

Article

Processing of Alumina Production Red Mud Sinter to Produce Solutions for Carbonation

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Abstract

The present study addresses the first stage in the development of a processing technology for low-grade high-silica bauxites aimed at producing solutions suitable for carbonation. The proposed process includes Bayer leaching of bauxite to obtain sodium aluminate liquor and red mud, followed by sintering of the red mud with limestone and soda ash. The resulting sinter is then leached with a recycled soda-caustic solution to produce an alkaline aluminate liquor and a residue. The alkaline aluminate liquor obtained from sinter leaching is subjected to carbonation to precipitate aluminum hydroxide, which is subsequently used as seed material during the decomposition of Bayer sodium aluminate liquor. The proposed technology reduces the volume of aluminate liquor directed to the Bayer decomposition stage while maintaining aluminum hydroxide yield, thereby improving the efficiency of the decomposition process.

Keywords: red mud; sintering; leaching; alkali; soda ash; sodium aluminate solution; aluminum hydroxide; carbonation

1. Introduction

The Bayer process is the simplest and most economical method for processing bauxite ores. It is based on the leaching of bauxite with recycled sodium aluminate liquor having a caustic ratio (α_k) of approximately 3.0, producing sodium aluminate solutions with a caustic ratio of 1.45–1.50 suitable for aluminum hydroxide precipitation through decomposition. During this process, silica is converted into sodium aluminosilicate hydrate (SASH), which constitutes the major component of the insoluble residue known as red mud (RM) that is subsequently disposed of in tailings storage facilities [1–7]. However, the Bayer process is economically feasible only for bauxites with a high silica modulus ($\mu_{Si} > 7$), where the silica modulus is defined as the ratio of alumina to silica content. During the processing of low-grade high-silica bauxites ($\mu_{Si} = 3\text{--}4$), losses of both alumina and alkali become excessive. Therefore, such ores are usually subjected to multistage mechanical and chemical beneficiation to upgrade them to Bayer-grade feedstock or are processed by the sintering method or combined Bayer–sintering technologies [8–16].



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The sintering process involves roasting bauxite with limestone and soda ash in rotary kilns at temperatures of 1100–1200 °C. Under these conditions, silica reacts with calcium oxide to form insoluble dicalcium silicate ($2\text{CaO}\cdot\text{SiO}_2$). The sinter is subsequently leached using a mixture of recycled process water and recycled soda-caustic liquor to produce sodium aluminate liquor with a caustic ratio of 1.45–1.50. Aluminum hydroxide is then recovered from the solution by decomposition [17]. Despite its effectiveness, this technology requires high fuel and electricity consumption, substantial operating and maintenance costs, and is associated with significant atmospheric emissions.

An alternative hydrometallurgical approach proposed by Ponomarev and Sazhin [2] replaces RM sintering with high-temperature pressure leaching in autoclaves at 280–300 °C using concentrated high-modulus sodium aluminate liquor ($\text{Na}_2\text{O}_{\text{caustic}} > 450 \text{ g/dm}^3$, $\alpha_k = 30$) with the addition of lime to promote the formation of calcium monosilicate ($\text{CaO}\cdot\text{SiO}_2\cdot 2\text{H}_2\text{O}$). However, this process has not been industrially implemented because of the lack of corrosion-resistant high-pressure equipment and its extremely high thermal energy demand.

At the Pavlodar Aluminium Plant (PAP) Aluminium of Kazakhstan JSC, a combined Bayer–sintering process is employed. In this technology, all bauxite feed is first processed through the Bayer circuit. The resulting red mud is separated from the sodium aluminate liquor and subsequently sintered with limestone and soda ash at 900–1100 °C [2]. After leaching, the sinter produces sodium aluminate liquor, which is directed to decomposition, while the remaining residue is discarded as waste unsuitable for further utilization. During sinter leaching, harmful impurities accumulate in the sodium aluminate liquor, negatively affecting the quality of the final alumina product.

The limitations of the existing technology employed at the PAP for processing low-grade high-silica bauxites necessitate further process improvement.

This paper presents the results of the first stage of the proposed technology, focusing on the determination of the optimum conditions for leaching RM sinter with a recycled soda-caustic solution to produce an alkaline aluminate liquor suitable for carbonation.

The scientific novelty of the proposed technology for processing low-grade bauxites lies in the leaching of red mud sinter using recycled soda-caustic solutions to produce alkaline aluminate liquors for the recovery of aluminum hydroxide by carbonation. The precipitated aluminum hydroxide can subsequently be used as seed material during the decomposition of Bayer sodium aluminate liquor.

Separate processing of the aluminate liquor obtained from the sintering route reduces the volume of aluminate liquor directed to the Bayer decomposition stage while maintaining the aluminum hydroxide yield, thereby improving the efficiency of the decomposition process.

2. Materials and Methods

The experimental methodology was based on an integrated approach to the processing of alumina production RM according to the technological flowsheet shown in Figure 1.

The proposed technology for processing RM sinter consists of the following stages:

- Leaching of the RM sinter using recycled soda–caustic liquor to produce sodium aluminate liquor suitable for carbonation and a solid residue for disposal;
- Autoclave desilication of the sodium aluminate liquor, producing a desilication residue that is recycled to the sintering stage;
- Deep desilication of the sodium aluminate liquor using a limestone slurry (CaCO_3), producing a residue that is discarded;
- Carbonation of the sodium aluminate liquor to produce aluminum hydroxide and regenerated recycled soda–caustic liquor. The present study focuses on the first stage of the proposed process, namely the leaching of RM sinter.

The elemental composition of the samples was determined by X-ray fluorescence spectroscopy (XRF) using an Axios PANalytical spectrometer (Malvern Panalytical, Almelo, The Netherlands) equipped with a 1 kW Rh anode X-ray tube. The phase composition of the samples was identified by X-ray diffraction (XRD) analysis.

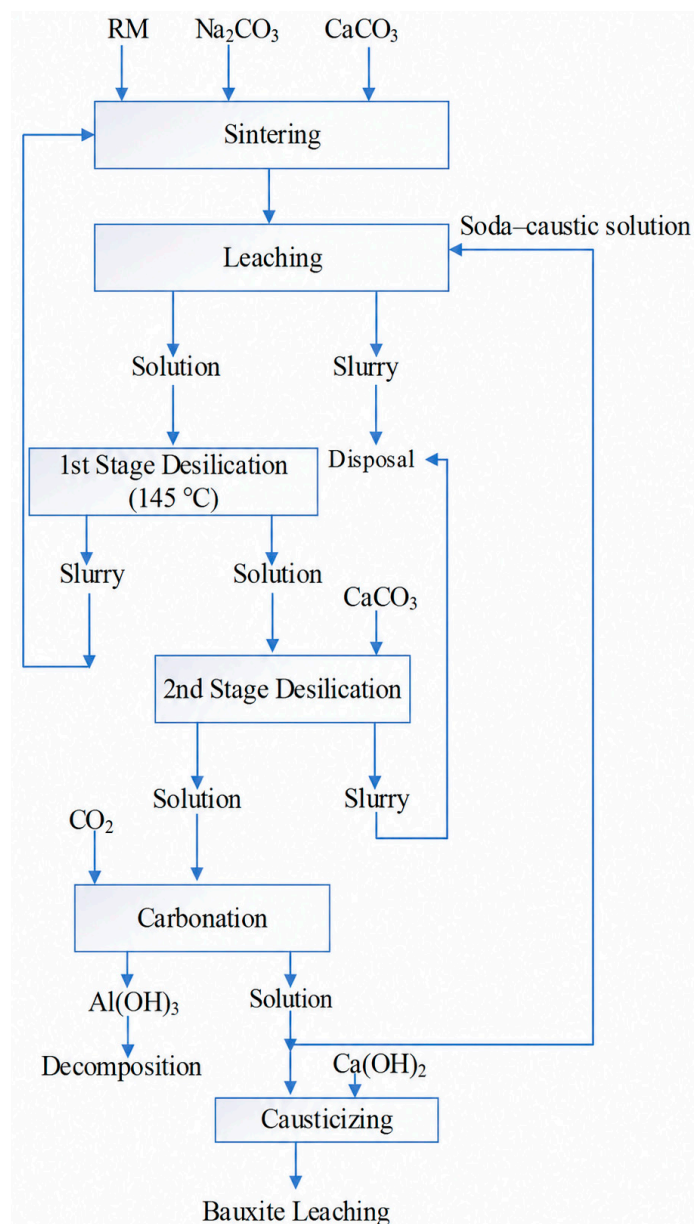


Figure 1. Process Flowsheet for the Carbonation Processing of Sodium Aluminate Liquor Obtained from RM Sinter Leaching.

The sinter was leached using recycled soda-caustic liquor obtained from the carbonation process with a total alkali concentration ($\text{Na}_2\text{O}_{\text{total}}$) of 150–200 g/dm³. The ratio of caustic alkali ($\text{Na}_2\text{O}_{\text{caustic}}$) to carbonate alkali ($\text{Na}_2\text{O}_{\text{carbonate}}$) was varied from 30:70 to 50:50, while the leaching temperature ranged from 30 to 100 °C, a liquid-to-solid ratio (L/S) of 2–10, and the leaching time from 30 to 180 min. The soda-caustic solutions were prepared by mixing reagent-grade NaOH and Na_2CO_3 in the required proportions.

The caustic modulus of the aluminate solution was calculated using the following formula [18]:

$$\alpha_{\text{ky}} = 1.645 \times \frac{\text{Na}_2\text{O}_{\text{ky}}}{\text{Al}_2\text{O}_3}$$

Notation:

$\text{Na}_2\text{O}_{\text{total}}$ —total alkali content;

$\text{Na}_2\text{O}_{\text{ku}}$ —caustic alkali content;

$\text{Na}_2\text{O}_{\text{ku}}$ —carbonate alkali content.

To ensure the reliability of the results, all experiments were performed in triplicate or quadruplicate, and the reported values represent the corresponding averages. Experimental data were processed by calculating material balances, product yields, and recovery efficiencies of the target components. The obtained results were used to analyze the distribution of elements among the different phases and to establish the optimum processing conditions.

3. Results

The objective of this study was to determine the optimum conditions for leaching RM sinter with recycled soda-caustic liquor to produce sodium aluminate liquor with a αk of 5–6, suitable for subsequent processing by the carbonation method.

The feed material used in this study was RM sinter produced from sodium carbonate and limestone at the PAP.

The chemical composition of the RM sinter (wt.%) was as follows: Na_2O , 16.10; Al_2O_3 , 11.50; SiO_2 , 12.19; Fe_2O_3 , 17.13; CaO , 20.03; TiO_2 , 3.17; V_2O_5 , 0.05; Ga_2O_3 , 0.011; SO_3 , 1.18; Cl , 0.145; and other products (o.p.), 18.494.

The phase composition of the RM sinter determined by X-ray diffraction (XRD) is presented in Figure 2.

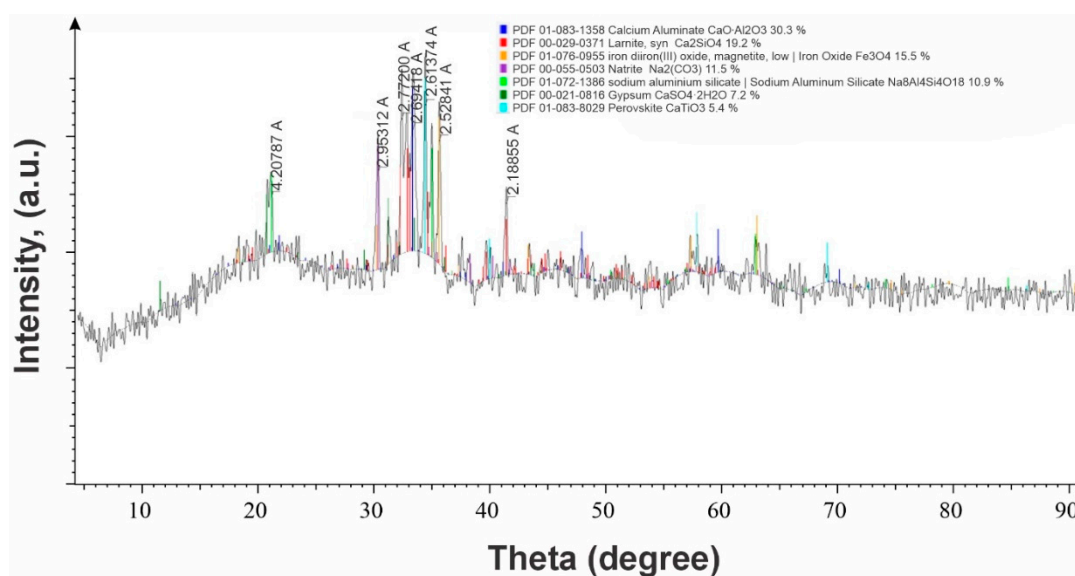


Figure 2. X-ray diffraction (XRD) pattern of the RM sinter.

The extraction of Al_2O_3 and SiO_2 from red mud sinter into the aluminate liquor depends on both the content of soluble aluminate phases in the sinter and the occurrence of secondary reactions leading to the formation of insoluble compounds. The resulting calcium and sodium hydroaluminosilicates exhibit low solubility in alkaline aluminate liquors, and their formation is responsible for the loss of alumina and alkali from the liquor to the residue.

The extent of these secondary reactions depends on the leaching conditions, including the composition of the soda-caustic solution, temperature, leaching time, and the L/S ratio of the pulp.

The effect of the total alkali concentration $\text{Na}_2\text{O}_{\text{total}}$ in the range of 150–250 g/dm³ on the extraction of Al_2O_3 and SiO_2 into solution was investigated at 50 °C with a leaching time of 30 min and L/S ratios between 3 (Figure 3, Table 1, experiments 1–4).

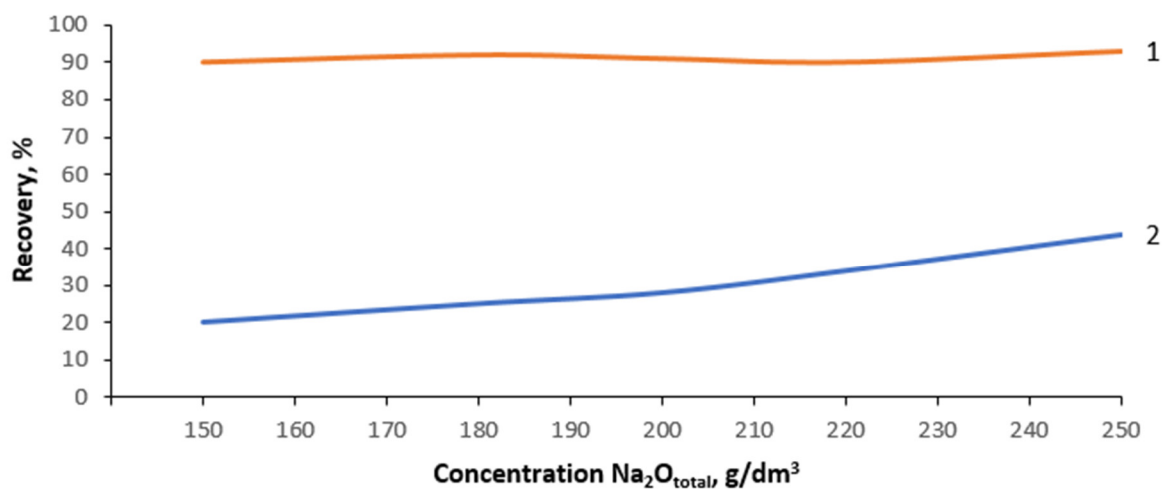


Figure 3. Extraction of Al_2O_3 and SiO_2 as a function of the total alkali concentration $\text{Na}_2\text{O}_{\text{total}}$: 1— Al_2O_3 ; 2— SiO_2 .

Table 1. Phase composition of the red mud sinter leaching residue as a function of leaching conditions.

Experiment No	Leaching Conditions				Content, X-Ray Phase Composition of Cake, %								
	Content Na ₂ O, %	Temperature, °C	Duration, min	Concentration Na ₂ O _{total} , g/dm ³	Ratio W:T	Dicalcium Silicate 2CaO · SiO ₂	Hydroaluminate Calcium 3CaO·2Al ₂ O ₃ · 4H ₂ O	Calcium Aluminate CaO·Al ₂ O ₃	Hydrogarnet 3CaO·Al ₂ O ₃ · Fe(SiO ₄)(OH) ₈	Sodium Hydroalumino­silicate Na ₂ O· Al ₂ O ₃ · 2SiO ₂ · 2H ₂ O	Calcite CaCO ₃	Perovskite CaTiO ₃	Magnetite Fe ₃ O ₄
1	50	50	30	150	3.0	44.6	14.2	12.3	-	-	5.1	12.4	16.5
2			60			38.5	18.3	14.1	-	-	-	12.3	16.8
3			120			36.1	19.8	15.1	-	-	-	12.2	16.8
4			180			34.3	20.4	16.4	-	-	-	12.3	16.6
5		75	30			38.2	20.6	11.8	-	-	-	12.7	16.7
6			60			37.7	21.0	12.0	-	-	-	12.5	16.8
7			120			26.1	22.3	14.3	-	8.1	-	12.5	16.7
8			130			13.5	24.7	15.2	-	11.8	5.7	12.5	16.6
9		100	30			36.9	of 14.9 the 12.9	-	18.9	-	-	12.7	16.6
10			60			31.5	12.9	-	31.3	-	-	12.5	16.8
11	70	50	120	200	3.0	14.9	12.9	-	31.2	5.6	6.1	12.5	16.8
12			180			11.8	13.2	-	31.3	6.8	7.9	12.3	16.7
13		75	30			41.5	10.4	11.6	7.3	-	-	12.3	16.8
14			60			40.4	11.1	10.8	8.8	-	-	12.3	16.7
15			120			40.0	12.1	9.9	9.0	-	-	12.4	16.6
16			180			34.0	9.7	-	-	-	7.1	11.7	38.0
17		100	30			-	-	-	53.3	-	-	15.6	31.1
18			60			-	-	-	-	52.8	-	21.3	24.2
19			120			40.1	9.5	9.6	6.4	5.6	-	12.3	16.5
20			180			27.4	11.3	6.0	8.8	7.1	10.7	12.3	16.4
21	30	50	30	250	3.0	15.6	15.8	5.1	11.1	10.2	13.5	12.5	16.7
22			60			41.8	-	9.4	-	19.6	-	12.5	16.7
23			120			19.8	-	-	-	15.2	35.9	12.5	16.6
24			180			-	18.8	-	-	7.9	44.2	12.4	16.7
25		75	30			40.9	14.8	10.0	-	-	4.9	12.5	16.7
26			60			36.3	13.2	14.8	-	-	6.5	12.5	16.7
27			120			33.3	13.0	16.5	-	-	8.3	12.4	16.5
28			180			32.6	12.8	17.2	-	-	8.4	12.5	16.5
29		100	30			31.4	12.6	17.5	-	-	9.5	12.6	16.4
			60			-	-	-	-	-	-	-	-

The results indicate that, under the selected experimental conditions, Al_2O_3 recovery is practically independent of the total alkali concentration within the investigated range of $\text{Na}_2\text{O}_{\text{total}}$ 150–250 g/dm³, remaining at 90–92%. In contrast, increasing the total alkali concentration significantly enhances the dissolution of SiO_2 , with its extraction increasing from 22% to 45%.

The leaching of RM sinter in a soda-caustic solution represents a complex multicomponent system. The leaching performance is influenced by numerous factors, one of the most important being the concentration of the soda-caustic leaching solution. The experimental results showed that increasing the alkali concentration had only a minor effect on Al_2O_3 extraction, whereas SiO_2 extraction increased noticeably. The observed behavior can be explained by the chemical and phase composition of the leaching residues. The analyses revealed that the total content of aluminum-bearing phases in the residues remained nearly constant, ranging from 29 to 31 wt.%. These phases determine the residual Al_2O_3 content in the residue after leaching. The presence of calcium aluminate ($\text{CaO} \cdot \text{Al}_2\text{O}_3$), originally contained in the sinter, indicates that the leaching process was not completed under the investigated conditions. The hydrate phase $3\text{CaO} \cdot 2\text{Al}_2\text{O}_3 \cdot 4\text{H}_2\text{O}$ is formed through the reaction of calcium aluminate with the alkaline solution (Reaction 1). The hydrogarnet phase, $3\text{CaO} \cdot \text{Al} \cdot \text{Fe} \cdot \text{SiO}_4 \cdot (\text{OH})_8$, results from the partial substitution of aluminum by iron in leaching solutions containing 200–250 g/dm³ Na_2O . The weak dependence of Al_2O_3 extraction on the Na_2O concentration indicates that alkali concentration is not the controlling factor governing alumina recovery. Instead, other process parameters, such as temperature and leaching time, are likely to have a greater influence.

The dissolution of SiO_2 is associated with the decomposition of dicalcium silicate ($2\text{CaO} \cdot \text{SiO}_2$) according to Reaction (2). The experimental results indicate that the extent of this reaction increases with increasing Na_2O concentration in the leaching solution.

The effect of temperature on the extraction of Al_2O_3 and SiO_2 was investigated using a leaching solution containing 150 g/dm³ total alkali ($\text{Na}_2\text{O}_{\text{total}}$) with a caustic alkali content ($\text{Na}_2\text{O}_{\text{caustic}}$) of 50% at a leaching time of 30 min (Figure 4, Table 1, Experiments 1, 5, and 9).

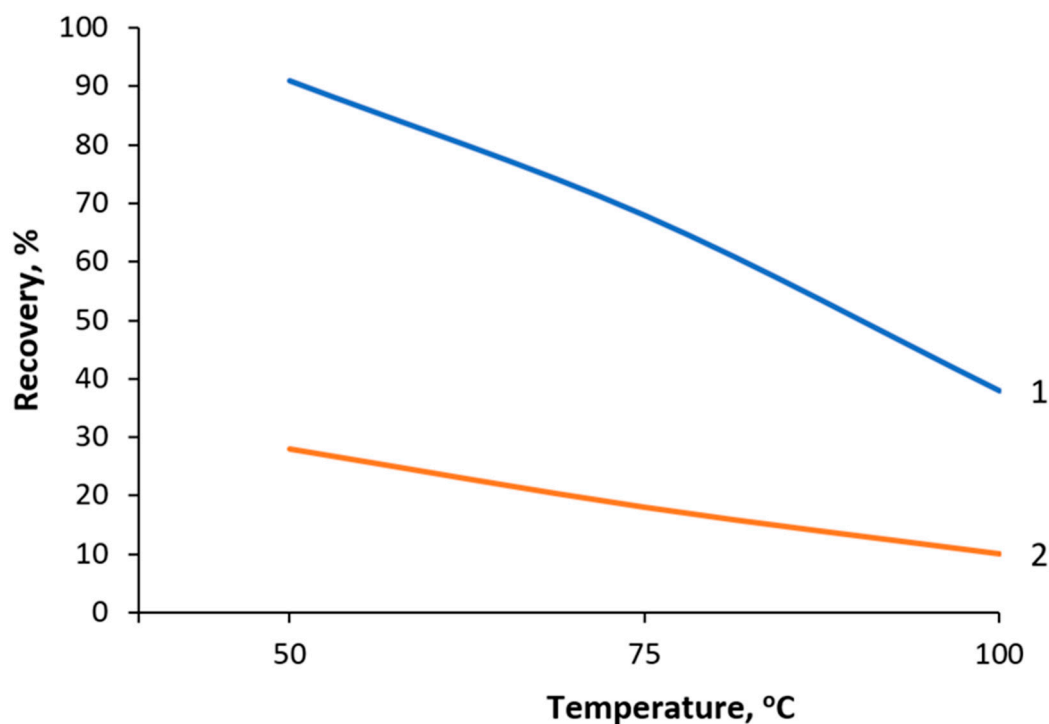


Figure 4. Extraction of Al_2O_3 and SiO_2 as a function of temperature: 1— Al_2O_3 ; 2— SiO_2 .

The decrease in Al_2O_3 extraction with increasing leaching temperature is attributed to the occurrence of secondary reactions leading to the formation of tricalcium hydroaluminate (Reaction 1) and hydrogarnet. The extraction efficiency of Al_2O_3 is governed by the amount of these sparingly soluble aluminum-bearing compounds remaining in the residue (Table 1). A similar relationship between Al_2O_3 extraction and leaching temperature has been reported under industrial operating conditions at the PAP, where leaching is carried out at temperatures not exceeding 85 °C in order to minimize alumina losses caused by secondary reactions [2]. At temperatures below 85 °C, however, the viscosity of the aluminate liquor increases, making solid–liquid separation more difficult. In the present study, the leaching temperature can be reduced because the recycled soda-caustic solutions used are less concentrated than conventional Bayer liquors.

Subsequent experiments aimed at determining the optimum leaching conditions were carried out using a soda-caustic solution containing 150 g/dm³ total alkali ($\text{Na}_2\text{O}_{\text{total}}$) with a caustic alkali content ($\text{Na}_2\text{O}_{\text{caustic}}$) of 50%, at a temperature of 50 °C and a leaching time of 30 min.

The effect of leaching time on the extraction of Al_2O_3 and SiO_2 into the aluminate liquor is presented in Figure 5 and Table 1 (Experiments 1–4).

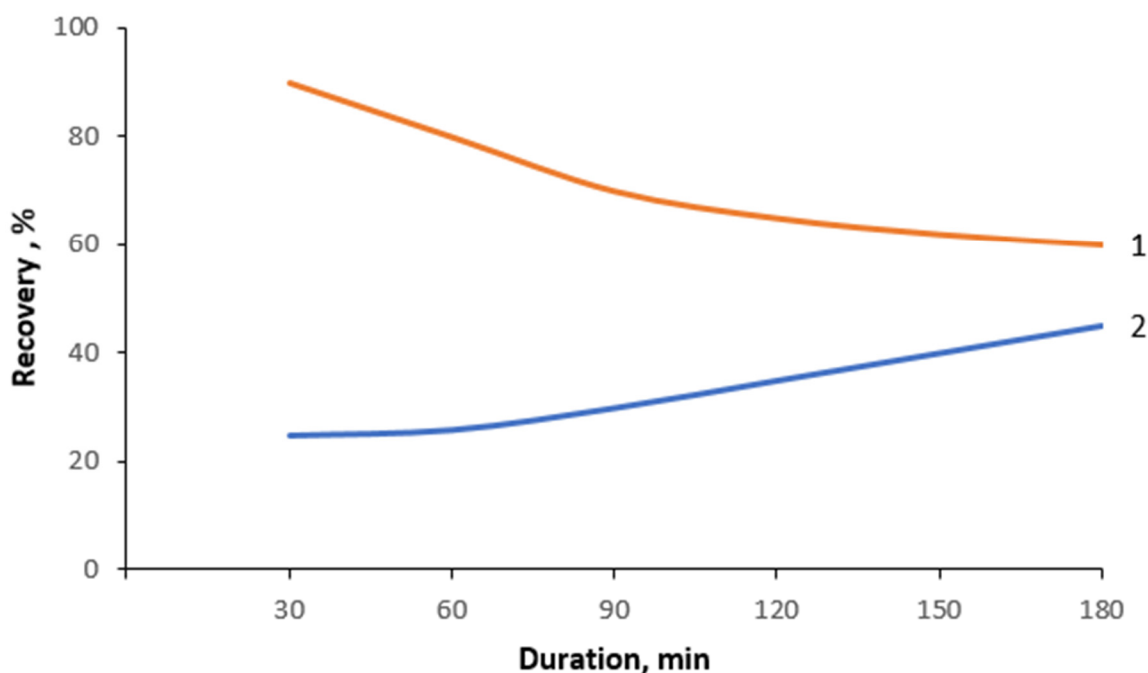


Figure 5. Extraction of Al_2O_3 and SiO_2 as a function of leaching time: 1— Al_2O_3 ; 2— SiO_2 .

The obtained results indicate that increasing the leaching time leads to a gradual decrease in Al_2O_3 recovery, while simultaneously increasing the dissolution of SiO_2 into the solution. The latter is undesirable because it adversely affects the quality of the sodium aluminate liquor and complicates its subsequent processing. The highest Al_2O_3 recovery was achieved at a leaching time of 30 min. A similar trend was observed at leaching temperatures of 75 °C (Experiments 5–8) and 100 °C (Experiments 9–12).

The effect of the L/S ratio in the leaching pulp on the extraction of Al_2O_3 and SiO_2 is presented in Figure 6 and Table 1 (Experiments 25–29). The L/S ratio determines the volume of solution that must be processed during subsequent stages of the process.

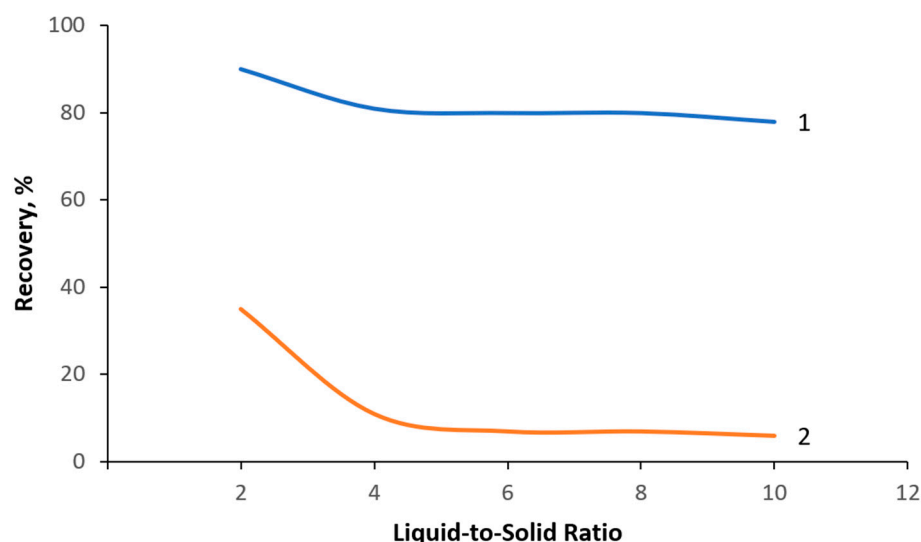
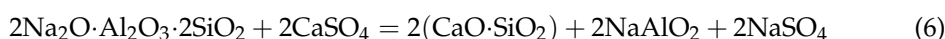
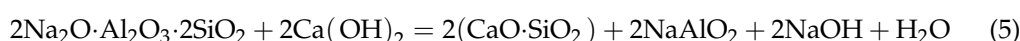
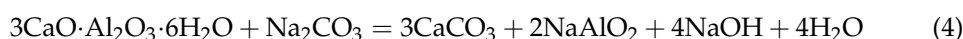
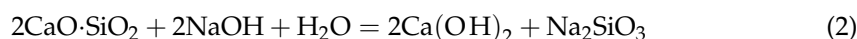


Figure 6. Extraction of Al₂O₃ and SiO₂ as a function of the L/S ratio: 1—Al₂O₃; 2—SiO₂.

The highest extraction of both Al₂O₃ and SiO₂ was obtained at an L/S ratio of 2:1. This behavior can be attributed to the higher concentration of dissolved reaction products in the leach liquor, which promotes their interaction with the mineral phases of the RM sinter.

The phase composition of the RM sinter leaching residues was investigated by X-ray diffraction (XRD) analysis (Table 1).

Based on the phase composition of the sinter and the leaching residues, the interaction between the components of the soda-caustic solution–sinter system can be described by the following reactions:



The X-ray diffraction analysis of the leaching residues confirmed the formation of phases consistent with the proposed Reactions (1)–(8), thereby establishing a relationship between the phase composition of the solid products and the extraction of Al₂O₃. The results indicate that Al₂O₃ extraction is governed by the total content of aluminum-bearing phases remaining in the leaching residue as a result of the secondary reactions (1)–(8). The smallest variation in the total content of aluminum-bearing phases was observed when the total alkali concentration (Na₂O_{total}) was varied, amounting to only 3.5%. In contrast, variations in leaching time and the L/S ratio resulted in changes of 26.0%, whereas changes in temperature produced a variation of 54.0%. These findings indicate that temperature is the most significant factor controlling the dissolution of aluminum-bearing phases and, consequently, the extraction of Al₂O₃.

The investigation of RM sinter leaching demonstrated that the optimum process parameters were a leaching temperature of 50 °C, a leaching time of 30 min, a L/S ratio of 2:1, and a soda-caustic solution containing 150–200 g/dm³ total alkali (Na₂O_{total}) with a caustic alkali content (Na₂O_{caustic}) of 50%.

Under the optimum leaching conditions, an aluminate liquor with the following composition (g/dm³) was obtained: 125 Na₂O_{caustic}, 115 Na₂O_{carbonate}, 240 Na₂O_{total}, 34.25 Al₂O₃, 1.10 SiO₂, 0.02 Ga₂O₃, and 0.045 V₂O₅, with a caustic ratio of 6.0 and a desilication coefficient (μ Si) of 31.1. The corresponding leaching residue had the following composition (wt.%): 0.798 Na₂O, 4.97 Al₂O₃, 15.6 SiO₂, 20.4 Fe₂O₃, 35.64 CaO, 8.93 TiO₂, 0.01 Ga₂O₃, 0.045 V₂O₅, and o.p.13.607. The Al₂O₃ extraction into the aluminate liquor reached 92%.

The X-ray diffraction (XRD) pattern of the leaching residue is shown in Figure 7.

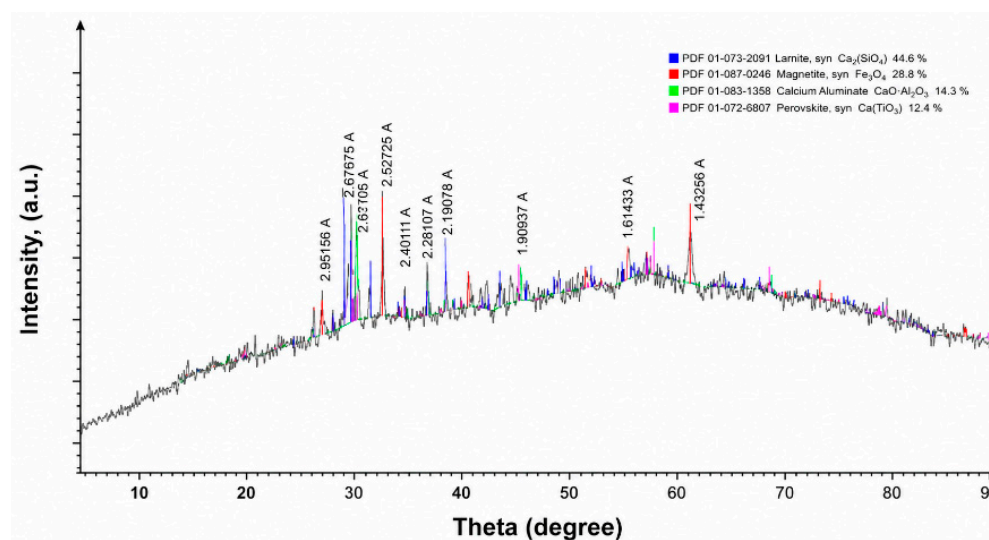


Figure 7. X-ray diffraction (XRD) analysis of the sinter leaching residue (leach cake).

According to the obtained results, leaching of the sinter under the optimal conditions in a soda–alkaline solution led to a decrease in the calcium aluminate content in the resulting leach cake and an increase in the content of dicalcium silicate (larnite). In addition, the carbonate, sodium aluminosilicate, and gypsum phases disappeared, indicating the occurrence of reactions.

The decrease in the calcium aluminate content is attributed to its reaction with alkali, forming tricalcium hydroaluminate and sodium aluminate (Reaction 1). The increase in the dicalcium silicate content is explained by the reaction between calcium aluminate and sodium silicate, producing dicalcium silicate and sodium aluminate (Reaction 3).

The sodium carbonate phase disappeared as a result of its reaction with tricalcium hydroaluminate, yielding sodium aluminate, alkali, and water (Reaction 4).

The disappearance of the sodium aluminosilicate phase is attributed to its reaction with calcium hydroxide, resulting in the formation of dicalcium silicate, sodium aluminate, alkali, and water (Reaction 5).

The gypsum phase disappeared owing to its reaction with sodium aluminosilicate, producing dicalcium silicate, sodium aluminate, and sodium sulfate (Reaction 6).

To evaluate the efficiency of the proposed soda-caustic leaching process for red mud sinter from the PAP, the results were compared with those obtained under the industrial operating conditions of the existing alumina refinery. At PAP, sinter is leached with recycled Bayer aluminate liquor having a caustic ratio (α_{ku}) of approximately 3.0 to produce a liquor with a caustic ratio of about 1.5 suitable for the decomposition stage. Under these operating conditions, the maximum alumina extraction achieved is approximately 86% [2].

4. Conclusions

The leaching behavior of red mud sinter in soda-caustic solutions was systematically investigated, and the optimum technological parameters for producing an aluminate liquor suitable for subsequent carbonation were established.

Temperature and leaching time were identified as the key process parameters. The maximum Al_2O_3 extraction of 92.3% was achieved at 50 °C using a soda-caustic solution containing 50% caustic alkali, with a leaching time of 30 min and a L/S ratio of 2:1.

Increasing the total alkali concentration had little effect on Al_2O_3 extraction but promoted the dissolution of SiO_2 into the aluminate liquor.

The X-ray diffraction analysis of the leaching residues confirmed the formation of phases consistent with the proposed reaction mechanism and established a correlation between the phase composition of the solid products and the extraction of Al_2O_3 . The results showed that Al_2O_3 extraction is governed by the total content of aluminum-bearing phases remaining in the leaching residue as a result of secondary reactions. Among the investigated parameters, temperature and leaching time were identified as the most significant factors controlling Al_2O_3 extraction into the aluminate liquor.

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Data Availability Statement: The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author.

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