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Low Grade Alumina Sources

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INNOVATIVE TECHNOLOGY FOR ALUMINA PRODUCTION FROM LOW-GRADE RAW MATERIALS

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Abstract

Increasing numbers of research institutes and industrial companies in the world are showing their interest in developing alternative methods for producing alumina from low grade raw materials. This paper examines the advantages and disadvantages of methods proposed by different authors for producing alumina by hydro chemical technology (alkaline and acid) compared with the alkaline sintering technologies.

1. Introduction

In the year of 2011 world bauxite mining amounted to 220 million tons, alumina refineries produced 98.3 million tons of Al_2O_3 and output of aluminium smelters was 44.1 million tons of primary aluminium [1].

Current estimation of bauxite reserves is 29 billion tons, in Africa (26%), South America and the Caribbean (25%), Oceania (21%), Asia (17%), and elsewhere (12%). Bauxite resources are estimated to be 55 to 75 billion tons, in Africa (32%), Oceania (23%), South America and the Caribbean (21%), Asia (18%), and elsewhere (6%) [1].

While the main resources of bauxite are concentrated in the Equatorial zone and the Southern hemisphere, most of the aluminum smelters are located in the Northern hemisphere (Table 1). This reflects long transportation distance between the bauxite reserves and aluminium production and consumption regions, often including necessity of both sea and inland transportation. For example total length of some logistic chains in UC RUSAL: Bauxite Mine - Alumina Refinery - Aluminum Smelter can take as much as 15 thousand kilometers.

Table 1

Distribution of aluminium production and bauxite reserves in the World

Regions	Aluminium production	Bauxite reserves
China	51 %	3 %
North America	13 %	<1%
CIS	9 %	1 %
Middle East	7 %	1 %
Europe	5 %	9 %
Asia excluding China	5 %	14%
Central & South America	5 %	25%
Australia	4 %	21%
Africa	1 %	26%

Ore deposits are moving more and more from the coast to the interior that increases cost of mined bauxite. According to Komlossy [2], above 50% of the world bauxite resource can not be economically exploited due to prohibitive expenses associated with need of infrastructure development. Many companies neglect already discovered bauxite mining areas due to their remote location and instead try to identify the deposits in geographically favorable location.

Another factor is steady decrease in quality of bauxite mined as best quality ore is consumed first. Notable in this respect is situation in China, where middle-low grade bauxite ($A/S < 7$) now occupies more than 70% of the resource [3] with sharp reduction in quality of bauxite in many plants just over last 3-4 years.

This increases the competition for access to quality bauxites globally. The most sensitive to this situation are the largest aluminum producers in China, Canada, Russia, and the Middle East regions due to not having sufficient bauxite resources of their own.

Meanwhile, most of the above major aluminum-producing countries have essentially inexhaustible resources of low-grade natural and man-made aluminous raw materials (low-grade bauxites and nonbauxitic ores) that were considered subeconomic to process until recently. According to Patterson [4], the nonbauxitic ores of aluminium include: aluminium phosphate rock, aluminous clay, alunite rock, coal waste washings and spoil, coal ash, dawsonite in oil shale, high-alumina clay, anorthosite, nepheline syenite, leucite, and saprolite. There are trillions of tones of aluminous shales, and hundreds of billions tones of alumina reach clay in the above aluminium-producing countries. In addition, there are tens of billions of tones of sillimanite group minerals.

Content of Al_2O_3 in alumina rich clays and other non-bauxitic rocks can be as high as 30-35%, content of alumina in such ores as nepheline and alunite rocks can be lower, in the range of 22-26%. While in the work of Edwards [5] published in 1930 typical alumina content in bauxite was 60 % giving it more distinctive advantage, now bauxite of 38-43 % of Al_2O_3 is not uncommon. Whereas, in the beneficiated non-bauxitic products like coal fly ash or sillimanite, it can reach over 60 % making them similar to almost extinct bauxite in terms of alumina content.

The lack of good quality bauxites and the need to secure a domestic raw material base is driving research in new technologies to process low grade ores into alumina, with the intention that these technologies will lead to a significant reduction of bauxite and alumina transportation costs, allow the extraction of more valuable components from the ore and reduce environmental impact. Establishment of commercial nepheline

and alunite processing technologies in the USSR, low grade bauxite processing in former USSR and China, semi-commercial stage trials of United States Bureau of Mines (USBM), CSIRO, Péchiney-Alcan and VAMI, as well as recent developments in fly ash processing technologies in Inner Mongolia (China) and hydrochloric acid process by Orbite Aluminae Inc. in Canada (latter two are referred in individual papers of current proceedings) are strong examples of this approach. Below alkaline and acidic methods are further explored.

2. Alkaline methods to process low grade ores into alumina

Apart from Bayer process for high and medium grade bauxites, alkaline methods are applicable to low grade bauxites, aluminosilicates and basic low grade ores including silicoaluminates of the alkaline or alkaline-earth metals such as the nepheline syenite and anorthosites. While there are several methods that have been tried on an industrial scale, only industrially established / prospective methods are described in this chapter.

2.1. History of development

The first method of extracting alumina from bauxite by sintering with soda ash, leaching of sodium aluminate from the sinter and decomposition of green liquor by carbonation was developed in **1854 by Luis de Le Chatelier** [6]. First industrial implementation of this method was at the Salendre plant in the South of France. Applicable to low silica bauxites, it was the only method for alumina production for decades before the Bayer process. This method is still used for caustic soda make up at UAZ alumina refinery in Russia, being economical due to lower price of soda ash compared to caustic soda.

In **1880 G. Mueller** (Germany) made this technology suitable for processing high silica ores due to addition of magnesite/dolomite to the sintering mix in order to bind silica into insoluble silicates of Mg or Ca [7]. In 1902 Mark Packard (Buffalo, USA) recommended optimal mix composition for sintering: $\text{CaO}:\text{SiO}_2=2.0$; $\text{Na}_2\text{O}:\text{Al}_2\text{O}_3=2.0$ [8]. The same lime ratio is still applicable now. In Russia this method was further developed by VAMI specialist work (A. Yakovkin, V. Mazel, F. Strokov et.al.), including dry and wet mix sintering. The method is efficiently used for alumina production at sinter branch at Bogoslovky alumina refinery.

A reduction melting method to process bauxites in Russia was developed in **1915 by Alexander Kuznetsov and Evgeny Zhukovsky**. According to this method, bauxite is mixed with carbonate of alkaline earth metal (Ba), coal and iron chips, mix is agglomerated and subjected to high temperature reduction melting in electrical furnaces. Result is ferrosilicon alloy and slag containing aluminate of the alkaline earth metal, latter is treated by soda liquor in order to extract alumina [9]. The method is suitable for complex processing of high iron bauxite. It was used to produce alumina from Tikhvin bauxite at Dniepr aluminium smelter before World War II but was substituted to Bayer process after the War due to its high energy consumption.

Karl Josef Bayer developed the method in **1887-1892** when he worked in Russia at the Tentelev Chemical plant (St. Petersburg) and Elabuga Chemical plant at Kama River. Two discoveries made the basis of the method by him: (i) ability of supersaturated green liquor to self-decompose [10], and (ii) ability of alumina

contained in bauxite to dissolve in solution of alkali at high temperatures [11] and possibility to use spent liquor for this purpose.

At the moment about 96 % of the world alumina is produced by this process, providing low capital and operating costs at production of high/medium grade bauxite. Low grade bauxites with high silica content make Bayer process uneconomical (lower alumina recovery and high caustic soda and heat consumption). This required development of Bayer – sintering combine methods for low grade bauxites and also methods to process non-bauxitic materials.

A series version of Bayer-sintering series method for processing low grade bauxites (A/S ~ 3) comprises Bayer processing and subsequent red mud lime sintering to recover alumina and soda back to the process. This method was developed by **Vladimir Mazel** and other VAMI engineers and commercially applied in Pavlodar plant in Kazakhstan (start up in **1964**). Application of this method for processing of bauxite with high silica content increases alumina chemical recovery up to 93% of total Al_2O_3 content. This plant shows excellent efficiency and is one of the lowest cost producers.

VAMI institute in USSR has elaborated [12] and implemented commercially the unique technology to process alunite ore into alumina, potassium sulfate, and sulfuric acid. The process was fully developed by **Georgy Labutin** and other VAMI engineers in **1953** and been used for engineering and construction of Kirovbad alumina refinery (Ganja, Azerbaijan) with production capacity of 150 000 t/y of alumina.

The development of the technology for processing of alkaline aluminosilicate ore started in USSR in early 1930's in order to utilise the nepheline concentrates after beneficiation of apatite-nepheline ores from Kola Peninsula, Russia. Development of basic technology was conducted in VAMI and GIPCH under guidance of **F. Strokov**, P. Vlodavetz, I. Lileev and others. In the middle of **1930's** it was decided to retrofit Volkhov alumina plant for processing of Kola nepheline concentrates but the works were interrupted by the war. Volkhov was converted to the process only in early 50-ies. After mastering at Volkhov, the technology was further improved by VAMI and used for construction of Pikalevo and Achinsk alumina refineries making them some of the most efficient alumina plants today [13].

2.2. Advantages and disadvantages of alkaline methods to process low grade ores

While the soda lime sintering process is the only industrially proven technology to process low grade ores (bauxites, nephelines, clays, etc.), using of this method to produce alumina from non-alkaline raw material (such as bauxite or clays) is not economic due to high energy intensity and capital costs. Only complex waste free processing of alkaline ores (nephelines, anorthosites) into alumina, soda, potash, cement, gallium and other sub products has proven to have production cost of alumina comparable with the production cost of Bayer processing of high quality bauxite due to the value of byproducts and reducing alumina transportation cost (if refinery is located close to the smelter). In order to realize this advantage, sufficient nepheline of suitable quality must be available along with local markets for all products and a cheap fuel source [13]. Capital cost for creation of

this industrial complex will be higher compared to the Bayer process for producing alumina only. Thus a potential project efficiency evaluation shall be made as an individual case, considering all these factors.

3. Acidic alumina recovery processes

Different methods of extracting alumina from non bauxite rocks largely including acid processes have been investigated in more than 20 countries. More than 16 different processes were tried in the period from 1910 till 1930 for local non bauxite sources (clay, leucite, alunite, cryolite, calcium aluminium phosphates, etc.) in United States, England, Italy, Germany and Japan [5].

During World War II the most critical task was to ensure security of domestic sources of alumina for aluminium producing countries leading to creation of 5 experimental plants to process anorthosite, kaoline, diasporic clay in USA. In Japan 13 different commercial plants were operated of which 4 operated acid processes although with low efficiency and alumina quality [14].

In 1960's – 1970's inflating price of bauxite inspired renewed research including R&D and industrial pilot studies by USBM, CSIRO, Péchiney, and Alcan. All were curtailed in the late 1980's due to worse economics (high energy and capital cost) when compared with the Bayer process, and/or lack of reliable solutions for some process areas.

In the former USSR, research on acid technology, production of alumina from kaolin clay and other non- bauxite ore began in the 20-ies of the last century and intensively developed until the discovery of deposits of Tikhvin and Ural bauxite and startup of their processing by conventional alkaline processes [9]. Further interest in acid technology was repeatedly renewed in 1970's leading to creation of proprietary technologies that have been brought to the stage of pilot testing. However, these studies were interrupted in the early 90's due to changes in the economic climate in USSR and Russia those days.

Some companies have recently made significant progress in the development of methods for the alumina production based on acidic technology. The most notable achievements belong to Canadian company Orbite Aluminae Inc. [15]. The company applied a range of advanced technology solutions of hydrochloric acid process to produce alumina and number of by-products (silica, hematite, and rare-earth metals) with HCl regeneration increasing thus the profitability of alumina production.

The most complete and detailed reviews of the alumina acid technologies were described both by Russian and foreign researchers [5, 16, 17]. The authors suggested the following systematic classification of the principal methods.

3.1. Sulfuric acid methods

The sulfuric acid processing of non-bauxite ore has considerable advantage, as far as the sulfuric acid is an effective and inexpensive reactant for leaching. The process was intensively investigated in USBM, VAMI, and Russian Academy of Science. An example of USBM flow sheet is presented in [17]. This method produced only crude alumina contaminated with iron that had to be refined again by a subsequent Bayer circle.

To improve the extraction of alumina in the solution and to achieve purification from iron in the sulfuric acid methods a variety of techniques were offered, some of them are described below. For example, the decomposition of the aluminum containing materials was performed by sintering with ammonium sulfate. Catalytic effect of additives like alkaline sulphates and sulfuric acid on the degree of sulphatization was established. The clays were initially calcined, and then subject to sulfatizing sintering with H₂SO₄. In optimal conditions, the extraction reached 90% of Al₂O₃. Purification from iron of the produced solution was offered by various ways, including solvent extraction using the di-(2-etipeksal) phosphoric acid in kerosene, and increasing pH by addition of NH₄OH from 0.1 to 0.2. Another way to purify solution from iron is to transfer Fe³⁺ into Fe²⁺ by sulfur dioxide gas with ~ 125 % of stoichiometrically required at 20-40 °C. During the subsequent heating of the solution to 160 °C basic aluminum sulfate is precipitated. A solution of sulfuric acid leaching of clay was purified from iron by sedimentation of as potassium jarosite in the hydrolysis of the reaction $3\text{Fe}_2(\text{SO}_4)_3 + \text{K}_2\text{SO}_4 + 12\text{H}_2\text{O} \rightarrow 6\text{H}_2\text{SO}_4 + \text{K}_2(\text{OH})_{12} \cdot \text{Fe}_6(\text{SO}_4)_4$. Part of the iron in the jarosite was partially replaced with aluminum because of their isomorphic substitution.

The purified aluminium sulfate solution was evaporated by 60% with crystallization of Al₂(SO₄)₃·nH₂O and calcination under 1100 °C, producing 99.9% alumina and generated gases were directed for sulfuric acid production. The aluminium sulfate crystallization normally produces fine crystal product with difficulty to be washed from mother liquor. Moreover it contains for 1 mol of Al₂(SO₄)₃ with 18 moles of H₂O and requires considerable heat for calcining. The sulfuric acid regeneration from the gas after aluminium sulfate decomposition is a complex and energy consuming process. Therefore aluminium sulfate recovery should be investigated using the hydrolysis of sulfate liquors under increased temperatures. The hydrolysis of sulfate liquor in an autoclave at temperatures up to 320 °C was reviewed in order to precipitate an insoluble basic aluminium sulfate.

Sulfuric acid and hydrochloric acid were jointly used in the H⁺ process developed by Aluminium Péchiney [18]. Alcan joined Aluminium Péchiney in building a pilot plant with 15 tons/day of Al₂O₃ capacity to demonstrate the possibility of its industrial implementation. The basic ideas of the process are presented at Fig. 1:

- 1) selective dissolution of alumina in a concentrated sulphuric acid solution that avoids the preliminary roasting of the ore;
- 2) conversion of aluminium sulphate into chloride with efficient purification of alumina in form of aluminium chloride hexahydrate;
- 3) the aluminum chloride separated from liquor was thermally decomposed generating alumina and hydrogen chloride directly recycled into the process. This alumina was of higher purity than the Bayer alumina.

In general the shortcomings of sulphuric acid method are: (i) difficulty to produce alumina free from iron and other impurities that required further alumina refining by alkali or use of hydrochloric acid; (ii) high energy consumption to decomposed 18-aqueous aluminium sulphate into alumina; (iii) it is not possible to regenerate sulfuric acid reactant without preliminary reconversion of SO₂ into SO₃.

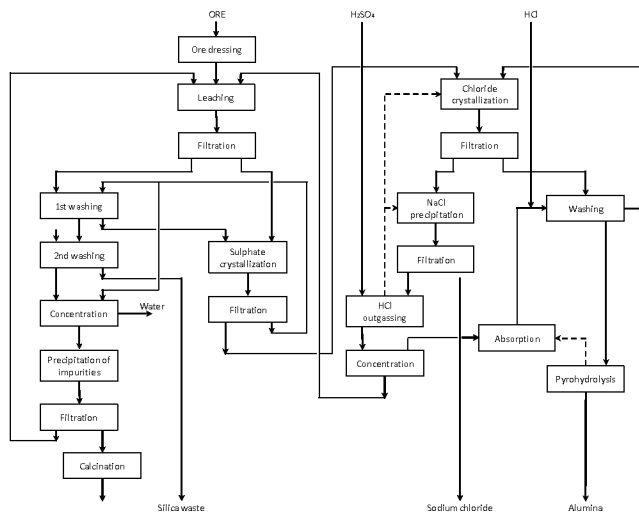


Fig. 1. H⁺ acid process for low quality ore processing into alumina

3.2. Nitric acid methods

The nitric acid methods can be implemented both in cyclic (recycling of the main reactant) and direct flow variants. In the last case for processing of alkali ores, for example nephelines, both alumina and nitric fertilizers may be produced. However the application of this approach is very limited and cannot be considered efficient. One of the cyclic schematic diagrams is presented on Fig. 2 [17].

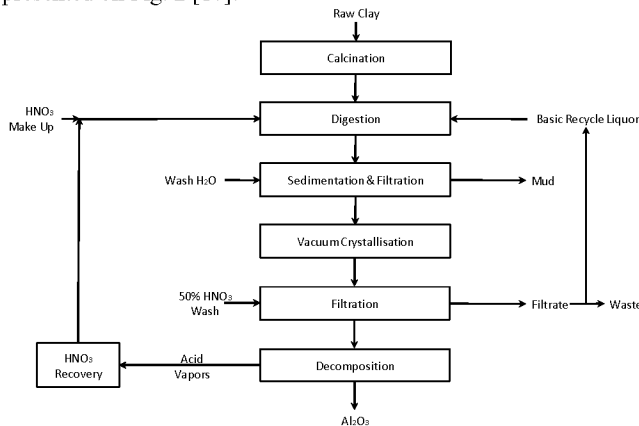


Fig. 2. Flow Chart for USBM Aluminium Nitrate/HNO₃ Leach Process for Alumina Extraction from Clay

A major shortcoming of cyclic nitric acid methods is high energy consumption for HNO₃ regeneration and the requirement to use special cleaning techniques for iron removal. Thus, to reduce the loss of working reagent the Kola ICTREMRM Institute of Russian Academy of science proposed using a powerful plasma reactor. However, despite the technical attractiveness and effectiveness of such solutions, the high energy consumption most probably would not make this method acceptable for industrialization.

3.4. Hydrochloric acid process

In recent years, much attention is paid to hydrochloric processing of shales, clay, lignite and other natural and man-made ore sources. This is due, first of all, to sufficient ease of HCl

regeneration, in spite of the increased requirements on the corrosion resistance of the equipment. Various options were studied for implementing the process for different types of raw materials. For 40 years from mid 1940's till mid 1980's USBM developed 6 potential processes including sulphuric, nitric and hydrochloric acid technology to produce alumina from clay, and tested them on specially constructed mini-plants starting from 1970's. The studies of different processes showed that hydrochloric acid leaching process of calcined clay followed by HCl gas sparging crystallization had the lowest capital and operating costs and had the greatest commercial potential of the 6 processes studied.

CSIRO Division of Mineral Chemistry has developed a process for the production of cell-grade alumina from high reactive silica bauxite using hydrochloric acid leaching [19]. The process is based upon leaching either calcined bauxite at atmospheric pressure or uncalcined bauxite under pressure, followed by crystallization of aluminium chloride hexahydrate using HCl sparging. Unlike other acid processes, effective iron separation is achieved in a single crystallization step. The aluminium chloride crystals are decomposed to alumina in a fluidized-bed and the hydrogen chloride produced is recycled. Having high energy consumption of 34.6 – 37.1 GJ/t this acid process was not considered to be a replacement for the Bayer process treating standard bauxite with low reactive silica. According to CSIRO assessment, when the reactive silica content approaches 10% then the economics favour the acid process.

In the 1960's VAMI developed and tested combined hydrochloric acid-alkaline technology (Fig 3). It allowed for the production of high quality alumina but was very high on energy due to the additional alkaline cycle. That is why the process was not industrially implemented. USSR started the construction of an industrial pilot plant with a capacity of up to 6,000 tons of aluminium chloride per year in Nikolayev (Ukraine) for chlorine electrolysis, but due to changes in the economic situation in the former USSR in the late 1980's, the project was not completed.

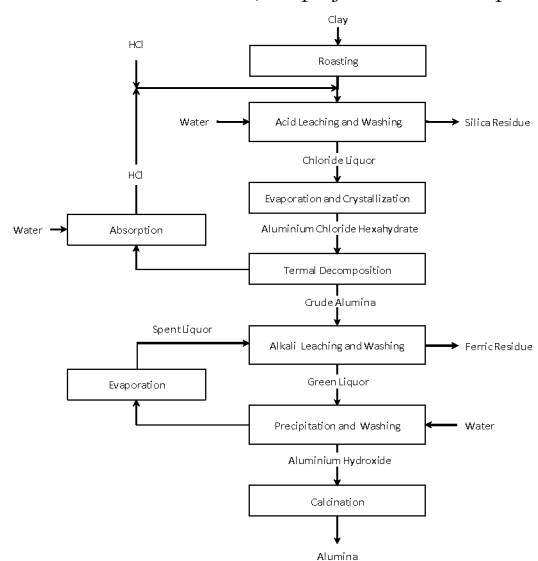


Fig. 3. Flow Chart for VAMI Combined technology of Alumina Extraction from Clay

At present the following optimized typical variant of flow sheet without alkaline tail is considered optimal (Fig 4).

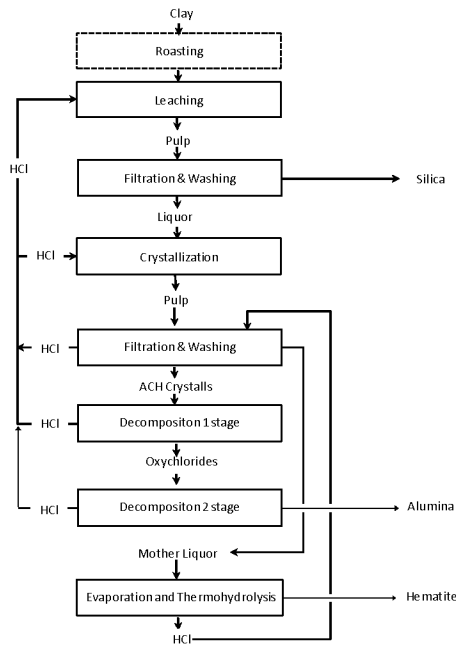


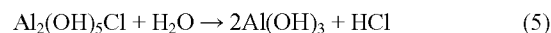
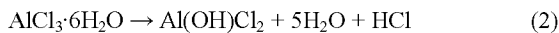
Fig. 4. Flow Chart for Hydrochloric Acid technology of Alumina Extraction from Clay

Recently, Orbite Aluminae Inc. has developed a hydrochloric acid technology using up-to-date advanced process and hardware solutions. One of the advantages of the technology helping in energy saving is using pressure leaching to avoid pre-calcination of ore. The technology was initially developed for local Grande-Vallée aluminium bearing clays (Quebec), but subsequently tested for other alumina bearing raw materials including bauxites, red mud and fly ash and tested at a pilot plant in Cap-Chat (Quebec). By the end of 2012 company has a project to convert the pilot plant into high purity alumina facility, while next step will be building the first commercial plant of 550 ktpa using this technology. UC RUSAL and Orbite Aluminae have discussions in connection with a possible partnership in this project, pursuant to which UC RUSAL would, among other things, offer process improvements based on its experience and know how.

During the last few years a number of interesting results have been obtained in our lab which allows reducing power consumption and improves the quality of the product. Thus, it has been established that thermal decomposition of aluminium chloride hexahydrate which is a version of its hydrolysis (thermal hydrolysis) which is commonly termed in technical literature as calcination and presented by an overall equation



is accomplished actually by a number of intermediate transformations of basic aluminium chlorides (oxychlorides) of different basicity and hydration degree. Some of these intermediate and not always successive reactions can be presented as follows:



Reaction (2) goes on with extraction of water in an amount which is excessive by stoichiometry to accomplish the overall reaction (1). However, according to the literature data and results of our own investigations the process ceases at a temperature of 100-500°C when the material is presented by a mixture of aluminium oxychlorides since water as a steam is rapidly blown away from the reaction zone without reaction with intermediate products of decomposition by reactions (3)–(5). Further transformations are feasible only when temperature is raised to 900-1000°C in the presence of flue gases. We developed conditions allowing the temperature to be reduced for the second stage of calcination and to obtain alumina with low content of chlorine and alpha phase.

Our laboratory studies showed that alumina obtained by a hydrochloric method is suitable for the production of aluminium in conventional cells. In spite of the fact that the crystal morphology (Fig. 5) differs from usual SGA, the values of bulk density and specific surface area correspond to those of conventional SGA while impurity content is considerably lower.

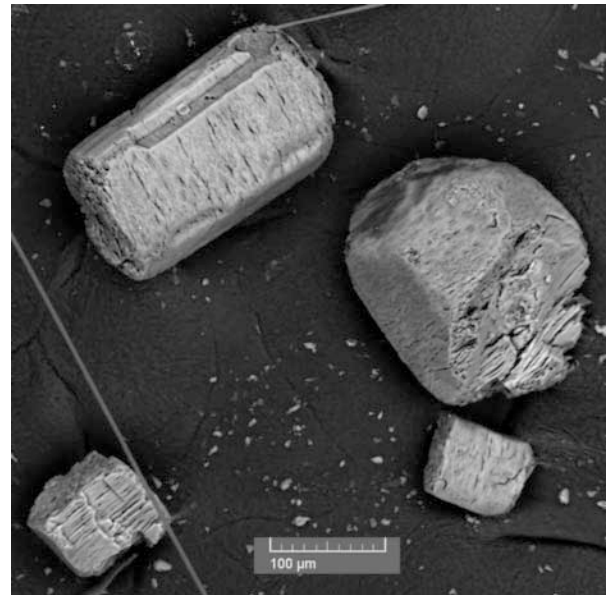


Fig. 5. Particles of calcined alumina produced by acid process

According to various authors the energy consumption to produce 1 ton of alumina in the processing of raw materials containing 20-50% Al_2O_3 by hydrochloric acid is 25-52 GJ. However, it can be significantly reduced using acid autoclave processing, without thermal activation of clay, by improving the crystallization process for $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ with a reduction of the number of stages, and decreased consumption of heat for the regeneration of hydrochloric acid. The results achieved in these areas are very encouraging. Orbite claims to achieve a degree of HCl regeneration exceeding 99 % while significantly reducing power consumption.

4. Benchmarking on Energy Efficiency

While benchmarking the energy consumption of different alumina processes is considered to be of primary importance. The total power consumption of an alumina plant (comprising consumptions of electrical energy and heat) are given below for different ores and processes.

Table 2
Benchmarking of energy efficiency of different alumina processes, RUSAL assessment

Technology	Ore	Power consumption, GJ/t alumina
Bayer process	Bauxites	8 - 18*
Hydrochloric acid processes	Clay	26 - 37
H ⁺ process	Clay	27 - 40**
Nitric acid processes	Clay	48 - 50**
Sulfuric acid processes	Clay	42 - 46**
Soda-lime sintering process	Nephelines, anorthosite	40 - 55

* or 13-23 GJ/t including energy required for production of caustic soda and lime used in the process; ** from [18]

As can be seen, energy efficiency of hydrochloric acid methods is still inferior to the Bayer process, which, however, can be more than compensated by the availability and low cost of raw materials, extraction of more components of ore in form of high purity products and, more importantly, by the reduced transport costs. For reference, in the RUSAL system the alumina supplied to a Siberian aluminum smelter from the nearby Achinsk Alumina Refinery using soda-lime sintering process has the lowest cost, due to lower transportation costs and production of soda ash and cement as by products of this technology, in spite of high energy and material intensity of the said production.

5. Conclusions

The growing interest in a number of countries to alternative alumina recovery processes from low grade ores is connected with establishing reliable domestic ore material base for their aluminum industry, saving on bauxite and alumina transportation costs and reducing the alumina cost using by-product revenues.

For processing of low grade bauxites ($3 < A/S < 5$) the Bayer-sintering combination methods are still the preferable industrially established technology.

Soda-lime sintering process is the preferable and commercially established technology for processing alkaline aluminosilicates (e.g. nephelines) assuring low self cost of alumina due to by-product value.

Sintering process is not economical to treat non-alkaline aluminosilicate ores, mainly due to high capital and operating cost, low content of alumina in the ore with prevailing silica content and absence of alkalis that can be produced as by-products. With reduction of silica and increase of alumina content of such materials (e.g. by beneficiation), sintering processes can find a limited application.

The most promising hydrochemical method to process the said low grade ores is using hydrochloric acid. The advantages of this method over other acids are: preferable capital and operation costs, and the possibility to produce alumina of higher quality compared with conventional cell grade alumina. The requirement for equipment with higher corrosive resistance at key process areas can be met with new materials and hardware solutions currently available. New innovative solutions shall be used to increase technical and economical efficiency of the process.

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