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Metallurgy

Lithium extraction methods and its application prospects: a review

[1*]Yersaiynova A.A., [1]Karshyga Z.B., [2]Muhammad N.A.A., [1]Yessengaziyev A.M., [1]Orynbayev B.M.

[1] *Institute of Metallurgy and Ore Beneficiation JSC, Satbayev University, Almaty, Kazakhstan*
[2] *University Pendidikan Sultan Idris, 35900 Tanjong Malim, Perak, Malaysia*

* *Corresponding author email: a.yersaiynova@satbayev.university*

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ABSTRACT
Lithium is the most important raw material for the production of modern electronics and electric vehicles. Today, it is impossible to imagine any mobile device without lithium batteries. The role of lithium in the global economy is only growing. The production of electric vehicles and batteries contributes to the reduction of carbon dioxide emissions. Nevertheless, end-of-life lithium-ion batteries pose a danger to the ecosystem. The article presents technological developments in the field of lithium extraction. The main sources of lithium are pegmatites, continental and geothermal brines, as well as clays, seawater and industrial brines. The main commercial lithium product is lithium carbonate (Li_2CO_3), which is obtained mainly from the mining, extraction and processing of spodumene ores and saltlake, oilfield brines. The effective role of lithium in addressing important issues such as pollution, climate change and the increasing depletion of natural resources used to produce lithium-ion batteries for these electric vehicles is also discussed.

Keywords: lithium outlook, lithium minerals, demand and use, lithium resources, key technologies.

Information about authors:

Yersaiynova Albina Abatkyzy
Doctoral student, laboratory of titanium and rare refractory metals, Institute of Metallurgy and Ore Beneficiation JSC, Satbayev University, Shevchenko str., 29/133, 050010, Almaty, Kazakhstan. Email: a.yersaiynova@satbayev.university; ORCID ID: https://orcid.org/0000-0003-0638-380X

Karshyga Zaure Baitaskyzy
Ph.D., Leading Researcher, Associate Professor, laboratory of titanium and rare refractory metals, Institute of Metallurgy and Ore Beneficiation JSC, Satbayev University, Shevchenko str., 29/133, 050010, Almaty, Kazakhstan. Email: z.karshyga@satbayev.university; ORCID ID: https://orcid.org/0000-0002-3025-7363

Muhammad Noorazlan Abd Azis
Ph.D., Associate Professor, Faculty of Science and Mathematics, Universiti Pendidikan Sultan Idris, Tanjung Malim, Perak, 35900, Malaysia. Email:azlanmn@fsmt.upsi.edu.my; ORCID ID: https://orcid.org/0000-0002-2792-4145

Yessengaziyev Azamat Muratovich
Ph.D., Head of the Laboratory of Titanium and Rare Refractory Metals of the Institute of Metallurgy and Ore Beneficiation JSC, Satbayev University, Shevchenko str., 29/133, 050010, Almaty, Kazakhstan. Email: a.yessengaziyev@satbayev.university; ORCiD ID: https://orcid.org/0000-0002-4989-4119

Orynbayev Bauyrzhan Munarbaiuly
Doctoral student, laboratory of titanium and rare refractory metals, Institute of Metallurgy and Ore Beneficiation JSC, Satbayev University, Shevchenko str., 29/133, 050010, Almaty, Kazakhstan. Email: Bauyrzhan.Orynbayev@stud.satbayev.university; ORCID ID: https://orcid.org/0000-0002-7730-9060

Introduction

Lithium and lithium-containing compounds are vital strategic resources for the economy, and their consumption is growing rapidly every year. Lithium is also one of the most important metals that set the direction of scientific and technological progress in the modern world. Lithium and its alloys are used in a wide variety of industrial applications, such as: batteries, ceramics and glass, lubricating greases, polymer production, medical, continuous casting mold flux powders, air treatment, and other uses [1].

In glass and ceramics manufacturing, lithium is used as a flux where it helps lower the melting point and increase durability [2]. Lithium can be used as an absorbent medium in industrial refrigeration systems as well as in humidity control and dehumidification systems. In metallurgy, lithium is used in the production of lightweight alloys used in aviation and aerospace, as well as in the production of high-strength, corrosion-resistant military components. In medicine, it is used to treat bipolar disorder, depression and other mental disorders [3]. Lithium is also used as a thickener in the production of lubricants. These lubricants have excellent heat resistance and water repellency properties, making them ideal for use in heavy equipment and automotive applications. Lithium is widely used today in the manufacture of batteries, especially lithium-ion batteries, which are used in electric transport, stationary energy storage systems, as well

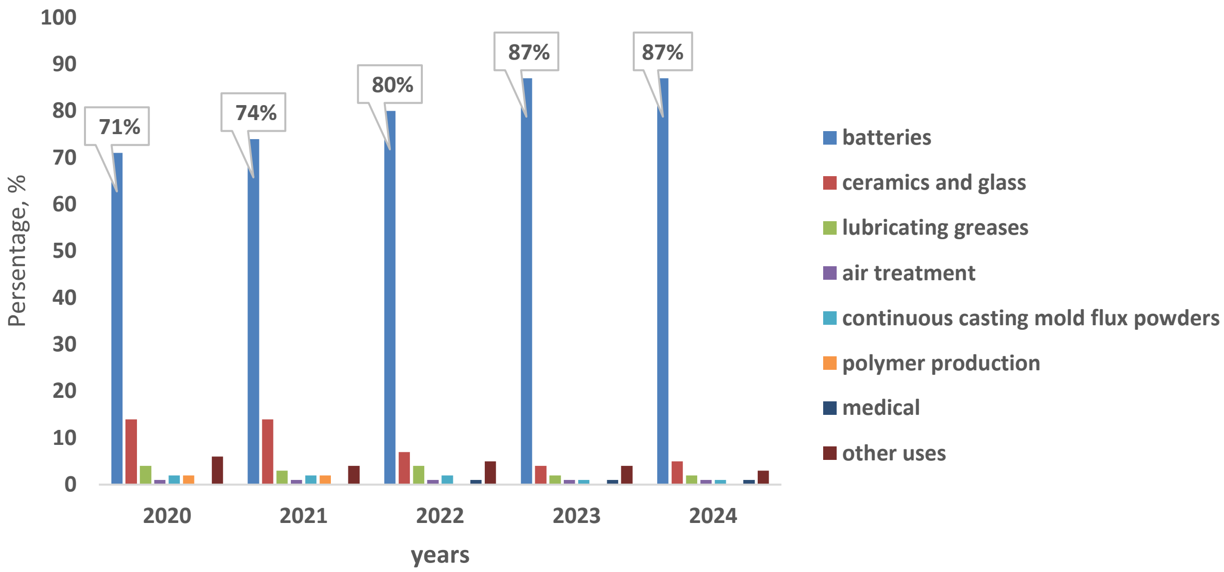

Figure 1 - Use of lithium and its compounds in the world from 2019 to 2023

as in compact power supplies for computer and telecommunications equipment. According to U.S. Geological Survey, Mineral commodity summaries over the last 5 years, the application area of lithium is changing [[4], [5], [6], [7], [8]]. Figure 1 shows a steady annual increase in the use of lithium in batteries, while other areas show a decrease.

Since Sony and Asahi Kasei introduced lithium-ion batteries in 1991, lithium-ion batteries have significantly changed many aspects of our daily lives [[9], [10]]. In recent years, the demand for lithium has been steadily increasing as global energy storage markets are growing rapidly [11]. The electric vehicle industry is experiencing particularly rapid growth, significantly accelerating this process [12].

Between 2021 and 2022, global fossil fuel-derived carbon dioxide (CO_2) emissions increased and reached the highest level in history [[13], [14]]. The main sources of atmospheric pollution are industrial enterprises and automobiles [15]. Industrial facilities emit gases and dust into the atmosphere, and vehicles emit significant amounts of exhaust gases [16]. Atmospheric pollutants include not only gaseous substances but also fine dust containing heavy metals such as lead (Pb), cadmium (Cd), and mercury (Hg), which pose a serious threat to human health [[17], [18], [19]]. Also, due to the emission of CO_x, NO_x and other gases into the atmosphere in large quantities, we intensively observe climate change every year: floods, forest fires, drought and other major disasters [[20], [21]]. In this regard, international organizations intend to switch to renewable energy sources to reduce the spread of greenhouse gases and to reduce the load of fossil fuels [22].

Increased demand for lithium has an impact on its price. The formation of lithium prices depends on its supply, which, in turn, may be constrained by its natural reserves, as they are concentrated in a limited number of countries.

Main lithium minerals and world reserves

The main sources of lithium are pegmatites, continental and geothermal brines, also clays, seawater and industrial groundwater [23]. The main commercial lithium product is lithium carbonate (Li_2CO_3), which is obtained mainly by mining, extraction and processing of spodumene ores and saltlake, oilfields brines.

More than 100 natural lithium minerals have been identified [[24], [25]]. Lithium is predominantly found in the form of silicates, less frequently in the form of phosphates, and lithium minerals from other classes are extremely rare [26]. Commercial lithium minerals are: spodumene, lepidolite, petalite, eucryptite and cinnwaldite, also other minerals are presented in Table 1 [27].

According to U.S. Geological Survey, Mineral commodity summaries U.S. Geological Survey lithium reserves worldwide in January 2024 are about 105 million tons [7]. Figure 2 shows that Bolivia, Argentina, USA and Chile have the largest lithium reserves. In Kazakhstan, according to the USGS, lithium reserves amount to 50 thousand tons.

Table 1 - The main lithium minerals

Mineral name	Chemical formula	Content of Li_2O, %
Spodumene	$LiAl[Si_2O_6]$	5.9-7.6
Bikitaite	$LiAlSi_2O_6*H_2O$	6.51
Holmquistite	$Li_2(Mg_3Al_2)Si_8O_{22}(OH)_2$	1.1
Lepidolite	$KLi_2Al(Al,Si)_4O_{10}(F,OH)_2$	4.1-5.5
Cinnwaldite	$K(Li,Fe,Al)_3(OH,F)_2[AlSi_3O_{10}]$	2.9-4.5
Tyniolite	$KLiMg_2[Si_4O_{10}](OH, F)_2$	3.70
Polylithionite	$KLi_2Al[Si_4O_{10}]F_2$	3.70-7.70
Bitite	$Ca_4(Li,Be,Al)_{12}[(Si,Al)_4O_{10}][OH]_2$	2.73
Kukeit	$LiAl_4[Si_3AlO_{10}](OH)_8$	0.80-4.33
Eucryptitis	$LiAl[SiO_4]$	6.1
Petalite	$LiAl(Si_4O_{10})$	3.4-4.1

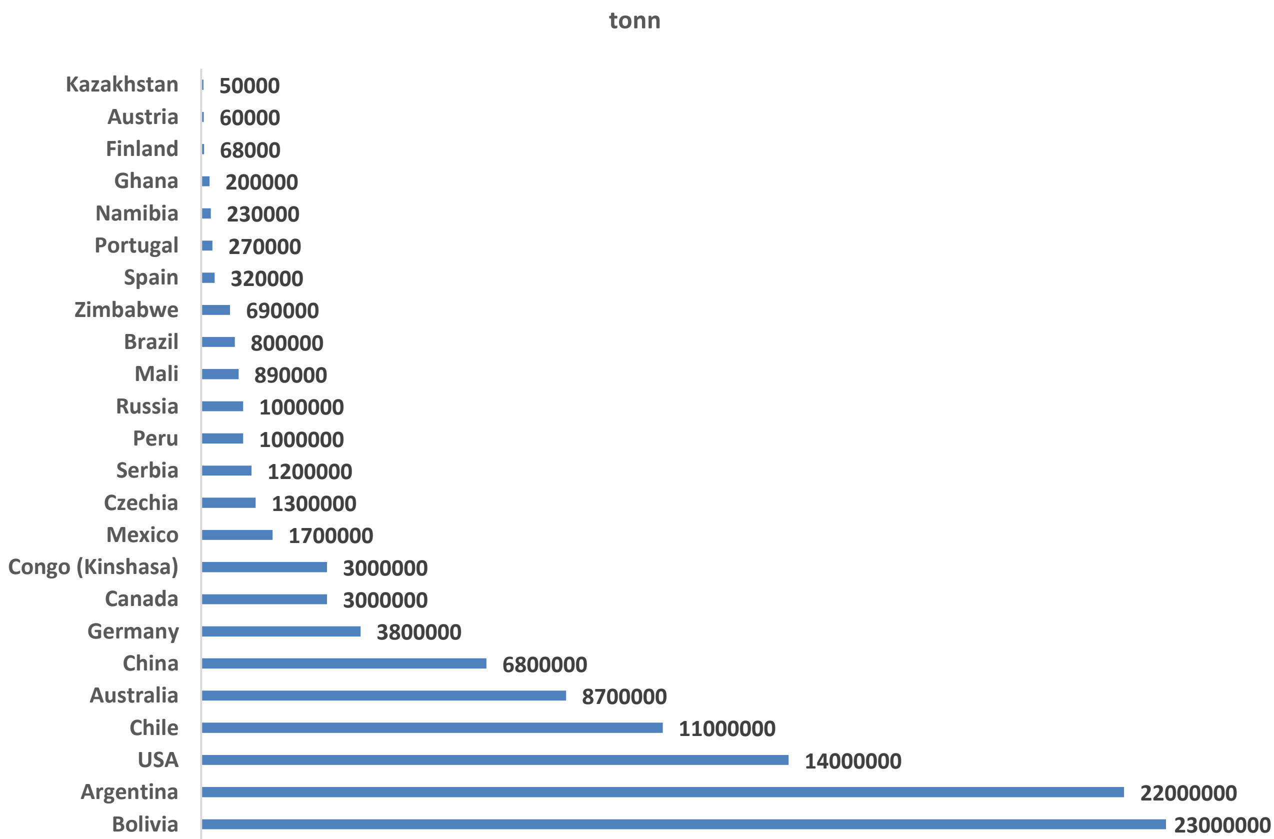

Figure 2 - Lithium reserves by country

President of Kazakhstan Kasym-Jomart Tokayev in 2022 during a meeting with the public of Zhetysu region stated about the large reserves of lithium in the bowels of Kazakhstan and the need for serious investment in the exploration and development of lithium, also instructed the Geological Service to intensify work in this direction [28].

Lithium deposits are mainly located: in East Kazakhstan region Akhmetkino, Akhmirovskoye, Bakennoye, Verkhne-Baimurzinskoye, Yubileynoye, Medvedka, Karasu, Kokkol, Targynskoye, in Aktobe region Verkhne-Irgizskoye. in Kostanay region Smirnovskoe and Drozhzhzhilovskoe, in Karaganda region lithium (Li) is established at the Zhanet deposit, in Almaty region at the Karagailyaktas deposit, in Zhambyl region lithium mineralization is established at the quartz vein deposit Maikol [29]. Currently, the above-mentioned sites are being explored, or the sites are mothballed or in reserve.

Mining activities are gaining momentum every year. Reserves of some rare elements have significantly decreased, while their demand has increased dramatically due to their increased use in new areas of engineering and technology. In this connection it is becoming more and more important to search for and involve in industrial development new types of mineral raw materials. One of such sources for many valuable elements necessary for various branches of economy can be underground industrial brines, which are a kind of “liquid ore” [30]. Its advantage over traditional ores is obvious: hydro-mineral raw materials have a polycomponent and can simultaneously serve as a source for the extraction of various valuable metals, such as lithium [31].

The comparison shows that the average brine deposit (1.45 million tons of lithium) is significantly larger than the average pegmatite deposit (0.11 million tons of lithium). Especially large brine deposits, such as Salar de Atacama in Chile and Uyuni in Bolivia, have a much larger total lithium resource (21.6 million tons of lithium) [[32], [33]]. Also lithium-bearing brine resources around the world are: Clayton Valley, USA, Salton Sea, USA, Salar de Atacama, Chile, HombreMuerto, Argentina, Salar de Uyuni, Bolivia, Searles Lake, USA, Great Salt Lake, USA, Dead Sea, Israel, Sua Pan, India, Bonneville, USA, Zabuye, Taijinaier, China. Thus, brine deposits have a much greater capacity for large-scale and long-term mining compared to pegmatite deposits. Lithium mining from brines is considered more favorable than mining from ores because it is more environmentally friendly and cost-effective [33].

On the territory of Kazakhstan 4 perspective provinces of industrial waters are identified: Pre-Caspian, Mangistau-Ustirt, Shu-Sarysu and South-Torgai. All provinces of industrial waters are associated with oil and gas bearing fields [34]. Locating lithium feedstock in or near industrial areas has significant advantages.

All natural sources of lithium raw materials have their own characteristics: the type of raw materials, the quantitative content of the main component, the accompanying impurity components and their quantity, and others.

Therefore, when processing existing raw materials and creating own production facilities, an individual approach is required with the study of modern technological developments and the involvement of new advanced solutions.

Lithium extraction methods

For spodumene concentrate, which is currently the main one among lithium concentrates, four treatment methods are known: sulfuric acid, sulfate, lime and chloride roasting. All these methods are used in industry, and the choice of a particular one depends on the economic efficiency [[35], [36]].

Sulfuric acid method. Lithium extraction by acid leaching is much more efficient when using β-spodumene as a starting material than α-spodumene [[37], [38], [39]]. Therefore, before sulfatization, natural spodumene is heat treated to convert the minerals into a more reactive form [[40], [41]]. After dicrypitation is followed by sulfatization with sulfuric acid, Decomposition reaction of spodumene with sulfuric acid to form lithium sulfate [[42], [43]]:

$$Li_2O{\cdot}Al_2O_3{\cdot}4SiO_2 + H_2SO_4 \rightarrow Li_2SO_4 + 4SiO_2 \cdot Al_2O_3 \cdot {\cdot}H_2O \quad (1)$$

$$Li_2SO_{4(s)} \rightarrow Li_2SO_{4(aq)} \quad (2)$$

$$Li_2SO_{4(aq)} + Na_2CO_{3(aq)} \rightarrow Li_2CO_{3(s)} + Na_2SO_{4(aq)} \quad (3)$$

In this process, H^+ ions from the acid chemoselectively replace Li^+ ions in spodumene as described in equation (1), forming water-soluble $Li_2SO_{4(s)}$ [42]. The resulting soluble compound is leached with water according to equation (2), after which the solution is treated with lime at 90°C to adjust pH and remove impurities. Further, lithium carbonate is extracted by adding sodium carbonate solution to the extract, which is reflected in equation (3) [[42], [43]]. The technological scheme of the sulfuric acid method of spodumene concentrate processing is presented in Figure 3.

The advantage of the sulfuric acid method is: no need to grind the ore due to the fact that preliminary decryption leads to its loosening, so that sulfatization with sulfuric acid is successfully carried out even in the treatment of large fractions, and there are no long high-temperature processes.

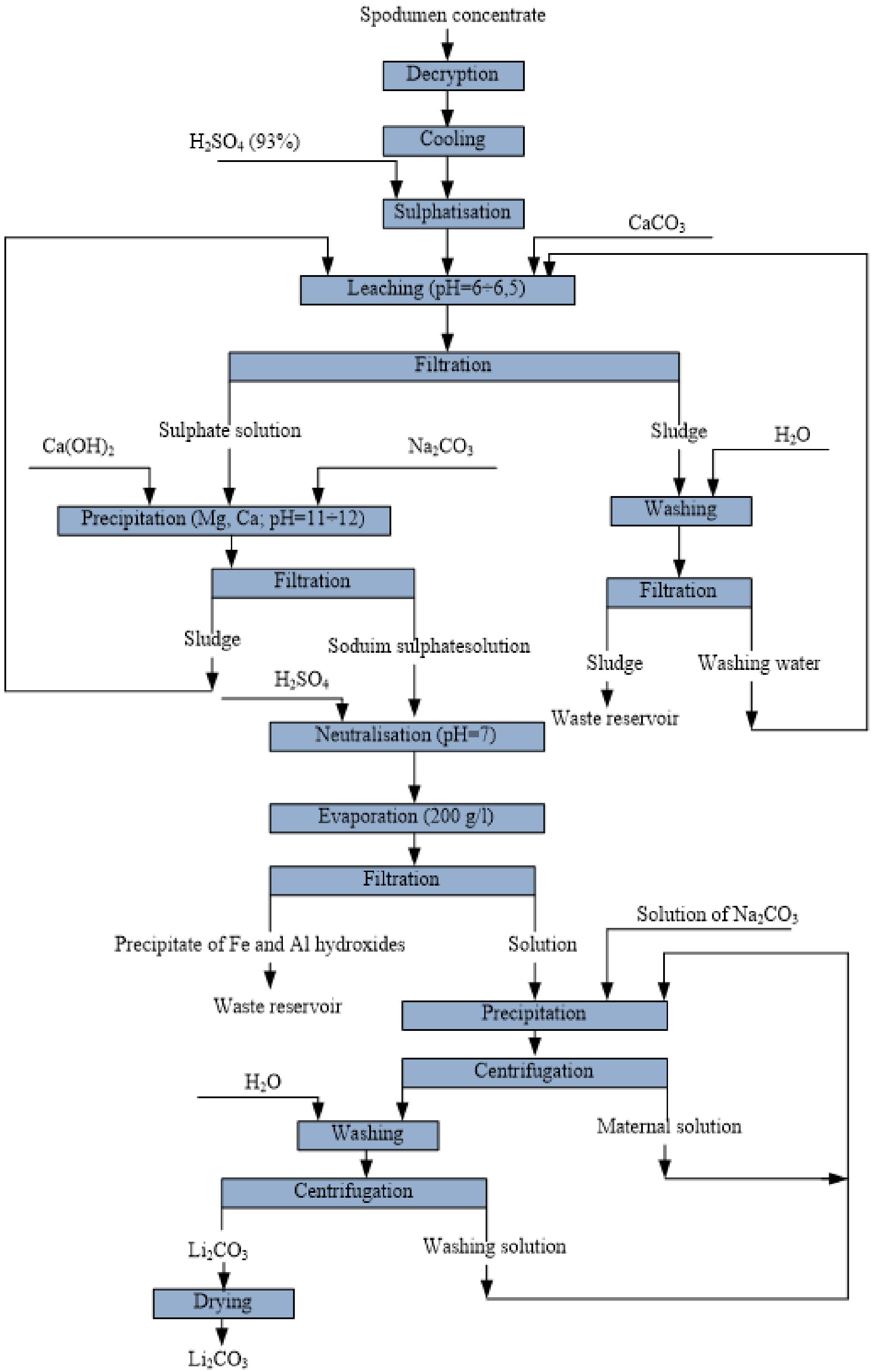

Figure 3 - Technological scheme of sulfuric acid method of spodumene concentrate processing

Sulfate method. This method is based on sintering of lithium ores and concentrates with potassium sulfate [[26], [44]]. The process is based on direct substitution of lithium with potassium [45]. Sintering of natural spodumene with potassium sulfate takes place at temperatures of 920 -1050°C, where α-spodumene under heating changes into β-spodumene, which reacts with potassium sulfate:

$Li_2O{\cdot}Al_2O_3{\cdot}4SiO_2 + K_2SO_4 = Li_2SO_4 + K_2O{\cdot} Al_2O_3{\cdot}4SiO_2$ (4)

The obtained sinter is quenched with water and leached, separating the insoluble residue by decantation or filtration. The solution containing lithium sulphate and impurities is purified with potassium or sodium hydroxides, after which lithium carbonate (Li_2CO_3) is precipitated with sodium carbonate according to equation (5). The precipitated lithium carbonate is squeezed, washed and dried:

$$Li_2SO_{4(aq)} + Na_2CO_{3(aq)} \rightarrow Li_2CO_{3(s)} + Na_2SO_{4(aq)} \quad (5)$$

The technological scheme of spodumene concentrate processing by sulphate scheme is presented in Figure 4.

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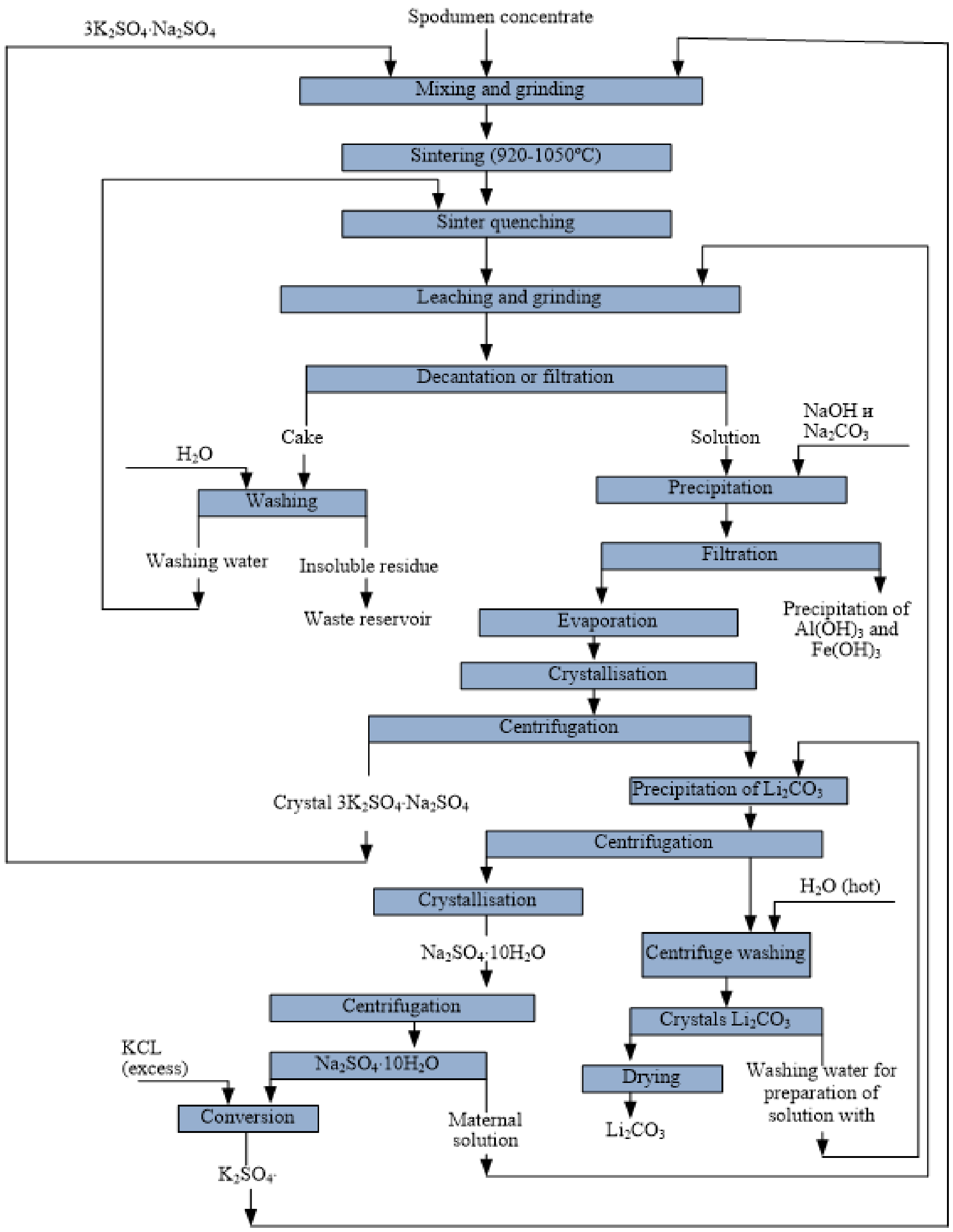

Figure 4 - Technological scheme of spodumene concentrate processing by sulfate scheme

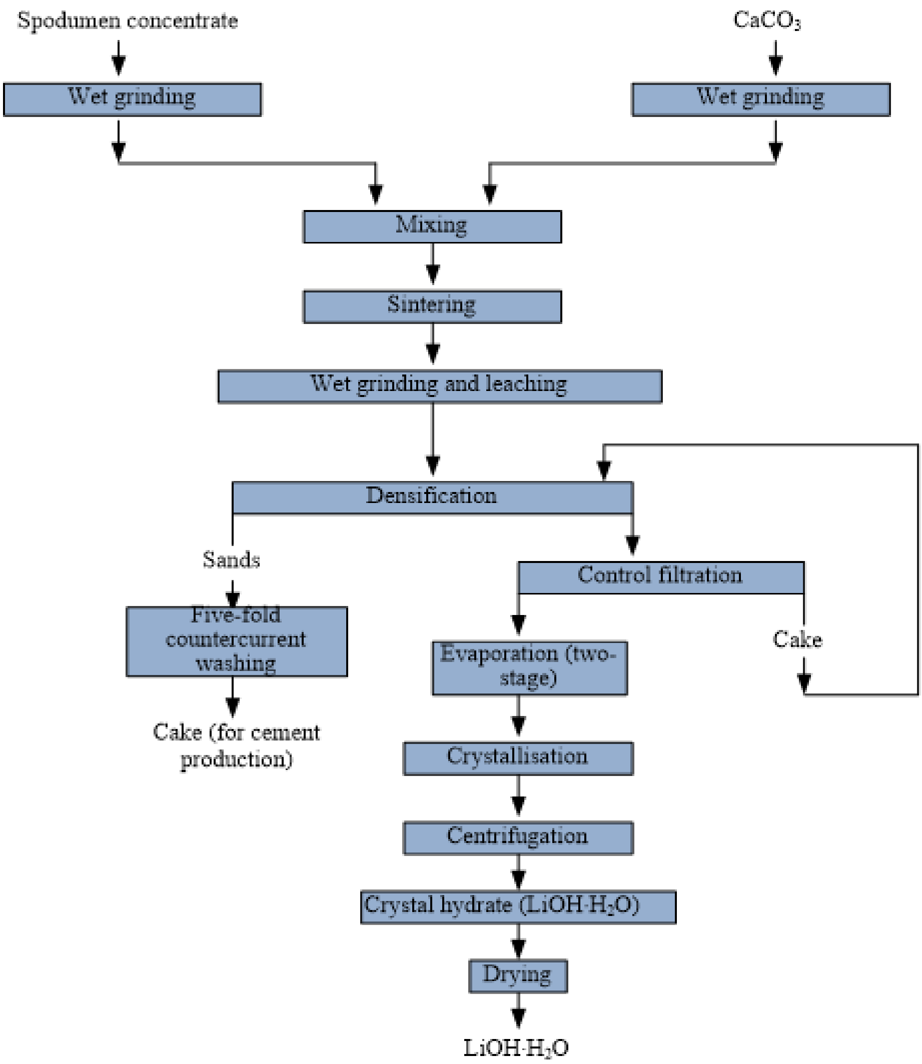

Figure 5 - Technological scheme of spodumene concentrate processing by lime method

Under optimal sintering conditions, the degree of mineral penetration reaches 90-95%, which indicates the high efficiency of the method. Also this method allows to obtain lithium carbonate with high purity, which is important for further use in industry. The disadvantage of the method is a large consumption of potassium sulphate.

Lime method. The lime method is based on the sintering of lithium concentrates with the adding of lime or limestone, and then decomposition of the resulting sinter with water [44].

The technological scheme of spodumene concentrate processing by the lime method is presented in Figure 5.

Sintering of spodumene with limestone is carried out at a temperature of 1150-1200°C. The following reaction takes place:

$$Li_2O{\cdot}Al_2O_3{\cdot}4SiO_2 + 8CaCO_3 \rightarrow Li_2O{\cdot}Al_2O_3 + 4(2CaO{\cdot}SiO_2) + 8CO_2 \quad (6)$$

The reaction of spodumene with lime can be described as follows:

$$Li_2O{\cdot}Al_2O_3{\cdot}4SiO_2 + 4CaO + 4H_2O \rightarrow 2LiOH + 2Al(OH)_3 + 4CaSiO_3 \quad (7)$$

The lime method has the following advantages: it is universal for opening of all lithium minerals, it does not require scarce reagents (only natural limestone is needed). However, the lime scheme has serious disadvantages: the initial concentrate should be rich in lithium content, and limestone should be of high quality, with low content of silicon, aluminum, iron [46]. Due to the low concentration of lithium in solutions after leaching, large volumes of process equipment and high energy consumption for evaporation are required.

Chlorinating method. One of the methods of processing ores and concentrates containing lithium is chlorinating roasting [[45], [47]]. This method was used to treat concentrates containing lepidolite and spodumene. The material consisting of lepidolite, NH_4Cl and $CaCO_3$ was roasted at 750 - 800°C and then leached with water [48]. The mineral decomposes by the reaction:

$$Li_2O\cdot Al_2O_3\cdot 4SiO_{2(тв)} + 4CaCO_{3(тв)} + 2NH_4Cl \rightarrow 2LiCl_{(тв)} + 4CaSiO_3 + Al_2O_3 + 4CO_{2(г)} + 2NH_3 + H_2O \quad (8)$$

Evaporation of the solution crystallizes a mixture of lithium and calcium chlorides, which can be used for electrolysis or lithium hydroxide production by causticization. This method has been commercialized in the USA for the extraction of lithium from spodumene concentrate.

Chlorination roasting allows to obtain sufficiently pure lithium chloride directly from ores without using expensive reagents. The disadvantages of this method include the difficulty of capturing lithium chloride and high aggressiveness of furnace gases.

In recent years, new methods like hydrofluoric acid method, autoclave method, and microwave firing have been proposed [[49], [50]]. However, there are still some disadvantages associated with these methods, such as the use of highly toxic reagents and high energy consumption.

Extraction of lithium from hydromineral raw materials become popular among lithium producers due to its low production costs.

According to literature data, lithium ions from hydromineral raw materials can be extracted using conventional methods such as: evaporation, precipitation, as well as direct lithium extraction methods: solvent extraction, electrochemical, adsorption and membrane methods [[51], [52], [53]].

The conventional method of extracting lithium from underground brine is to pump it to the surface and further place it in huge ponds. Over a long period (up to a year or more), the water gradually evaporates, allowing the concentration of lithium to rise to a level sufficient to allow it to be precipitated by chemicals. However, this method is only applicable in sunny, dry regions [54].

As a result of the limitations of conventional technologies, direct lithium extraction (DLE) techniques have been developed to address these shortcomings and provide more efficient, environmentally friendly and cost-effective recovery of lithium from resources with relatively low grades of this metal.

Membranes. The membrane method is classified based on their driving mechanisms: electrodialysis, which relies on electrical potential, and nanofiltration, which operates under pressure. The main membrane technologies are nanofiltration, microfiltration and ultrafiltration. Nanofiltration membranes (NF) are capable of passing monovalent ions while simultaneously trapping multivalent ones. This property allows them to be used to separate lithium from divalent ions such as Mg^{2+} and Ca^{2+} [55]. In recent years, they have become a key method for lithium extraction, showing a rapid growth of interest from researchers and progressing steadily towards full-scale industrial utilization [56]. This interest is due to the low energy consumption, excellent cyclic stability, high separation efficiency, large porosity, permeability, surface area, and good mechanical stability of membranes [57]. In addition, they are easily adaptable to different operating conditions However, this process has its disadvantages, especially that membranes clog quickly when working with saturated solutions.

Solvent extraction. This method, known as liquid-liquid extraction, uses the difference in solubility properties of different salts to isolate lithium. The extraction process usually involves separating the solution into two separate phases: aqueous and organic. During this process, the impurities remain in the aqueous phase, while the lithium ions pass into the organic phase. Extractants include neutral organic phosphorous compounds, β-diketones, crown esters, and ionic liquids. One of the widely used extractants is $TBP/FeCl_3$ [58]. This is a promising method for lithium production, but it has drawbacks, including the need for an additional extractant purification step, possible loss of target material, and risks of environmental contamination when using organic reagents.

Electrochemical method. Electrochemical processes are one of the most studied and effective methods of lithium extraction, characterized by a high degree of extraction and ease of operation, as well as environmental safety [59]. The process is based on a principle similar to battery reverse charging technology, where an external electrostatic field is used to stimulate the movement of ions between a pair of specially designed electrodes designed to extract lithium ions [60]. The process is usually carried out cyclically: firstly, the target ions are introduced into the working electrode, and then released back into the solution during the regeneration stage to repair the electrode. In electrochemical methods, the performance of electrode materials plays a key role [61]. Therefore, it is necessary to optimize electrode materials that have a high ability to capture and release lithium, as well as demonstrate stability and corrosion resistance.

Adsorption. Methods of lithium sorption from brines can be carried out using both organic and inorganic sorbents. But organic ionites are selective not only to lithium, but also to other ions, which can complicate the separation and purification of extracted lithium, as well as lead to the formation of organic waste, which may require special methods of disposal or treatment, which increases the environmental burden of the process [62].

For the treatment of low lithium concentrations, the ion exchange adsorption method can be applied, where ionic sieves are used as adsorbents [63]. Lithium ion sieves are considered one of the most promising materials for lithium extraction from low lithium brines due to their high adsorption capacity and excellent lithium selectivity. Studies show that manganese oxide (LMO) and titanium oxide (LTO) based sorbents as an effective method for lithium extraction from brines [[64], [65]].

The greatest success has been achieved in developing ion sieves based on lithium manganese oxide (LMO). In particular, the lithium ion sieve material ($H_2Mn_2O_4$), derived from the spinel structure $LiMn_2O_4$ after acid treatment, exhibits high selectivity for Li^+ ion adsorption in aqueous solutions. This makes it a promising candidate for lithium extraction from natural brines. Such materials possess a high adsorption capacity.

Thus, according to the review, innovative approaches such as direct lithium extraction technologies present opportunities to develop a more sustainable and efficient lithium supply chain. This highlights the dynamic and ever-evolving nature of the lithium mining industry as it adapts to technological advancements and environmental challenges.

Conclusions

As the literature review has shown, lithium is currently one of the highly demanded metals of strategic importance, as well as contributing to solving environmental problems and improving the quality and level of our daily lives. Significant lithium reserves available in Kazakhstan allow to create production facilities for extraction and processing of domestic lithium-containing raw materials to produce valuable lithium products. Existing processing methods are designed for raw materials of appropriate composition, so research and development of technology acceptable for Kazakhstani lithium raw materials is an urgent task. The development of efficient and environmentally friendly lithium extraction technologies will enhance Kazakhstan's position in the global lithium supply chain, contributing to economic growth and technological advancement. Future studies will focus on optimizing extraction processes, minimizing environmental impact, and exploring innovative approaches to utilize Kazakhstan’s lithium resources effectively.

Conflicts of interest. On behalf of all authors, the corresponding author states that there is no conflict of interest.

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Литийді алу әдістері және оны қолдану перспективалары: шолу

[1]Ерсайынова А.А., [1]Қаршыға З.Б., [2]Muhammad N.A.A.,
[1]Есенгазиев А.М., [1]Орынбаев Б.М.

[1] Металлургия және кен байыту институты АҚ, Сәтбаев университеті, Алматы, Қазақстан
[2]Пендикан Сұлтан Идрис университеті, Перак, Малайзия.

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ТҮЙІНДЕМЕ
Литий қазіргі заманда электроника мен электр көліктері үшін маңызды шикізат болып табылады. Бүгінгі күні литий батареялары жоқ кез келген мобильді құрылғыны елестету мүмкін емес. Литийдің әлемдік экономикадағы рөлі тек қана артып келеді. Электрлік көліктер мен батареяларды өндіру көмір қышқыл газының шығарындыларын азайтуға көмектеседі. Дегенмен, қызмет ету мерзімі біткен литий-иондық батареялар экожүйеге қауіп төндіреді. Мақалада литийді өндіру саласындағы технологиялық әзірлемелер берілген. Литийдің негізгі көздері пегматиттер, континенттік және геотермалдық тұзды ерітінділер, сондай-ақ саз, теңіз суы және өнеркәсіптік тұзды ерітінділер болып табылады. Литийдің негізгі коммерциялық өнімі литий карбонаты (Li_2CO_3) болып табылады, ол негізінен сподумен кендері мен тұзды ерітінділерді өндіруден және өңдеуден алынады. Сондай-ақ, қоршаған ортаның ластануы, климаттың өзгеруі және электр көліктеріне арналған литий-иондық аккумуляторларды өндіру үшін пайдаланылатын табиғи ресурстардың сарқылуы сияқты маңызды мәселелерді шешуде литийдің тиімді рөлі талқыланады.

Түйін сөздер: литий перспективалары, литий минералдары, сұраныс және пайдалану, литий ресурстары, негізгі технологиялар.

Авторлар туралы ақпарат:

Ерсайынова Альбина Абатқызы — *Докторант, титан және сирек қиын балқитын металдар зертханасы, Металлургия және кен байыту институты АҚ, Сәтбаев университеті, Шевченко көш., 29/133, 050010, Алматы, Қазақстан. Email: a.yersaiynova@satbayev.university; ORCID ID: https://orcid.org/0000-0003-0638-380X*

Қаршыға Зәуре Байтасқызы — *Ph.D., жетекші ғылыми қызметкер, қауымдастырылған профессор, титан және сирек қиын балқитын металдар зертханасы, Металлургия және кен байыту институты АҚ, Сәтбаев университеті, Шевченко көш., 29/133, 050010, Алматы, Қазақстан. Email: z.karshyga@satbayev.university; ORCID ID: https://orcid.org/0000-0002-3025-7363*

Мухаммед Нуразлан Абд Азис — *Ph.D., қауымдастырылған профессор, ғылым және математика факультеті, Пендикан Сұлтан Идрис университеті, 35900 Танджонг Малим, Перак, Малайзия. Email: azlanmn@fsmt.upsi.edu.my; ORCID ID: https://orcid.org/0000-0002-2792-4145*

Есенгазиев Азамат Муратович — *Ph.D., титан және сирек қиын балқитын металдар зертханасының меңгерушісі, Металлургия және кен байыту институты АҚ, Сәтбаев университеті, Шевченко көш., 29/133, 050010, Алматы, Қазақстан. Email: a.yessengaziyev@satbayev.university; ORCiD ID: https://orcid.org/0000-0002-4989-4119*

Орынбаев Бауыржан Мұнарбайұлы — *Докторант, титан және сирек қиын балқитын металдар зертханасы, Металлургия және кен байыту институты АҚ, Сәтбаев университеті, Шевченко көш., 29/133, 050010, Алматы, Қазақстан. Email: Bauyrzhan.Orynbayev@stud.satbayev.university; ORCID ID: https://orcid.org/0000-0002-7730-9060*

Методы получения лития и перспективы его применения: обзор

[1]Ерсайынова А.А., [1]Қаршыға З.Б., [2]Muhammad N.A.A.,
[1]Есенгазиев А.М., [1]Орынбаев Б.М.

[1] АО Институт металлургии и обогащения, Satbayev University, Алматы, Казахстан
[2]Университет Пендидикан Султан Идрис, Перак, Малайзия

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АННОТАЦИЯ
Литий - важнейшее сырье для производства современной электроники и электромобилей. Сегодня невозможно представить ни одно мобильное устройство без литиевых батарей. Роль лития в мировой экономике только возрастает. Производство электромобилей и аккумуляторов способствует сокращению выбросов углекислого газа. Тем не менее отслужившие свой срок литий-ионные батареи представляют опасность для экосистемы. В статье представлены технологические разработки в области добычи лития. Основными источниками лития являются пегматиты, континентальные и геотермальные рассолы, а также глины, морская вода и промышленные рассолы. Основным коммерческим продуктом лития является карбонат лития (Li_2CO_3), который получают в основном при добыче, извлечении и переработке сподуменовых руд и рассолов. Также обсуждается эффективная роль лития в решении таких важных проблем, как загрязнение окружающей среды, изменение климата и растущее истощение природных ресурсов, используемых для производства литий-ионных батарей для электромобилей.

Ключевые слова: перспективы лития, минералы лития, спрос и использование, ресурсы лития, ключевые технологии.

Информация об авторах:

Ерсайынова Альбина Абатқызы — *Докторант, лаборатория титана и редких тугоплавких металлов, Институт металлургии и обогащения АО, Satbayev University, ул.Шевченко, 29/133, 050010, Алматы, Казахстан. Email: a.yersaiynova@stud.satbayev.university; ORCID ID: https://orcid.org/0000-0003-0638-380X*

Қаршыға Зәуре Байтасқызы	*Ph.D., ведущий научный сотрудник, ассоциированный профессор, лаборатория титана и редких тугоплавких металлов, Институт металлургии и обогащения АО, Satbayev University, ул. Шевченко, 29/133, 050010, Алматы, Казахстан. Email: z.karshyga@satbayev.university; ORCID ID: https://orcid.org/0000-0002-3025-7363*
Мухаммед Нуразлан Абд Азис	*Ph.D., ассоциированный профессор, факультет естественных наук и математики, Университет Пендидикан Султан Идрис, 35900 Танджонг Малим, Перак, Малайзия. Email:azlanmn@fsmt.upsi.edu.my; ORCID ID: https://orcid.org/0000-0002-2792-4145*
Есенгазиев Азамат Муратович	*Ph.D., заведующий лабораторией титана и редких тугоплавких металлов, Институт металлургии и обогащения АО, Satbayev University, ул.Шевченко, 29/133, 050010, Алматы, Казахстан. Email: a.yessengaziyev@satbayev.university; ORCiD ID: https://orcid.org/0000-0002-4989-4119*
Орынбаев Бауыржан Мұнарбайұлы	*Докторант, лаборатория титана и редких тугоплавких металлов, Институт металлургии и обогащения АО, Satbayev University, ул.Шевченко, 29/133, 050010, Алматы, Казахстан. Email: Bauyrzhan.Orynbayev@stud.satbayev.university; ORCID ID: https://orcid.org/0000-0002-7730-9060*

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