# Review on hydrometallurgical processing technology of lateritic nickel ore for the last 20 years in the world

**K. K. Mamyrbayeva**, *Ph.D., Associate Professor of Department "Metallurgical Processes, Heat Engineering and Technology of Special Materials", Mining and Metallurgical Institute[1], e-mail: k.mamyrbayeva@satbayev.university*
**A. N. Kuandykova**\*, *Master's Degree, Ph.D. Student of Department "Metallurgical Processes, Heat Engineering and Technology of Special Materials", Mining and Metallurgical Institute[1], e-mail: almira.kuandykova@stud.satbayev.university*
**T. A. Chepushtanova**, *Ph.D.,Candidate of Technical Sciences, Head of Department "Metallurgical Processes, Heat Engineering and Technology of Special Materials", Associate Professor, Mining and Metallurgical Institute[1], e-mail: t.chepushtanova@satbayev.university*
**Y. S. Merkibayev**\*, *Ph.D., Head of Laboratories of Department "Metallurgical Processes, Heat Engineering and Technology of Special Materials", Mining and Metallurgical Institute[1], e-mail: y.merkibayev@satbayev.university*

[1]*Satbayev University, Almaty, Kazakhstan.*

In recent years, there has been an increased demand for nickel-cobalt-containing batteries, which in turn adds a lot of interest in the production of these metals in large volumes. Due to a contraction of the volume of nickel sulfide ores, the majority of producers consider laterite deposits as a potential source of nickel and cobalt. However, processing laterites by traditional pyrometallurgical methods is economically unprofitable. Research has shown that the use of combined and hydrometallurgical methods can be promising. This article is devoted to a review of the results of research and articles on the extraction of nickel and cobalt from laterite ores over the past 20 years. Current technological schemes for the production of nickel and cobalt using combined manufacturing processes are presented, as well as the results of scientific research on increasing the recovery degree of nickel and cobalt from low-grade refractory laterite ores using preliminary activating roasting, various leaching reagents, oxidizing agents, bacteria, etc. Growing interest in nickel production has also been observed in Kazakhstan and research work on the extraction of target metals from local laterite ores is being intensively carried out. According to the investors' forecast, the first mining in the republic and full commissioning of nickel production are expected in 2025.

***Key words:*** leaching, nickel, cobalt, hydrometallurgy, laterites.

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## Introduction

Nickel with its unique properties [1] is a strategic metal for many modern industrial processes — for the production of non-ferrous alloys and alloy steels [2—4], electrochemical processes [5—6], commercial chemicals, catalysts [7—8], nanomaterials for solar batteries [9—10]. Recently, the growth of nickel consumption has particularly increased in the field of clean energy development [11] — manufacturing of electric vehicles and lithium-ion (Li-ion) batteries [12—13]. According to the IEA (International Energy Agency), by 2050, metal consumption for clean energy technologies will increase by 10.8—30.1 times for nickel and by 9.9—32.9 times for cobalt [14].

Experts estimate that the total world reserves of nickel exceed 100 million tons, and low-grade oxidized nickel ore (laterites) represents the bulk of the world's nickel reserves (85%) [15]. Despite this, more than half of the primary nickel production comes from sulfide ores. In nickel-containing ores, cobalt is the main accompanying metal, the content of which does not exceed about 0.08%, due to the high growth in demand for cobalt at present, it is also a promising metal for extraction.

Laterite deposits are widespread in tropical regions of the world, such as: Caledonia, Australia, Cuba, Brazil, Colombia, Greece, the Philippines, Indonesia and India [16—17]. Most of the laterite deposits found on the Earth's surface are of the limonite type [(Fe,Ni)O(OH)-$n$H$_2$O]. Iron oxides, namely goethite [FeO(OH)], are the main chemical component of limonite minerals, with which nickel is closely related [18—19]. As a rule, silicate-saprolite types containing 1.5—2.5% nickel lie under the limonite zone [20]. In nickel deposits, on the other hand, the most common nickel sulfide mineral in the earth's crust is nickel pentlandite [(Fe,Ni)$_9$S$_8$], in which the bulk of nickel is present in the form of iron-sulfur complexes. Overall, nickel sulfide ores provide about three-quarters of the world's nickel production, of which pentlandite accounts for almost 90% of nickel sulfide ores [21—23].

Higher quality lateritic minerals are processed industrially using pyro- and hydrometallurgical methods, the choice of which depending on the chemical and mineralogical composition of the ore. Pyrometallurgical

\*Correspondence author.

methods are used in the processing of ores with a nickel content above 1.5 (wt.%). Ores are processed through certain energy-intensive processes such as drying, calcination, roasting, high-temperature reduction and smelting, and in the case of hydrometallurgical methods, mineral acids, inorganic and organic solvents, or combinations thereof are used to treat ores to leach metals. Basically, hydrometallurgical processing is used for ores with low nickel content with the ratio of the main components $SiO_2/MgO$, Fe/Ni and Ni/Co 1.5 and 12 and more than 30, respectively [24].

The most common concentration process used before industrial hydrometallurgical processing of nickel laterites is to remove the coarse fraction from the feedstock, which has a lower Ni content than the finer material. This leads to the generation of larger wastes, which are either stored or processed through heap leaching. A number of other methods for physically separating nickel laterites are absorber-float (or dense medium) separation, gravity separation, magnetic separation, electrostatic separation (roasting) and flotation. However, none of these technologies, although providing some degree of upgrading, have not been fully utilized in hydrometallurgical technologies, which allows us to assume that the complex composition of nickel laterites does not require preliminary benefication [25—26].

In industry, the extraction of nickel from laterite minerals is carried out by such processes as: Caron process, high-pressure acid leaching (HPAL), atmospheric acid leaching (AAL-atmospheric acid leaching), biochemical leaching (BL-Bio chemical leaching), and heap leaching (HP-Heap leaching). Among hydrometallurgical processes for laterite processing, the HPAL process is the most effective for recovering nickel and cobalt, but high capital investment and operating costs represent a significant disadvantage. Other methods are characterized by low selectivity and recovery of valuable metals associated with high consumption of reagents, as in the case of AAL and HL [27—30]. These metallurgical processes are economically viable and industrially acceptable when nickel recovery is carried out from relatively higher quality oxidized nickel minerals (lateritic minerals).

## 1.1. Caron technology

Caron technology is a comprehensive technology for processing lateritic nickel ores, including pyro- and hydrometallurgical processes. Laterite ores are mainly processed using this technology. The ore is roasted to selectively reduce Ni and Co to metallic forms. These metals are then leached with a solution of ammonia or ammonium carbonate. A small amount of iron is reduced to form an alloy with Ni and Co [31—33]. First, the ore is calcined at a temperature of 850 °C, after cooling to 150—200 °C, the ore is treated with an ammonia/ammonium carbonate solution, after filtering the solid (cake) from the liquid (productive solution), Co is extracted in the form of cobalt sulfide, and Ni — nickel carbonate.

Nickel carbonate is further calcined to obtain the final product —nickel oxide.

During the process, ammonia can be recirculated so reagent costs are low and the leaching step is highly selective for Ni and Co. Recovery rates of Ni and Co can be relatively low compared to HPAL (<90% Ni, <80% Co) and the energy requirement for roasting the ore is very high, which increases operating costs [34—35].

Caron technology was first introduced at the Nicaro plant in Cuba (the final product is nickel oxide), then at the Yabulu plant in Australia [36] (with the production of NiO powders, NiS — CoS sulfide concentrate), at the Sao Miguel Paulista plant in Brazil (with the production of cathode nickel powder, currently stopped), at the Punta Gorda plant, Cuba (producing NiO powders).

In general, the Caron technology has its advantages, such as a high recovery degree of Ni and Co, and the possibility of ammonia solution recirculation. The disadvantages of the technology are the electrically intensive processes of drying and reduction roasting, the production of relatively poor productive solutions (with Ni content up to 2 $g/dm^3$, Co up to 0.3 $g/dm^3$), large losses of Ni and Co during separation from iron hydroxides by filtration, exposure to ammonia solutions on the environment. To solve the problem of ammonia's impact on the environment, in most enterprises the ammonia solution after leaching is returned to the beginning of the process with preliminary regeneration, that is, to leaching a new batch of ore.

Research is being conducted towards completely replacing ammonia with safer reagents, one of them is monosodium glutamate [37].

## 1.2. HPAL High Pressure Acid Leaching

High pressure acid leaching is based on sulfuric acid leaching of Ni and Co from laterites at high temperature (240—270 °C) and pressure (2000—4000 kPa) in an autoclave. Under such conditions, the rate of basic chemical reactions accelerates and the leaching process becomes effective: it is completed in just 60—90 minutes. The recovery of Ni and Co under these conditions is more than 95%. At temperatures above 200 °C in an acidic solution (pH = 2—3), iron precipitates in the form of hydroxide, which reduces acid consumption and allows the selective extraction of Ni and Co from the productive leaching solution [38—39].

The HPAL process is suitable for processing limonite ores, since silicate and clay ores contain acid-intensive minerals (micas, chlorites, etc.), which affect the overall economic efficiency of the process.

The main advantage of the HPAL technology is the high leaching rate (from 30 to 120 minutes) [40—41]. Traditional leaching methods last much longer and, as a result, have lower recovery rates [42].

In the Philippines, limonite ore from the Mindoro deposit is processed using high pressure sulfuric acid leaching (HPAL) technology, leaching time is 30 minutes, nickel

recovery is 96.5—97.5%. Saprolites mined at this deposit are leached at atmospheric pressure; Nickel extraction in 6 hours of leaching reaches up to 97—98%. The end products of the enterprise are nickel cathode and cobalt sulfate. The enterprise's capacity is estimated at 52.7 thousand tons of nickel and 15.9 thousand tons of cobalt sulfate. Average recovery is 89% for nickel and 91% for cobalt [43—44].

High acid consumption of 350—500 kg per tonne of ore is one of the main disadvantages of HPAL. In addition, the capital costs of HPAL can be high due to the highly corrosive nature of the leaching medium, which requires expensive titanium-lined autoclaves [45].

Despite this lack of technology, the Coral Bay nickel production plant using HPAL technology has been successfully operating in the Palawan deposit, Indonesia since 2005. The main product is a sulphide concentrate containing 57% nickel and 4% cobalt. Nickel production currently amounts to 10.000 tons per year [9—10]. The flow chart for nickel extraction using HPAL technology is shown below **(Fig.)** [45].

### 1.3. Heap and atmospheric acid leaching

Heap leaching is a well-established process used to process many non-ferrous metals, namely copper, gold and uranium. Heap leaching of nickel laterite is simple and flexible and can be applied to many laterite deposits that currently have no viable route to mining, and as evidence of this the method is currently used by Glencore

(Minara and Xstrata), Vale, BHPB and Anglo American for nickel laterites [46].

The performance of heap leaching is influenced by many factors, such as the granulometric and mineralogical composition of the ore, the type, concentration and permeability of the solvent, the duration of leaching, etc. Although heap leaching is mainly used for the processing of oxidized ores, in the production of nickel —for laterite ores, research has established that mineralogy Nickel laterites have a strong impact on the recovery of nickel and cobalt. It was shown in [47] that high rates of Ni extraction (88—96%) during sulfuric acid leaching were achieved mainly from low-goethite ores, but goethite content was not the only factor influencing Ni extraction. A study of elements substituted in goethite has shown that greater extraction of Ni is achieved from ores in which Fe is partially replaced by Ni or Co. But the proportion of Co recovered during leaching did not strongly correlate with the content of a particular mineral phase. This is consistent with the findings of [48].

Today, the optimal solvent for heap leaching of laterite ores is sulfuric acid. Conducted studies on heap sulfuric acid leaching of laterites have shown that nickel recovery up to 85% can be achieved in less than 40 days with an iron content in solution of less than 5%. The consumption of sulfuric acid varies from 10 to 25 kg of recovered nickel [49].

Despite the positive aspects, heap leaching of nickel remains at the stage of study for the processing of many
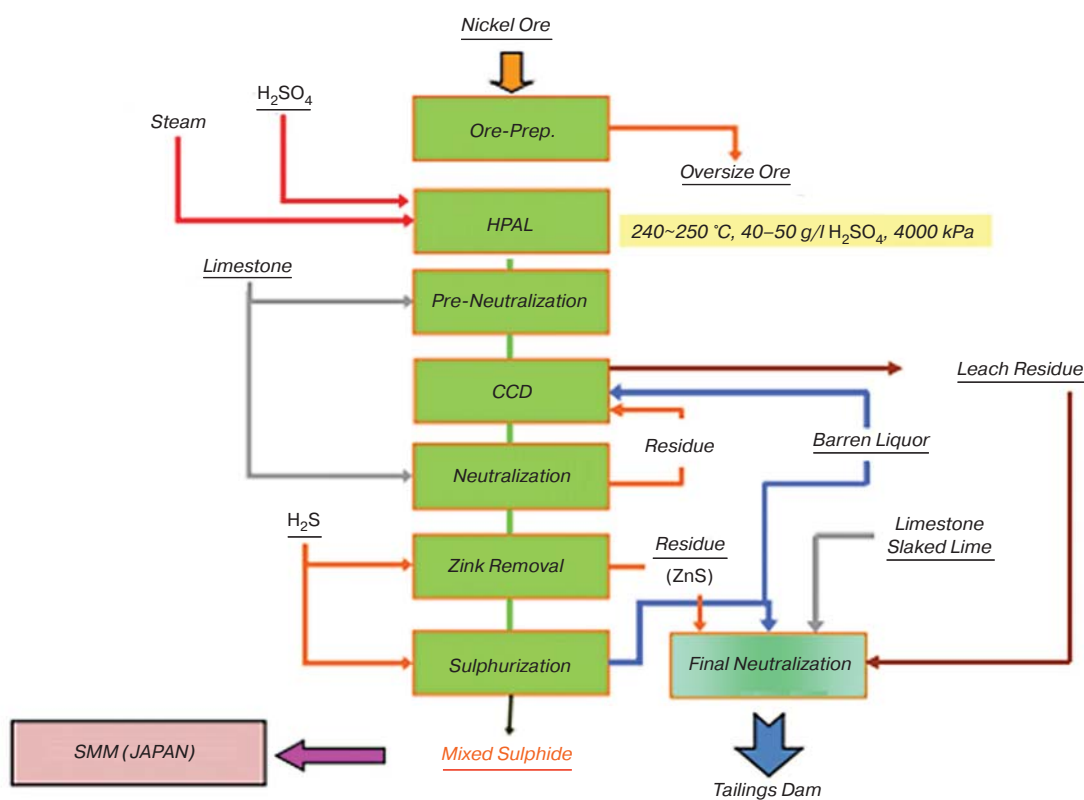


**Fig.** HPAL technology for processing nickel ores [45] (*original picture uses by Creative Commons Attribution License rules*)

deposits. A method is proposed for the agglomeration of laterite ore [50−52], double aeration [53], the use of new types of solvent, bacteria [54], etc., which have a positive effect on the extraction of nickel, cobalt, etc.

For low-grade nickel laterite ores, stirred tank atmospheric sulfuric acid leaching is the preferred hydrometallurgical method based on lower capital and operating costs compared to HPAL. In this leaching method, sulfuric acid is preferred due to its availability and efficiency [55]. Limonite and saprolite ores are leached with concentrated sulfuric acid in stirred tanks at atmospheric pressure. The process is carried out at a temperature of 100 °C, and the leaching time is up to 12 hours using 2 mol/l $H_2SO_4$ and a $S : L$ ratio of 1 : 4. In the first stage, Ni and Co are leached from the limonite ore, then the excess sulfuric acid in the suspension is neutralized by the addition of saprolite ore, releasing more Ni and Co into solution. This process can be applied to low-grade ores (<1.5% Ni), with Ni and Co leaching recoveries greater than 90% for Ni and greater than 80% for Co. The main disadvantages of this process are the long leaching time (2 hours or more) and high acid consumption compared to HPAL (500−700 kg per ton of ore) [56].

Over the past decade, numerous studies have been carried out to develop technological schemes for non-oxidizing acid leaching of nickel-containing ores. J. Mäkinen et al. [57] used the addition of fluorides as an oxidizing agent in sulfuric acid leaching of laterite ore to increase the efficiency of nickel extraction. Fluorides have an intensifying effect on the dissolution process of nickel silicate minerals and as a result, more than 90% nickel recovery was achieved.

H. Basturkcu et al. [58] found that the greatest leaching effect in the recovery of nickel from the Kaldag region (Turkey) laterite ore is achieved using various salt additives. As such additives, that activate the leaching process, NaCl, $Na_2S_2O_5$, $Na_2SO_4$ and KCl were tested. The research results showed that the addition of NaCl or KCl led to a reduction in the duration of leaching by half (4 hours instead of 8) and a decrease in the degree of iron leaching by 16%. Under these conditions, the recovery degree of nickel did not change and amounted to 98.2%.

In the non-oxidizing sulfuric acid processing of laterite ore, in which the main nickel-containing mineral is nontronite, calcium fluoride was used as an intensifying additive. By leaching ore with the addition of calcium fluoride in an amount of 10 kg/t, an increase in nickel extraction by more than 10% was achieved, that is, the recovery degree increased from 80−83% to 94%. This reagent is capable of slowly decomposing in sulfuric acid solutions to form hydrofluoric acid. The latter well dissolves the ferrous silicate mineral nontronite with the formation of complexes of the type $[FeF_6]^{3-}$ [59] and leads to an inevitable increase in the recovery degree of nickel, which was built into the crystalline structure of this mineral.

J. Li et al [60] managed to extract nickel up to 92.3% when leaching laterite ore from Yunnan Province, China with a solution of hydrochloric acid.

As the practice of existing nickel production enterprises has shown, the use of mineral acids as solvents is effective for nickel extraction, but on the other hand, they create serious environmental problems. In this regard, recent research has been carried out to find alternative leaching methods using organic acids, which, in addition to being relatively cheap and widely available, have environmental advantages. For example, the possibility of recovering nickel and cobalt by leaching low-grade serpentine-rich sulfide ore using an alkaline glycine leaching system has been explored [61]. Many researchers have confirmed that citric acid, along with mineral acids, is also an effective reagent for leaching nickel from saprolites and laterites [62].

### 1.4. Biochemical leaching

Recently, there has been increasing interest in studies of nickel and cobalt leaching in the presence of microorganisms. Bioleaching is very attractive because it is environmentally friendly, but the rate of leaching process is low. Studies have shown that heterotrophic microorganisms such as Bacillus, Enterobacter (Aerobacter), Pseudomonas, Coulobacter, etc. are used for leaching of laterite ores [63−65].

In [66], lateritic nickel ore was used for bacterial leaching using a mixed consortium of mesophilic acidophiles that are members of the genus Thiobacillus. For the experiments, laterite ore was taken in various forms, such as raw, roasted ore at 400−600 °C. Leaching experiments were carried out under the following conditions: stirring speed 400-500 rpm, temperature 35 °C. It was found that the extraction of nickel and cobalt from raw ore is low and amounts to 9.47% and 41.12%, respectively. Leaching of roasted ore with a pulp density of 10% in the presence of microorganisms led to a noticeable increase in the solubility of nickel-cobalt containing minerals, the maximum recovery of nickel and cobalt reached 77.23% and 73.22%, respectively.

The authors [67] showed that when leaching laterite ore using two different microbes acidithiobacillus ferrooxidans and delftia acidovorans at a temperature of 90 °C for 3 hours with a stirring speed of 370 rpm and a $S : L$ ratio = 1 : 1, maximum nickel extraction were 83.65% and 80.18% respectively, while cobalt recovery reached 86.93% and 83.94%. Based on leaching experiments in the presence of delftia acidovorans, at 30 °C and 150 rpm incubator for 30 days, metal recovery rates decreased and amounted to 57.09% and 60.19%, respectively, and when using at.ferrooxidans microbes, the final nickel and cobalt recovery rates were 59.41% and 64.85%, respectively. The most effective bacteria for leaching laterite ores turned out to be acidophilic bacteria (at. ferrooxidans). The data are consistent with the results of the study [68−70]. Bioleaching rates, along with bacterial types, are strongly influenced by nickel and cobalt mineralogy [54]. It has been established that Mn- and Mg-containing minerals of nickel and cobalt with low iron content, such as serpentines, are better bioleached by the reduction of

divalent iron generated by bacteria, compared to other minerals of these metals. Aerobic bioleaching of laterites with at.thiobacillus and at.caldus resulted in lower pH and therefore higher extraction of Ni and Co than anaerobic bioleaching with a ferrooxidans. Therefore, the bioleaching rates of limonite nickel laterites vary significantly depending on the mineralogical composition of the ores [54]. Ni recovery from laterites decreases with increasing goethite content, but in some lateritic deposits goethite can be unusually reactive and therefore quickly dissolved by acid. Reductive bioleaching needs some improvements to become a viable technology applicable to the processing of goethite-rich laterite ore.

Nickel bioleaching is a cost-effective process, as it does not require large expenses, is environmentally friendly, safe and is an alternative method to traditional nickel extraction [71]. To improve process performance, research has recommended the use of ultrasound when carrying out biological leaching [72].

### 1.5. Chloride leaching

Due to its strong reducing properties, hydrochloric acid is an effective solvent for many metal oxides and salts, allowing high rates of extraction of valuable components into solution at atmospheric pressure. Today, there are several enterprises that use the technology of chloride leaching of laterites. Such companies include BHP Billiton, Jaguar Nickel, Nichromet Solutions Inc, Intec Ltd, Anglo American (Anglo Research Nickel process — ARNi), etc. [73—75].

Hydrochloric acid leaching is effective for processing laterite ores; one of the main arguments against the use of hydrochloric acid as a solvent on an industrial scale is that hydrochloric acid is aggressive to the equipment used and therefore there is a need to use more expensive materials in the equipment.

BHP Minerals developed a chloride-based heap leach process [76], in which nickel and cobalt were extracted from the chloride solution by ion exchange or precipitation with magnesium oxide. Iron was either also precipitated with the help of magnesium oxide, or itself was subjected to pyrohydrolysis after liquid extraction. These methods of iron removal, while technically quite possible, required large quantities of magnesium oxide in the first case, and in the second case, the pyrohydrolysis method requires a lot of expensive energy. In this regard, both methods turned out to be economically infeasible. The project of Jaguar Nickel Inc. is the closest to industrial implementation, which is developing a new processing technology using hydrochloric acid leaching of laterite ore for a plant in Guatemala (Central America) to produce nickel and cobalt. [77].

As the results of the conducted studies have shown, hydrochloric acid is an effective leaching reagent, but the introduction of hydrochloric acid leaching technology in many fields is hampered due to high cost, strict safety requirements and adverse environmental consequences.

At the State Scientific Research and Production Enterprise "Kazmekhanobr", studies of the behavior of the main components of laterites during hydrochloric acid leaching using electrodialysis have continued. This combined leaching method makes it possible to increase the extraction of nickel into solution to 82.3%; cobalt up to 72.7%, at the same time, the extraction of iron into the solution remains virtually unchanged [78].

### 2. Nickel and cobalt production in Kazakhstan

Kazakhstan has significant mineral resources, which contain about 1.5 million tons of nickel (1.4—2% of global reserves) and 100 thousand tons of cobalt, which is 1.4% of global reserves. Almost 100% of nickel and cobalt is found in laterite ores with a nickel content of 0.9—1.01% and cobalt of 0.05—0.06%. In terms of nickel content in ore, Kazakhstan is significantly inferior to the main countries with significant nickel reserves. Nickel ore deposits are concentrated in the northern regions of Kazakhstan and are mainly concentrated in the Bugetkol (Aktobe region), Gornostaevsky (East Kazakhstan region) deposits, as well as in the Kempirsay (Aktobe region) and Ekibastuz-Shidertinsk (Pavlodar region) group of deposits.

Scientists in Kazakhstan have previously conducted a number of studies devoted to the development of innovative and environmentally friendly technology for processing nickel-cobalt-containing raw materials in Kazakhstan [79—80].

The authors of [79] conducted studies on non-oxidative leaching of nickel from laterites of the Bugetkol deposit using concentrated solutions of sulfuric acid. Agitation leaching was carried out with sulfuric acid solutions with concentrations of 24.5; 68.6; 152; 230; 661.5; 823; 1778 g/l with constant stirring, leaching time was 6 hours, $S : L$ ratio = 1 : 10, ore sample weight 25 g, temperature 20 °C To open nickel minerals, the optimal acid concentration is 661.5 g/l, recovery degree of nickel into the solution was 97.37%.

Studies on the leaching of nickel, cobalt and iron from laterites of the Kempirsay nickel-bearing massif using a hydrochloric acid solution [80] have established that at a temperature of 65 °C and a process duration of 2—4 hours, the extraction of target components into the solution is: Ni — 99.8 %, Co — 98.7 %, Fe — 99.3 %.

Satpayev University is conducting research on complex processing of nickel-containing ores of Batamsha deposit with nickel content of 0.8—1.04%, cobalt 0.08—0.1%. Preliminary studies on sulfuric acid leaching of laterite ore showed that maximum nickel extraction is achieved at a sulfuric acid concentration of 250 kg/t, leaching temperature 80—85 °C, it was found that at a concentration of 100—250 g/dm$^3$ of sulfuric acid in the temperature range 70—85 °C the recovery degree of nickel and cobalt is in the range of 55—95%. The dissolution of nickel and cobalt from raw ore occurs in diffusion and transient modes ($E = 7$—35 kJ/mol).

SAT & Company and the Kaznickel Company in the Gornostaevskoye deposit of nickel-cobalt-containing ores located in the East Kazakhstan region are carrying out primary work on the extraction of nickel by underground leaching with the release of ferronickel as the final product. The planned production capacity is from 50 to 150 thousand tons per year of ferronickel.

Along with laterite ores, tailings and waste from existing plants can serve as sources of nickel and cobalt. More than 7 thousand tons of nickel and 14 thousand tons of cobalt per year [39—40] are lost in the Sokolovsko-Sarbaisky Mining Production Association in Kazakhstan, along with tailings and technogenic raw materials. Research is being conducted to extract nickel and cobalt from the products of sulfatizing roasting of the tailings of this enterprise.

## Conclusion

Nickel and cobalt, due to their unique properties, are widely used in various fields and it is expected that in 30 years the consumption of nickel and cobalt will increase to an average of 300%.

Due to the depletion of rich sulfide ores, an increase in the production of nickel and cobalt is expected due to the introduction of laterite ores into processing. It is known that the production of nickel and cobalt from laterites is considered unprofitable due to their complex mineralogical composition and low content of target metals. However, as the practice of operating enterprises over the past 20 years shows, laterite processing can be carried out using combined and hydrometallurgical processes, namely HL, Caron process, AAL, HPAL technologies, bacterial leaching, chloride leaching and others.

Currently, several enterprises using hydrometallurgical and combined technologies operate in China, Australia, the Philippines and other countries, which produce metallic pure nickel, nickel oxides and various nickel-containing compounds.

Scientific research conducted since 2000 on the extraction of nickel and cobalt from low-grade laterite ores has shown that the choice of processing method strongly depends on the mineralogical composition of laterites. Nickel is readily leached from smectite and saprolite laterites, which contain large amounts of magnesium and silica. Limonite ores with a large amount of ferrous minerals are poorly leached. Common effective leach reagents are sulfuric acid and hydrochloric acid. The recovery rates of target metals from laterites are influenced by factors such as the type and concentration of the leaching reagent, temperature, leaching duration, solid-to-liquid phase ratio, and ore size. To activate the processes of leaching of metals from high-refractory laterites, the addition of high-potential oxidizers and preliminary activating roasting in the temperature range 350—700 °C were effective. In recent years, research has been carried out on laterite leaching using new organic solvents, as well as bacterial leaching with new types and combinations of bacteria, which turned out to be less environmentally friendly and longer lasting.

In the Republic of Kazakhstan there are mainly laterite deposits with low nickel content (no more than 1.5% and cobalt 0.8%). However, the complexity of the ore composition and low content of target metals in them, and the lack of optimal processing technology still hinder their production. Conducted research and extensive testing of the developed technologies have shown that sulfuric acid leaching and the use of combined technologies to obtain pure sulfide and sulfate compounds of nickel and cobalt are acceptable for the processing of Kazakh laterite ores. The possibility of involving in the production of middlings and tailings of existing plants of Kazakhstan as sources of nickel and cobalt is also shown. To process these materials, it is proposed to use a combined technology, including oxidative-sulfatizing roasting and further sulfuric acid leaching.

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