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Processing of Low-Quality Gibbsite-Kaolinite Bauxites

Symbat Dyussenova [1,*], Rinat Abdulvaliyev [1], Ata Akcil [2], Sergey Gladyshev [1] and Galiya Ruzakhunova [1]

[1] Institute of Metallurgy and Ore Beneficiation JSC, Satbayev University, Almaty 050010, Kazakhstan; rin-abd@mail.ru (R.A.); gladyshev.sergey55@mail.ru (S.G.); g.ruzahunova@satbayev.university (G.R.)
[2] Mineral-Metal Recovery and Recycling (MMR&R) Research Group, Mineral Processing Division, Department of Mining Engineering, Suleyman Demirel University, TR32260 Isparta, Turkey; ataakcil1@gmail.com
* Correspondence: dusenova_s@mail.ru

Abstract: The results of studies on the processing of gibbsite-kaolinite bauxite are presented. The developed technology includes preliminary chemical activation and thermal transformation during enrichment to obtain a concentrate suitable for processing by the Bayer method. As a result of the chemical activation of gibbsite-kaolinite bauxite in a solution of sodium bicarbonate, a change in the phase composition occurs, which made it possible to improve the results of gravity enrichment with the production of a coarse-grained gibbsite fraction. The transformation of bauxite in the temperature range of 900–1000 °C is explained by the decomposition reactions of siderite, gibbsite, kaolinite, calcite, dolomite and sodium ferro-sulfide oxide, as well as the formation of sodium aluminosilicate, hematite, quartz and the chemically stable phase of corundum. The optimum firing temperature of bauxite is 950 °C, after which, as a result of alkaline treatment during chemical enrichment, the extraction of SiO_2 into solution was 74.9%. A silicon modulus of enriched bauxite 10.9 units was obtained. As a result of the autoclave leaching of gibbsite-kaolinite bauxite after a two-stage enrichment, the maximum extraction of alumina into solution was 87.4%. The yield of red mud during the processing of bauxite enriched and calcined at 950 °C was 37.62%. During the autoclave leaching of bauxite without enrichment, the yield of red mud was 71%.

Keywords: roasting; bauxite; chemical activation; leaching; silica module

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1. Introduction

Alumina is produced all over the world with the alkaline method [1,2]. This is due to the simplicity of the method instrumentation, which does not require special grades of steels or other scarce materials. Industrial alkaline methods intended to produce alumina from bauxites, depending on the quality of the processed raw materials, are divided into the:

- hydrochemical method (Bayer method);
- sintering method;
- combined method (combination of the Bayer method and sintering method in parallel and sequential versions) [3].

The classical Bayer method is suitable only for high-quality bauxites with a silicon modulus greater than 7. The silicon modulus (μ_{Si}) of samples is determined from the relation of Al_2O_3/SiO_2. It is advisable to process bauxites with a silicon modulus less than seven by sintering or a combined method.

The sintering method is more versatile and can be applied to any high-silicon aluminum raw material. The main disadvantage of this technology is the high prime cost (320–330 USD per 1 ton Al_2O_3) of the obtained alumina, associated with high energy consumption, material consumption of equipment, losses of alkali and alumina with waste sludge.

Bauxites from various deposits differ significantly in their qualitative composition and quantitative content of components; therefore, various methods or processing methods

are used for them [3–5]. Processing methods are known for highly sideritized bauxites, including their roasting, cooling, magnetic separation, leaching and processing of red mud. The main advantage of these schemes is a good separation of bauxite components. The disadvantages are labor-intensive crushing, screening, magnetic separation, flotation processes and additional investment costs.

There is a hydrometallurgical caustification method for sideritized bauxites [6,7]. The essence of this method is in the fact that bauxite is subjected to soda-lime leaching using soda ash and limestone in autoclaves. However, the use of this method results in significant alumina losses due to the formation of tricalcium hydroaluminate.

One of the technological options intended to process sideritized bauxite is thermal causticization. The essence of the method is the roasting of bauxite at the temperature of ~900 °C in a tubular rotary kiln with soda in the required amount. A method of thermal conditioning for chloride-containing bauxites was proposed in [5] that enables the removal of carbonates by roasting with chlorides. This roasting method is selective, as only carbonate constituents are removed.

It was shown in [5–8] that the sintering method using calcium- and corundum-containing additives enables the processing of high-iron bauxites. However, the disposal of corundum-containing waste and the use of rotary sintering furnaces show that the proposed method is not promising. Low-quality bauxites can be processed by the separate leaching of clay and stony fractions of bauxites. However, the separation of bauxite fractions in an alkaline medium does not provide the required separation degree of the clay part [5,9–12].

The bauxites of the Krasnogorsk deposit currently used at the Pavlodar aluminum plant (hereinafter referred to as PAP) of Aluminum of Kazakhstan JSC (the Republic of Kazakhstan) in the production with the sequentially combined method named Bayer-sintering, are distinguished by a low silicon modulus and an increased content of harmful components: siderite, chamosite, hematite, pyrite, organic and other impurities, and their quality constantly decreases, which results in a sharp deterioration in the composition of solutions, middlings and a decrease in the technical and economic parameters [13]. With this circumstance, a complex of theoretical and technological studies is performed to develop an effective technology, since the above methods processing low-quality bauxite have a number of disadvantages that make it difficult or impossible under production conditions.

The optimal solution to involve the majority of bauxite deposits in Kazakhstan into the processing industry is our proposed technology intended to process low-quality gibbsite-kaolinite bauxite with a preliminary two-stage beneficiation, including innovative technical solutions that enable to remove harmful impurities, to transform the phase composition, to remove silicon during subsequent alkaline processing and to obtain material suitable for the alumina production in the simplest and most efficient Bayer process.

The proposed technology for processing low-quality gibbsite-kaolinite bauxites, with preliminary two-stage enrichment, makes it possible to obtain a material suitable for the production of alumina by the simplest and most efficient Bayer method.

2. Materials and Methods

X-ray fluorescence analysis was performed with a Venus 200 PANalytical B.V. spectrometer with wave dispersion (Malvern PANalytical B.V., Almelo, The Netherlands). Chemical analysis of the samples was performed on an optical emission spectrometer with inductively coupled plasma Optima 2000 DV (PerkinElmer, Waltham, MA, USA). X-ray experimental data were obtained with a Bruker D8 ADVANCE (Bruker Corporation, Billerica, MA, USA) apparatus using copper radiation at an accelerating voltage of 36 kW and a current of 25 mA. Thermodynamic calculations of the reactions accepted for analysis were performed using the HSC Chemistry special program (Outokumpu Oyj, Helsinki, Finland).

The initial data for calculations were obtained from the NIST Standard Reference Database 13 website [14,15]. Beneficiation (desiliconization) of the calcined coarse-grained fraction was performed in a 1800-mL thermostatted beaker while stirring with a mechan-

ical stirrer at 400 rpm in a solution containing 10% NaOH at the temperature of 100 °C, L:S = 6:1 (Liquid:Solid) for a duration of 2 h. The circulating solutions were mixed before the experiments by placing the canisters on the roller tables of the laboratory mill for 30 min.

The chemical activation and leaching of beneficiated bauxites were performed in 300-mL autoclaves installed in a thermostatically controlled tunnel with automatic temperature control 0–300 °C and stirring of the autoclaves over the head. This autoclave was designed and manufactured for laboratory research at the Institute of Metallurgy and Ore Beneficiation (Almaty, Kazakhstan).

Heat treatment was performed in a tubular rotary kiln that enables to create conditions for uniform roasting as a result of mixing (Zhengzhou SuTong Electric Equipment Company, Zhengzhou, China). The material of the reaction tube was stainless steel. Accepted designations: $\alpha_{ку}$—caustic modulus; μ_{Si}—silicon modulus; $Na_2O_{ку}$—caustic alkali; $Na_2O_{кб}$—carbonate alkali; and $Na_2O_{об}$—total alkalinity. The novelty of the technology lies in the two-stage enrichment of bauxite by gravity and chemical methods (Figure 1).

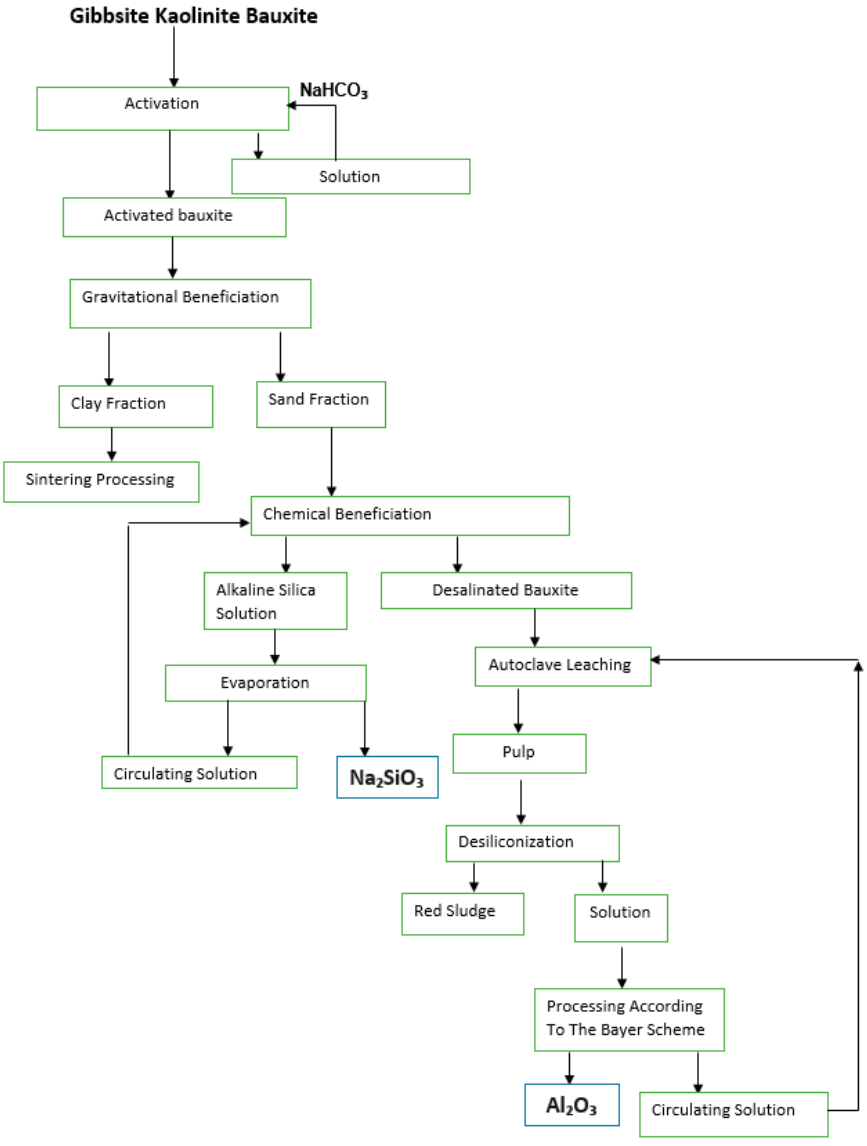


Figure 1. Two-stage beneficiation technology of bauxites.

Before gravity enrichment, preliminary chemical activation is conducted in a sodium bicarbonate solution. The selected coarse-grained fraction is sent for thermal transformation

followed by chemical enrichment in an alkaline solution. The clay fraction is sent to the sintering process of the main alumina production.

As a result of the performed operations, a conditioned product with a silicon modulus of more than 7 was obtained that meets the requirements for processing by the simplest and most economical Bayer method. In the process of research, the material composition of the original bauxite and the resulting middlings of the technology-bauxite after activation, clay and sand fractions, enriched bauxite, pulp, red mud, alumina and solutions were analyzed by XRD and chemical analysis.

3. Results and Discussion

3.1. Physical and Chemical Composition

A representative sample of gibbsite-kaolinite bauxite from the Krasnogorsk deposit was used for the study: % wt—Al_2O_3 42.0; SiO_2 11.5; Fe_2O_3 19.5; CaO 1.08; Na_2O 0.22; MgO 0.18; K_2O 0.03; TiO_2 2.05; SO_3 0.24; Cl-0.04; other 23.16; and μSi 3.65.

The X-ray phase bauxite composition is presented in Figure 2.

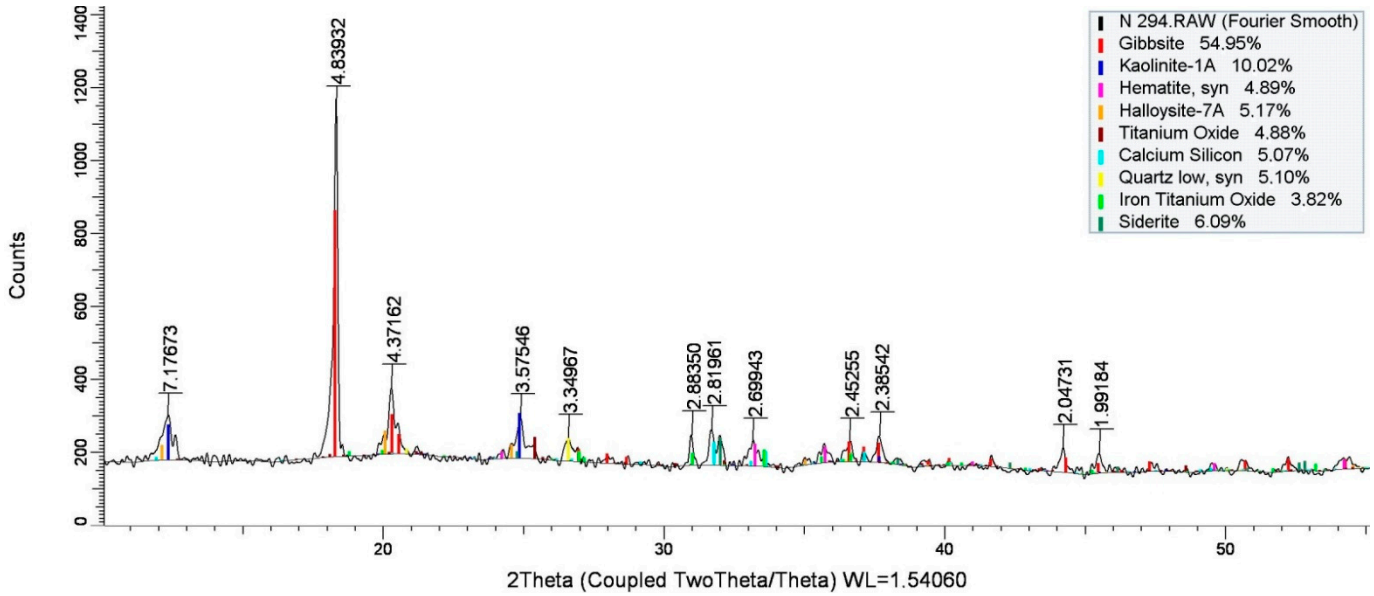


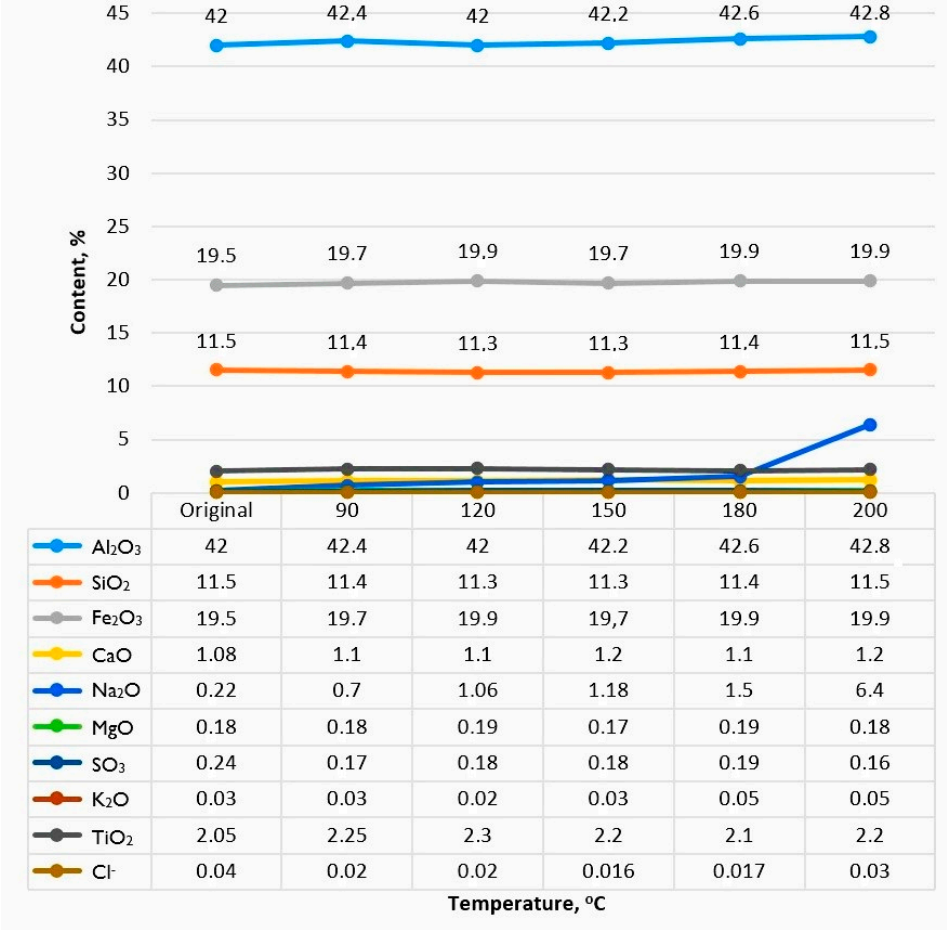
Figure 2. X-ray diffraction pattern of bauxite from the Krasnogorsk deposit. The data were previously given in the publication [16].

3.2. Chemical Activation

Chemical activation of bauxite before gravity beneficiation was performed in a sodium bicarbonate solution containing 120 g/dm^3 $NaHCO_3$ at temperatures of 90–200 °C [16,17]. The $NaHCO_3$ concentration was taken using the solubility limit. Changes in the chemical and phase compositions of bauxite depending on temperature and duration are shown in Figures 3–6 (Data from the publication [16] were used here).

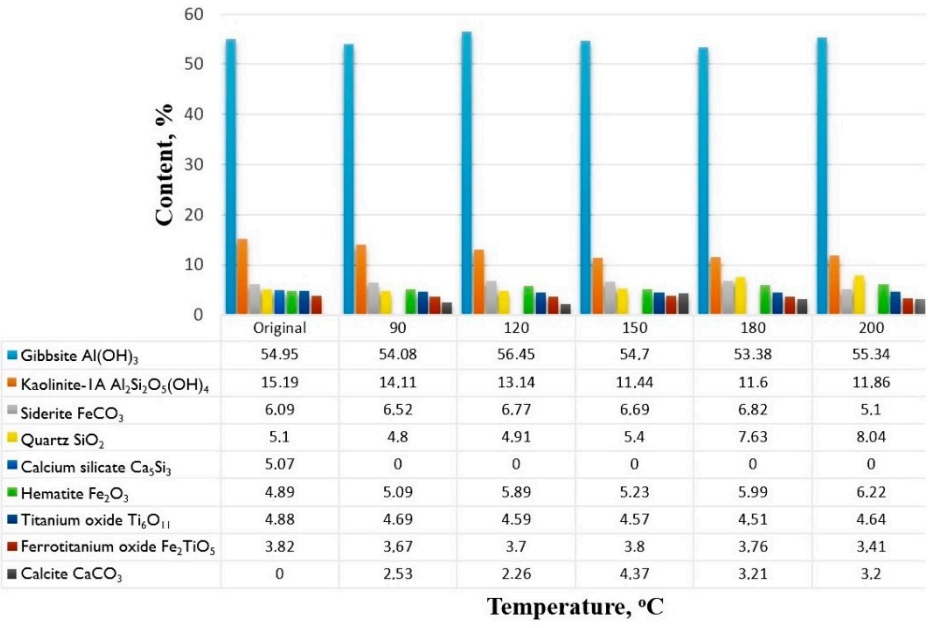
Studies of the influence of the chemical activation temperature on the change in the chemical and phase composition were performed for a duration of 60 min. The conditions were chosen based on previous studies [18]. Studies of the effect of the chemical activation duration were performed at the temperature of 120 °C, at which the maximum reduction in the hard-to-open kaolinite phase was obtained.

X-ray phase analysis results of the coarse-grained fraction of bauxite after chemical activation are presented in Figure 7.



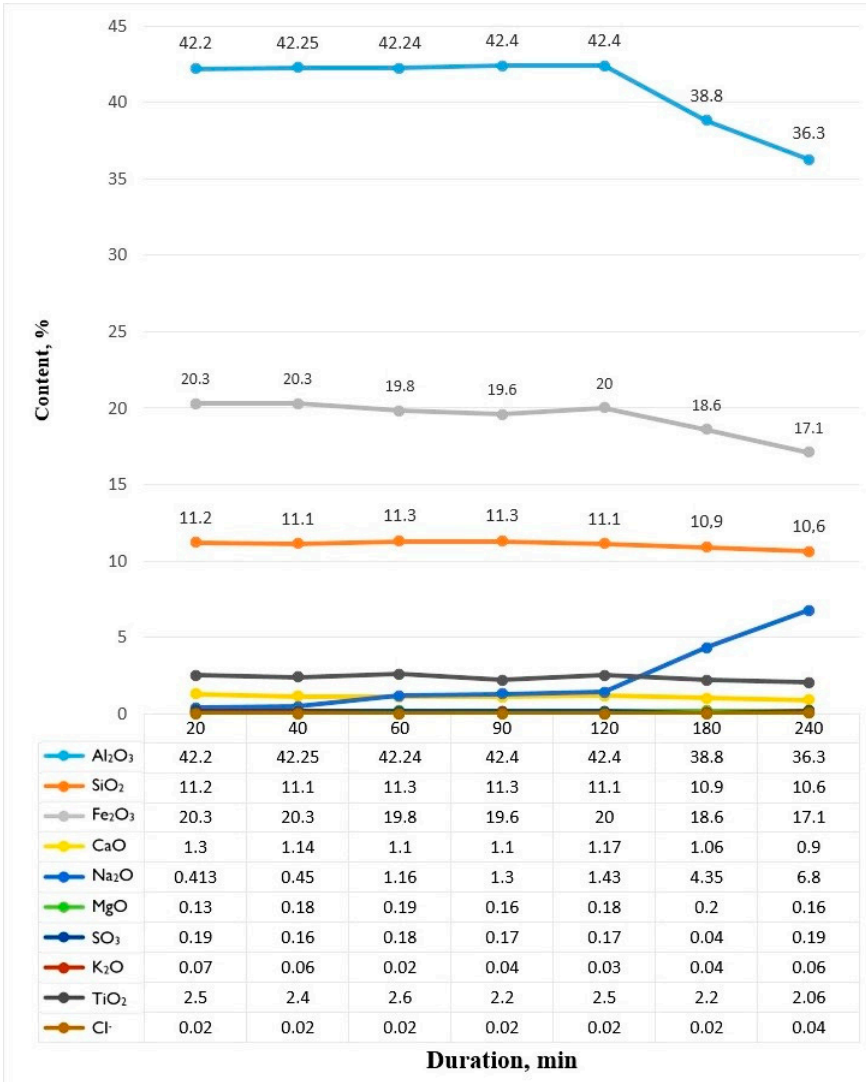
	Original	90	120	150	180	200
Al_2O_3	42	42.4	42	42.2	42.6	42.8
SiO_2	11.5	11.4	11.3	11.3	11.4	11.5
Fe_2O_3	19.5	19.7	19.9	19,7	19.9	19.9
CaO	1.08	1.1	1.1	1.2	1.1	1.2
Na_2O	0.22	0.7	1.06	1.18	1.5	6.4
MgO	0.18	0.18	0.19	0.17	0.19	0.18
SO_3	0.24	0.17	0.18	0.18	0.19	0.16
K_2O	0.03	0.03	0.02	0.03	0.05	0.05
TiO_2	2.05	2.25	2.3	2.2	2.1	2.2
Cl^-	0.04	0.02	0.02	0.016	0.017	0.03

Figure 3. The chemical composition of bauxite samples depending on the activation temperature.



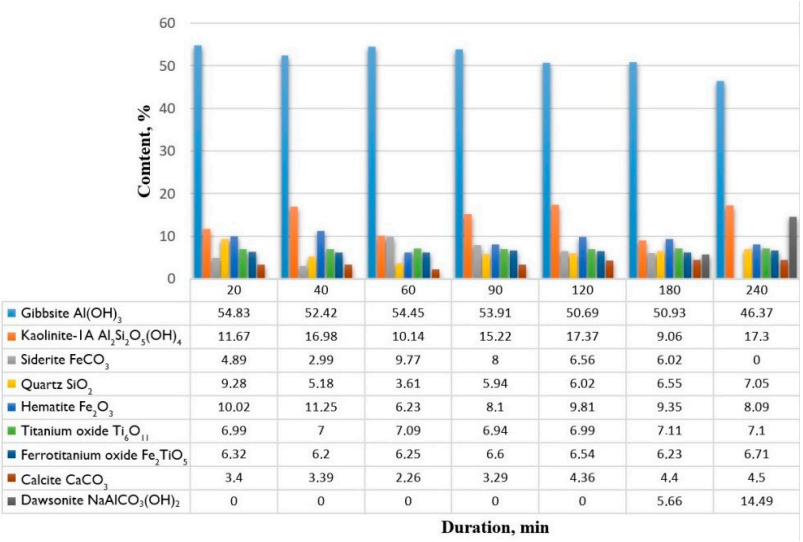
	Original	90	120	150	180	200
Gibbsite $Al(OH)_3$	54.95	54.08	56.45	54.7	53.38	55.34
Kaolinite-1A $Al_2Si_2O_5(OH)_4$	15.19	14.11	13.14	11.44	11.6	11.86
Siderite $FeCO_3$	6.09	6.52	6.77	6.69	6.82	5.1
Quartz SiO_2	5.1	4.8	4.91	5.4	7.63	8.04
Calcium silicate Ca_5Si_3	5.07	0	0	0	0	0
Hematite Fe_2O_3	4.89	5.09	5.89	5.23	5.99	6.22
Titanium oxide Ti_6O_{11}	4.88	4.69	4.59	4.57	4.51	4.64
Ferrotitanium oxide Fe_2TiO_5	3.82	3.67	3.7	3.8	3.76	3.41
Calcite $CaCO_3$	0	2.53	2.26	4.37	3.21	3.2

Figure 4. Phase composition of bauxite samples depending on the temperature of chemical activation.



	20	40	60	90	120	180	240
Al_2O_3	42.2	42.25	42.24	42.4	42.4	38.8	36.3
SiO_2	11.2	11.1	11.3	11.3	11.1	10.9	10.6
Fe_2O_3	20.3	20.3	19.8	19.6	20	18.6	17.1
CaO	1.3	1.14	1.1	1.1	1.17	1.06	0.9
Na_2O	0.413	0.45	1.16	1.3	1.43	4.35	6.8
MgO	0.13	0.18	0.19	0.16	0.18	0.2	0.16
SO_3	0.19	0.16	0.18	0.17	0.17	0.04	0.19
K_2O	0.07	0.06	0.02	0.04	0.03	0.04	0.06
TiO_2	2.5	2.4	2.6	2.2	2.5	2.2	2.06
Cl^-	0.02	0.02	0.02	0.02	0.02	0.02	0.04

Figure 5. The chemical composition of bauxite samples depending on the duration of activation at the temperature of 120 °C.



	20	40	60	90	120	180	240
Gibbsite $Al(OH)_3$	54.83	52.42	54.45	53.91	50.69	50.93	46.37
Kaolinite-1A $Al_2Si_2O_5(OH)_4$	11.67	16.98	10.14	15.22	17.37	9.06	17.3
Siderite $FeCO_3$	4.89	2.99	9.77	8	6.56	6.02	0
Quartz SiO_2	9.28	5.18	3.61	5.94	6.02	6.55	7.05
Hematite Fe_2O_3	10.02	11.25	6.23	8.1	9.81	9.35	8.09
Titanium oxide Ti_6O_{11}	6.99	7	7.09	6.94	6.99	7.11	7.1
Ferrotitanium oxide Fe_2TiO_5	6.32	6.2	6.25	6.6	6.54	6.23	6.71
Calcite $CaCO_3$	3.4	3.39	2.26	3.29	4.36	4.4	4.5
Dawsonite $NaAlCO_3(OH)_2$	0	0	0	0	0	5.66	14.49

Figure 6. The phase composition of bauxite samples depending on the duration of chemical activation at the temperature of 120 °C.

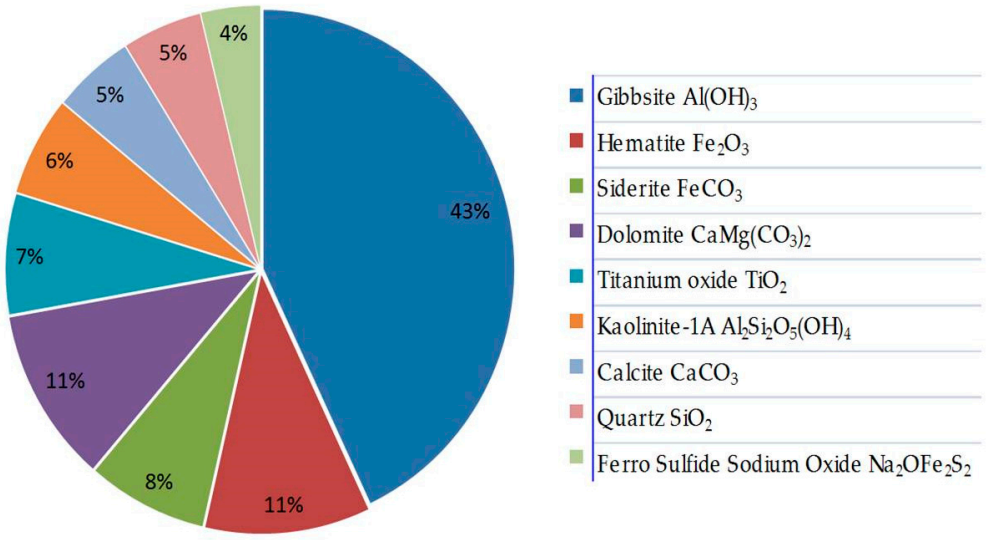


Figure 7. Phase composition of the coarse-grained fraction of bauxite.

3.3. Preliminary Roasting and Chemical Beneficiation

The coarse-grained fraction of bauxite after gravity beneficiation was subjected to thermal transformation to obtain a high-quality bauxite concentrate with a silicon modulus of more than 7 suitable for processing according to the Bayer scheme. The transformation included preliminary roasting at the temperature of 900–1000 °C for 2 h. The technology provides for the use of sintering furnaces, which are installed at the PAP. The operating temperature of the furnaces at the inlet is 900–950 °C. The duration was chosen taking into account the time spent on heating the material and bringing it to the temperature of the sintering zone, 1000 °C [19].

The phase composition of bauxite depending on the roasting temperature is specified in Figure 8.

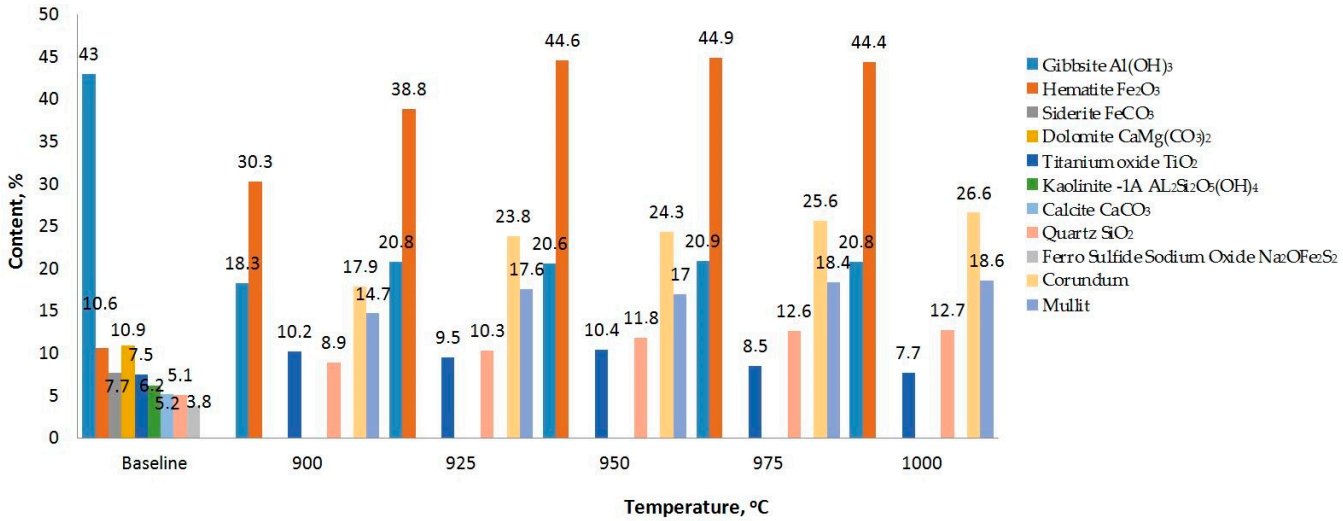


Figure 8. The phase composition of bauxite depending on the roasting temperature.

The mechanism of transformation of the phase composition can be represented as the following reactions based on the results of bauxite roasting:

$$Na_2O \cdot 2FeS + 3O_2 \rightarrow Na_2O + 2FeO + 2SO_2 \qquad \Delta G = -1434.9 - 0.436\ T \tag{1}$$

$$Na_2O + 2Al(OH)_3 + 2SiO_2 \rightarrow Na_2O{\cdot}Al_2O_3{\cdot}2SiO_2 + 3H_2O \quad \Delta G = 18.81 - 0.119\ T \tag{2}$$

$$FeCO_3 \rightarrow FeO + CO_2 \quad \Delta G = 79.64 - 0.179{\cdot}T \tag{3}$$

$$4FeCO_3 + O_2 \rightarrow 2Fe_2O_3 + 4CO_2,\ \Delta G = -265.44 - 0.442 \cdot T \quad (4)$$

$$Fe_2O_3 \rightarrow 2FeO + 0.5O_2,\ \Delta G = 292 - 0.692\ T \quad (5)$$

$$2Al(OH)_3 \rightarrow Al_2O_3 + 3H_2O \quad (6)$$

$$Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O \overset{500–600\ ^oC}{\rightarrow} Al_2O_3 \cdot 2SiO_2 + 2H_2O \quad (7)$$

$$2(Al_2O_3 \cdot 2SiO_2) \overset{850–925\ ^oC}{\rightarrow} 2Al_2O_3 \cdot 3SiO_2 + SiO_2 \quad (8)$$

$$2Al_2O_3 \cdot 3SiO_2 \overset{1000\ ^oC}{\rightarrow} 2(Al_2O_3 \cdot SiO_2) + SiO_2 \quad (9)$$

The phases of siderite, gibbsite, kaolinite, calcite, dolomite and ferro-sulfide sodium oxide disappear as a result of roasting. A chemically stable phase of corundum is formed. The contents of hematite, quartz and mullite increase. The increase in the content of quartz can be explained by the decomposition of kaolinite with the formation of mullite in accordance with Equations (7)–(9).

The appearance of a chemically stable phase of corundum enables to prevent the transition of Al_2O_3 into solution during chemical beneficiation and to ensure the maximum removal of silica. Chemical beneficiation of calcined bauxite was performed with an alkaline solution containing 10% NaOH at the temperature of 100 °C for 2 h and L:S = 6. We used the factory alkaline solution of PAP with a content of 496 g/l Na_2O_{ky} to obtain an alkaline solution containing 10% NaOH. The chemical beneficiation results of calcined bauxites are shown in Table 1 and Figure 9.

Table 1. The composition of bauxite after chemical beneficiation (with Extraction of SiO_2 into solution).

Bauxite Roasting Temperature, °C	Content, %										Extraction of SiO_2 into Solution, %
	Al_2O_3	SiO_2	Fe_2O_3	CaO	SO_3	Na_2O	Cl	TiO_2	Other	μ_{Si}	
900	45.77	10.27	22.49	2.03	0.61	3.99	0.01	3.05	11.76	4.45	55.2
925	52.72	9.36	26.58	1.95	0.48	3.76	0.01	3.13	2.01	5.63	56.0
950	58.06	5.33	27.12	2.31	0.12	0.49	0.01	3.3	0.08	10.9	74.9
975	57.47	5.37	27.14	2.25	0.09	0.53	0.01	3.14	0.01	10.6	74.6
1000	60.35	5.69	27.49	2.1	0.09	0.59	0.01	3.44	0.03	10.6	74.3

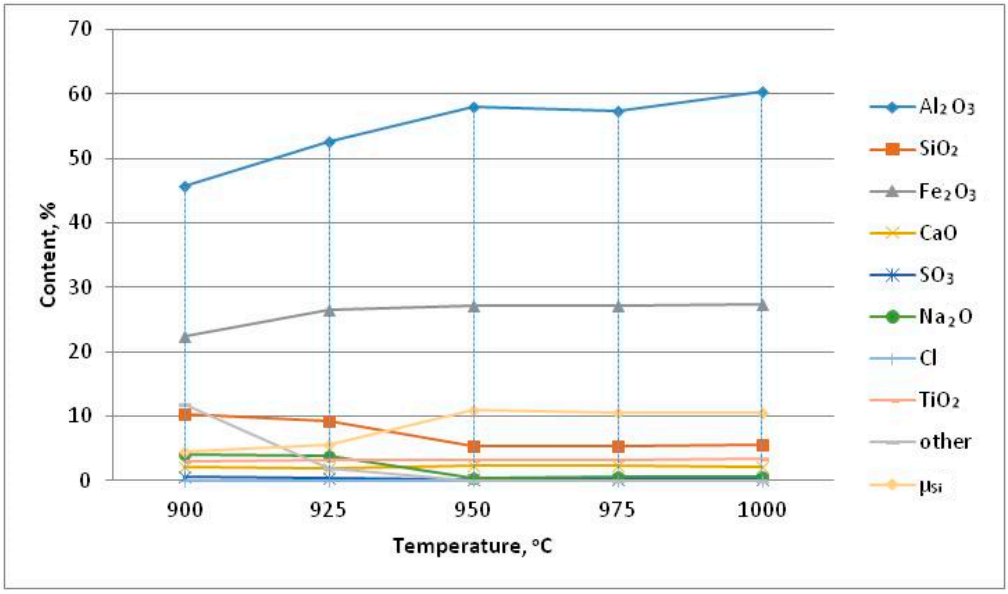


Figure 9. The composition of bauxite after chemical beneficiation.

The dependence of the change in the silicon modulus of bauxite and the degree of extraction of SiO_2 into the solution as a result of chemical beneficiation on the roasting temperature are shown in Figure 10.

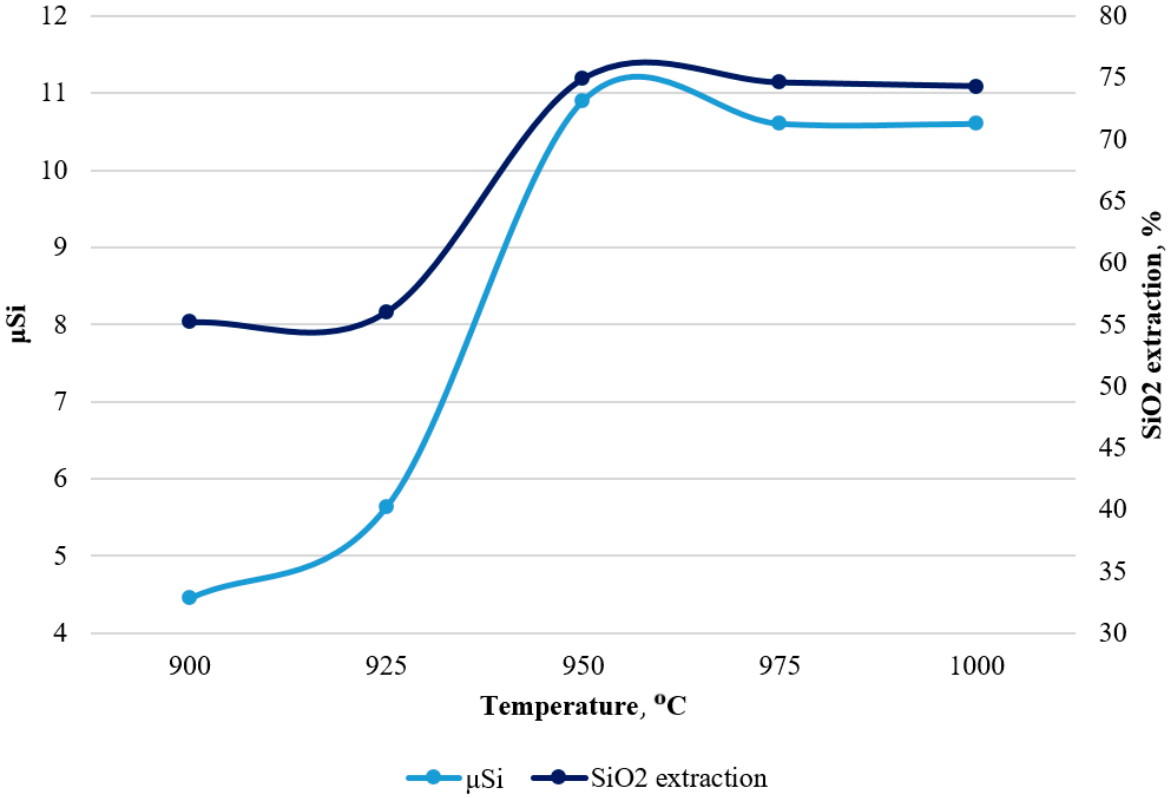


Figure 10. Changes in the silicon modulus (μ_{Si}) of bauxite and the extraction degree of SiO_2 (legend right) into solution as a result of chemical beneficiation.

It follows from the obtained chemical beneficiation results that a roasting temperature of 950 °C is sufficient to obtain a silicon modulus of bauxite of more than 7 units, while the extraction of SiO_2 into the solution was 74.9%. Autoclave leaching of beneficiated bauxite was performed at the temperature of 280 °C during 2 h in a recycled alkaline-aluminate solution of PAP composition, wt g/dm^3: 123.9 Al_2O_3; 326.6 $Na_2O_{об}$; 6.4 $Na_2O_{кб}$; 320.2 $Na_2O_{ку}$; and $\alpha_{ку}$-3.099. The circulating solution was dosed based on the calculation of a solution with $\alpha_{ку}$-1.48 to be obtained. The autoclave leaching pulp was diluted with distilled water to obtain an aluminate solution for decomposition with a content of $Na_2O_{ку} \approx 120$ g/dm^3.

Agitation mixing of the diluted pulp of autoclave leaching was performed at the temperature of 105 °C for 2 h. The compositions of aluminate solutions and solid phases (red mud) of autoclave leaching of bauxite after pulp dilution are presented in Tables 2 and 3 and Figures 11 and 12.

Table 2. The compositions of aluminate solutions (with $\alpha_{ку}$ and μ_{Si}).

Bauxite Roasting Temperature, °C	Content, g/dm^3					$\alpha_{ку}$	μ_{Si}
	Al_2O_3	$Na_2O_{об}$	$Na_2O_{кб}$	$Na_2O_{ку}$	SiO_2		
900	116.325	132.5	9.5	123.0	1.0	1.74	116
925	118.3	132.8	9.9	122.9	0.99	1.70	119.4
950	130.8	133.04	11.04	122.0	0.88	1.54	140
975	133.9	133.4	11.3	122.1	0.85	1.50	148
1000	133.5	134	11.5	122.5	0.820	1.51	158

Table 3. The compositions of solid phases (red mud) with μ_{Si} and Al_2O_3 extraction.

Bauxite Roasting Temperature, °C	Solids Content, %								μ_{Si}	Extraction into Solution Al_2O_3, %
	Al_2O_3	SiO_2	Fe_2O_3	CaO	SO_3	Na_2O	Cl	TiO_2		
900	21.403	21.869	30.986	1.556	0.2	16.95	2.771	3.623	0.98	66.1
925	20.31	15.73	34.15	2.8	0.3	16.74	2.58	3.45	1.21	70.3
950	14.1	10.65	54.5	3.11	0.5	15.82	2.306	3.554	1.29	87.8
975	12.7	10.7	54.22	3.2	1.09	15.01	2.35	3.45	1.505	88.1
1000	15.0	11.4	54.8	3.8	0.8	12.44	1.401	4.009	1.89	87.0

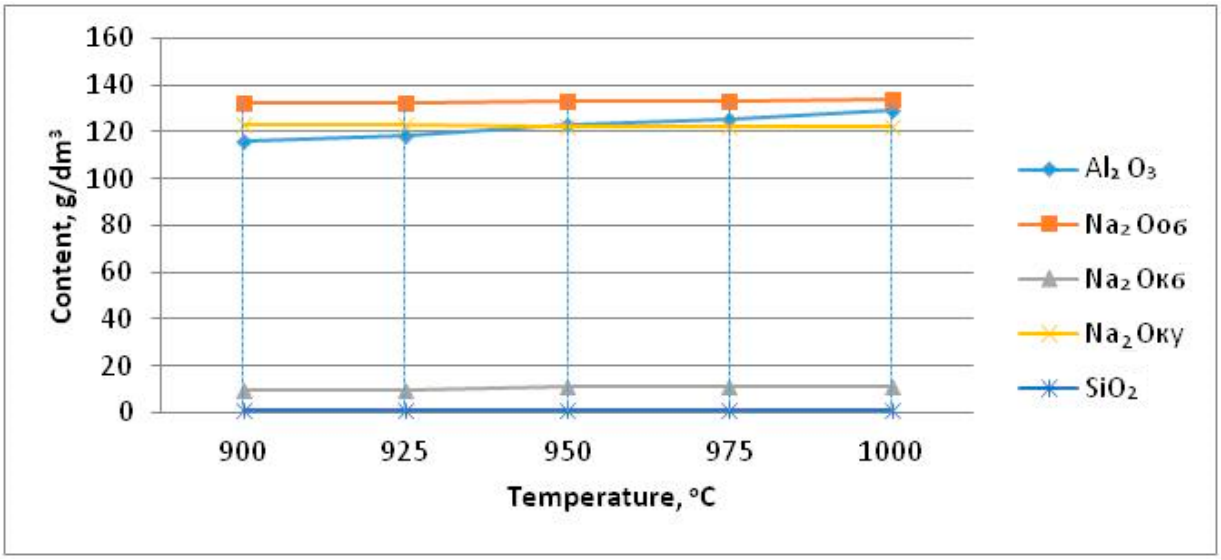


Figure 11. The compositions of aluminate solutions.

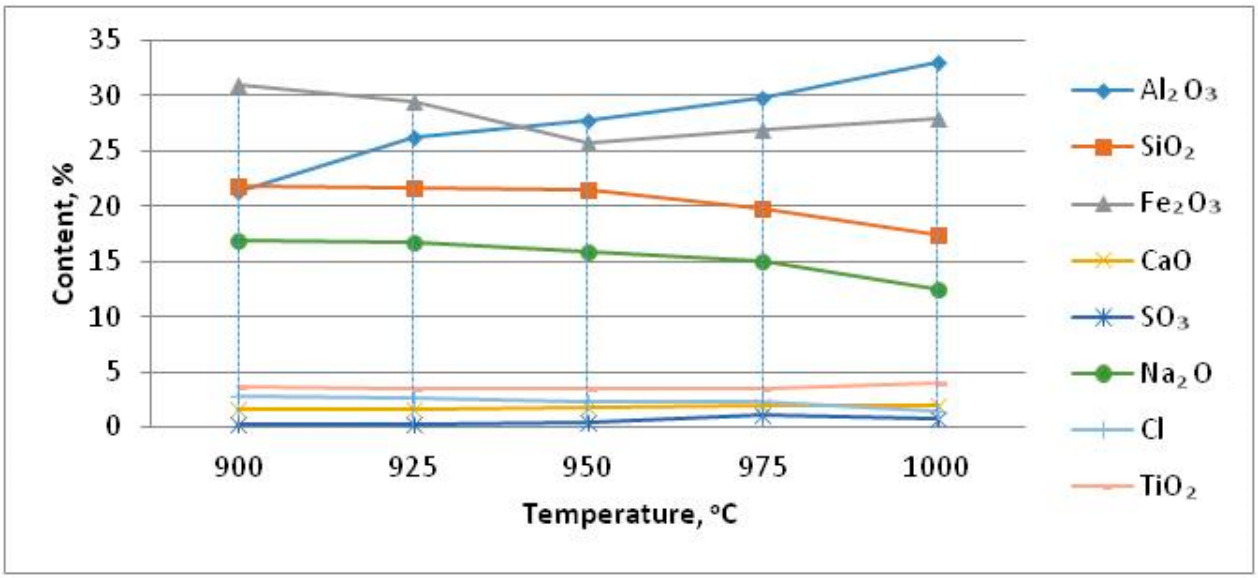


Figure 12. The compositions of aluminate solutions.

Analysis of the autoclave leaching results for pre-calcined bauxite showed that the optimal conditions are the roasting temperatures of bauxite 950–975 °C, while the alumina extraction into the solution was 87.8–88.1%. A higher roasting temperature resulted in an increase in the proportion of the sparingly soluble Al_2O_3 phase, respectively, to a decrease in extraction.

The yield of red mud during the processing of bauxite enriched and calcined at 950 °C was 37.62%. During autoclave leaching of bauxite under the accepted conditions without enrichment, the yield of red mud was 71%, i.e., carrying out preliminary enrichment significantly reduces the amount of environmentally harmful waste from alumina production—red mud.

The technology uses methods of gravitational enrichment with the separation of an environmentally friendly product—kaolinite (clay) fraction and chemical enrichment in a recycled alkaline solution, which is regenerated with the release of extracted silica into a commercial product (for example, liquid glass).

4. Conclusions

The technology was developed to process gibbsite-kaolinite bauxite, including preliminary chemical activation and thermal transformation during beneficiation, to obtain a concentrate suitable for processing by the Bayer method. A change in the phase composition occurs as a result of the chemical activation of gibbsite-kaolinite bauxite in a sodium bicarbonate solution that made it possible to improve the gravity beneficiation results with the production of a coarse-grained gibbsite fraction.

The chemical activation mode is selected from the conditions excluding the formation of an undesirable phase—dawsonite. The bauxite transformation within 900–1000 °C is represented by the reactions of decomposition of siderite, gibbsite, kaolinite, calcite, dolomite and ferro-sulfide sodium oxide, as well as the formation of sodium aluminosilicate, hematite, quartz and the chemically stable phase of corundum.

The optimum roasting temperature of bauxite is 950–975 °C, after which a bauxite silicon modulus of more than 7 units was obtained as a result of alkaline treatment during chemical beneficiation. The extraction of SiO_2 into solution was 74.9%. The maximum extraction of alumina into a solution of 87.8–88.1% was obtained as a result of autoclave leaching of gibbsite-kaolinite bauxite after a two-stage beneficiation.

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