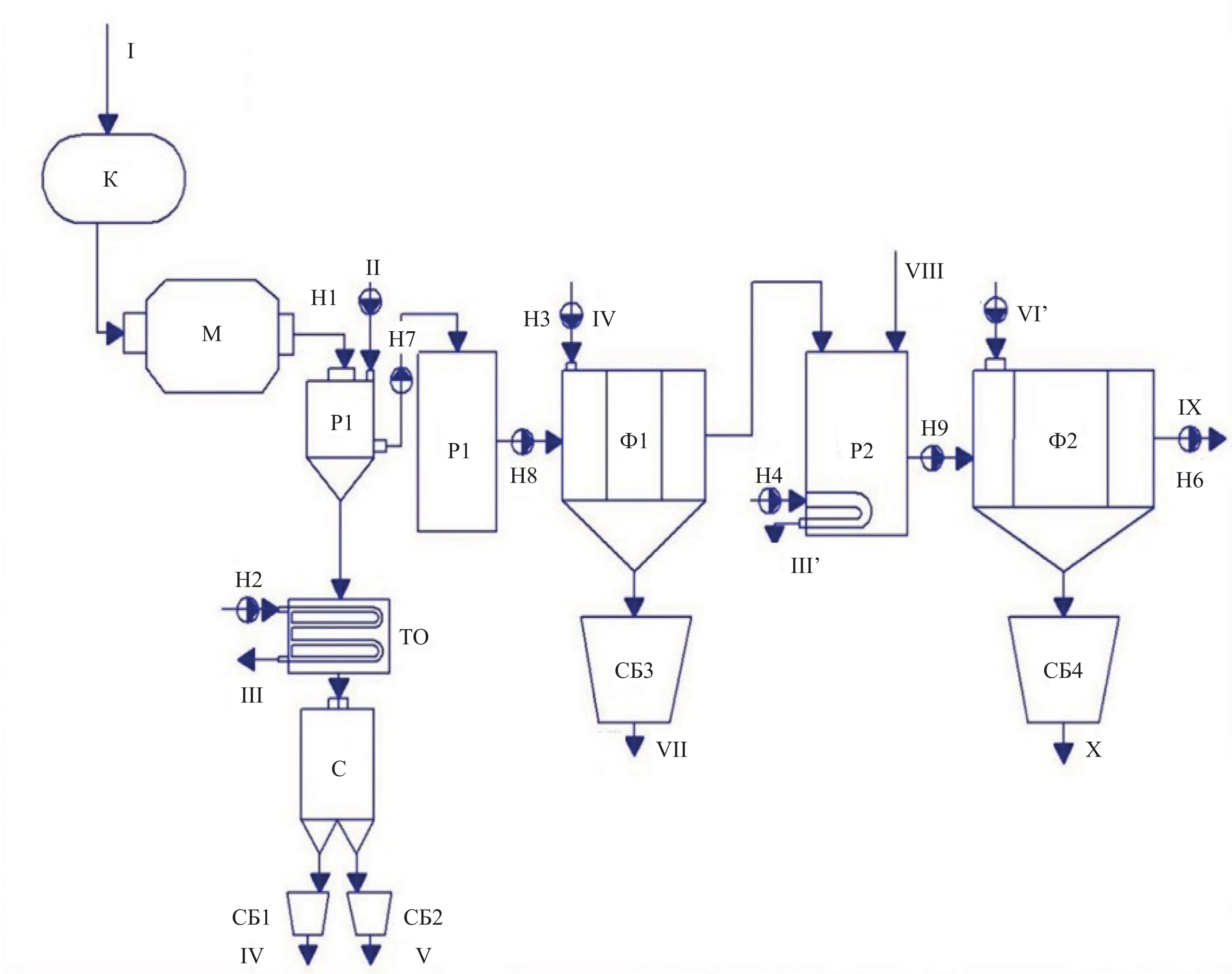

Fig. 4. Technological scheme of the LIB processing process:
I — spent LIB; II — water; III, III' — heat-carrying medium; IV — trapped plastic; V — trapped metals;
VI, VI' — water for washing the press filter; VII — trapped metal oxides, graphite; VIII — Na_2CO_3;
IX — filter for water purification; X — Li_2CO_3.

According to Figure 4, the technological scheme of the LIB processing process with the extraction of lithium carbonate consists of five stages: grinding, separation, filtration, precipitation and capture of wet Li_2CO_3 sediment. At the first stage, the equipment used is: a cryocamber, a ball mill, a vertical hammer crusher, a heat exchanger and a circulation pump. In the second stage, an electrostatic separator is used to separate metal particles and plastic. At the third stage, the main equipment used is a suspension pump and a press filter. They are necessary to capture manganese dioxide, graphite, metal particles and plastic residue. A reaction vessel is used to precipitate Li_2CO_3.

The material balance for the developed method has been calculated (Table 2). This calculation was based on a forecast that 1 ton of LIB with specific characteristics would be processed each day at the production site.

Table 2

Summary of the material balance of the recycling process

Input			
Products of processing	t/day	t/year	% wt
1. Waste in the form of lithium-ion batteries, including:	1.000	25.000	67.980
– Manganese dioxide	0.150	3.750	
– Graphite	0.060	1.500	
– Lithium compounds	0.110	2.750	
– Solvents	0.180	4.500	
– Metal particles	0.350	8.750	
– Plastic	0.150	3.750	
2. Water	0.403	10.075	27.396
Na_2CO_3	0.068	1.700	4.624
Total	1.471	36.775	100.000
Output			
Products of processing	t/day	t/year	% wt
1. Wet sediment Li_2CO_3	0.081	2.025	5.506
2. Filtrate	0.627	15.675	42.868
3. Sediment (on press filter 1)	0.254	6.350	17.267
4. Metal particles	0.349	8.725	23.725
5. Plastic	0.149	3.725	6.962
6. Water vapor released during the sludge drying process	0.011	2.750	3.672
Total	1.471	36.775	100.000

As a result of introducing the technology for LIB processing, 2.025 tons/year of wet Li_2CO_3 sludge, 6.350 tons/year of graphite, manganese dioxide, metal particles, and plastic will be produced each year from 25 tons of waste. Additionally, 8.725 tons/year of metal particles, 3.725 tons/year of plastic will be captured and separated at the primary stage. The resulting filtrate (15.675 tons/year) was proposed to be sent to the water treatment plant of the main production plant for the production of lithium-ion batteries N, where multilevel water purification takes place.

Discussion and Conclusion. As a result of the study, an analysis of various methods for recycling lithium-ion batteries has been carried out. The advantages and disadvantages of each method have been discussed. A search for the most promising method for LIB recycling has been conducted using the method of multi-criteria choice. A method for recycling lithium-ion batteries with the extraction of valuable components using a hydrochemical method has been presented. This system ensures safe recycling of used lithium-ion batteries with minimal negative impact on the environment and maximum recovery of valuable components. Calculations show that 2.025 tons/year of wet Li_2CO_3 sediment and 6.350 tons/year of graphite, manganese dioxide, metal particles and plastic sediment would be extracted from 25 tons/year of spent LIB.

Thus, the introduction of a closed-loop economy into the process of LIB processing can reduce the negative impact on the environment, as well as bring financial benefits when extracting valuable components. Recycling plays an important role in reducing waste and promoting the efficient use of energy.

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FS Melnikova: analysis of literary data, description of the theoretical part of the research, calculation of material balance, design of a scientific article.

NV Kostryukova: creation of a technological scheme of LIB processing process.

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