Bayer + Pedersen the Perfect Match for the Future of Alumina Production, with Benefits

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Abstract

Within the EU-funded ENSUREAL project (GA No. 767533), significant efforts have been undertaken to recover and improve expertise from the past (Norwegian Pedersen process from the 1920s) and reach more sustainable alumina production.

The current work uses the data from ENSUREAL's upscaling study and demonstrates a possible way to use bauxite residues from the Bayer process as feed material for the Pedersen process. Generated and available data allowed for a case evaluation considering the foreseen process equipment. An initial approximate assessment of the economic feasibility was conducted.

Treatment of bauxite residue via the electrical route of the Pedersen process produces a significant amount of pig iron. This approach fittingly meets the increasing need to electrify and decarbonize the steel industry. Combining these two processes presents the alumina industry with a promising opportunity for diversification or joint-venture activities for making better use of the raw materials.

Considering a merging of the Bayer and Pedersen processes, the typical alumina extraction rate from the bauxite increases from approximately 80% to more than 95%. Valuable pig iron is generated, covering process oncosts and generating additional revenue. The utilization of the by-product gray mud is under evaluation, and initial positive market feedback has been received. In addition to decreasing its environmental footprint, shifting alumina production to a zero-waste process reduces the financial efforts for disposal and remedial activities.

According to Charles Darwin, "It is the long history of humankind (and animal kind, too) that those who learned to collaborate and improvise most effectively have prevailed," a perspective that could be applicable to the alumina industry as well.

Keywords: Alumina production, Merging Bayer and Pedersen processes, Valorization of bauxite residues, Grey mud, Pig iron coproduction, Diversification of alumina production.

1. Introduction

On May 23, 1925, Harald Pedersen of Trondheim, Norway, filed a priority patent application (with number NO252399X [1]) describing a process of leaching a slag with sodium carbonate to ultimately produce alumina. The US patent US1618105A dated February 15, 1927, is available online and documents this first mention of the Pedersen process. During that period, globalization was not an issue of concern, and, contrary to current times, easy and cheap access to transport was not available. Indeed, one of the advantages of the Pedersen process is the greater flexibility in raw materials, and no need for high-grade bauxites, which allows alumina plants to more easily use, for example, locally available resources. Melting and producing the slag is an equalization step, allowing the generation of a stable input stream for further processing steps. This might also explain why the US Bureau of Mines established, in 1949 [2], intensive testing procedures aiming to make low-grade bauxites available for alumina production. After the Second World War and at the start of the Cold War, such research undoubtedly had a more strategic than economic component.

In parallel to these efforts, an industrial Pedersen plant was in operation in Norway from 1928 to 1969 [3]. The growing and improved Bayer plants were most probably more economical in the 1960s. In addition, less emphasis was placed on sustainability at that time, consequently the Pedersen process fell into a slumber.

In 2017, a project consortium under the lead of SINTEF united its forces and was awarded funding by the European Commission to restart implementing the Pedersen technology for alumina production. The aim was to prove that low-grade bauxite can serve as a raw material for alumina production and that a zero-waste approach avoiding not easily treatable side streams, such as bauxite residues, is possible.

Following the Pedersen process route, the process concept, shown in Figure 1, was investigated within the ENSUREAL project. In the first step, limestone, bauxite, and coke are mixed to generate a slag, which, after melting, gains specific properties suitable for the leaching of the Al components. Mixing and pelletizing are the initial steps to reach a homogeneous feed material and ensure a dust-free rotary kiln treatment. Within the rotary kiln, the prereduction of the iron and calcination of the limestone takes place. Subsequently, the hot material is fed directly into a submerged arc furnace (SAF); this step involves slag design and pig iron production. The subsequent slag cooling stage seems trivial at first glance, but is critical for the further processing steps.

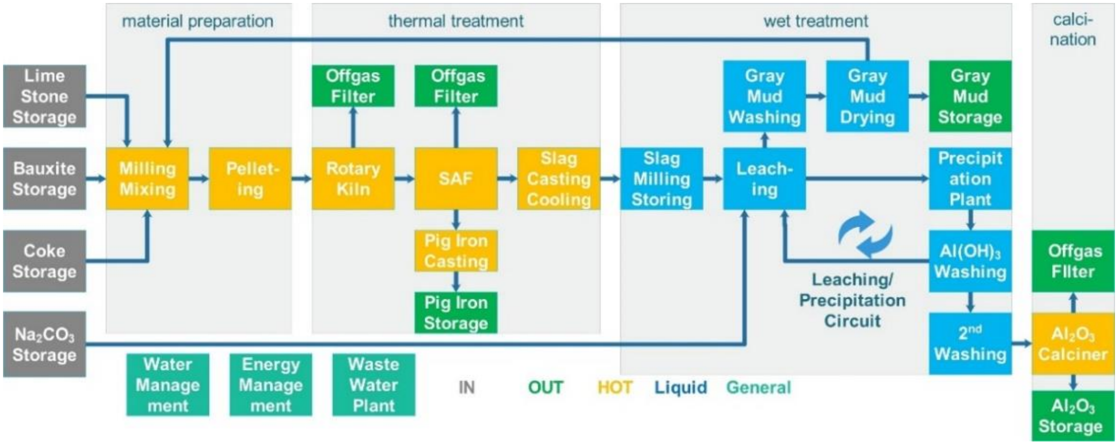


Figure 1. Block flow diagram of the Pedersen process investigated within the ENSUREAL project [4].

Next, milling the slag generates a fine and easy-to-leach material; leaching is executed in a sodium carbonate solution, producing a concentrated solution of sodium aluminate and generating calcium carbonate, the so-called gray mud, as a by-product. Aluminum tri-hydroxide (alumina hydrate) is precipitated by sparging carbon dioxide gas, present in the rotary kiln off-gases, into the pregnant sodium aluminate solution. The precipitate is then separated from the recovered sodium carbonate solution and calcined, and the solution is transferred back to the leaching stage, closing the loop.

Within the ENSUREAL project, finalized in Q1 2022, tests using bauxite and, alternatively, bauxite residues were conducted. The project aimed to investigate a stand-alone process for alumina production using this technology.

Nevertheless, innovations do not end with a project deadline, and the idea to combine a Bayer with a Pedersen plant had already started to grow during the final project stage. In the current paper, we utilized the available data to take the idea one step further and not merely to set up two adjacent plants and use the side stream from the first as feed for the second.

The process idea summarized in the following sections notably merges the two technologies. Shared equipment for the critical process steps, such as using only one crystallization unit for particle growth and only one calciner, generates benefits for the whole process setup. Regarding the mindset, each process should focus on what it can do the most effectively, with the emphasis in the Bayer and Pedersen processes being on the aspects involving alumina and iron, respectively. Applying such a joint approach makes a holistic valorization of the feed material possible.

2. Summary of the Experiments

The practical tests in the ENSUREAL project were carried out at both laboratory and demonstration scale at the alumina plant of the project partner MYTILINEOS. For further upscaling and to reach EU standard technical readiness level (TRL) 8 and 9, executing further continuously operated tests with the designated equipment will undoubtedly be necessary. For the present purposes, the generated results, in combination with literature from the past century, provide a solid basis for the subsequent process and project development steps.

2.1 Sample Preparation

Figure 2 shows the Greek bauxite and bauxite residue samples prepared by MYTILINEOS. In addition, bauxite ores from other regions, such as Jamaica, Turkey, and India, were tested at laboratory scale according to the available infrastructure of the project partners. SINTEF conducted the prereduction tests using hydrogen, and MYTILINEOS used an indirectly heated rotary kiln, as shown in Figure 3, with a capacity of 300 kg/h for prereduction and preheating.

Figure 2. Bauxite residue sample from MYTILINEOS tested.

Figure 3. Indirect heated rotary kiln used for the preheating and prereduction.

2.2 Smelting and Cooling

Smelting and cooling tests revealed significant differences depending on the mixtures, the scale of the tests, and cooling rates. Relevant details are included in the published literature [5-7], and patents [8,9] by the original Pedersen plant team and provide a significant indication of the way forward for industrial implementation. Reaching the correct composition together with a suitably low cooling rate will lead to a self-disintegrating slag and proper leaching and desilication behavior. Figure 4 depicts an electrical furnace used at the MYTILINEOS plant, and Figure 5 shows a self-disintegrating slag sample.

Figure 4. Smelting tests at the MYTILINEOS pilot plant.

Figure 5. Self-disintegrating slag sample.

2.3 Leaching

The chemical reactions associated with the leaching process are given in Equations 1 to 4:

$$(CaO.Al_2O_3)_s + (Na_2CO_3)_{aq} + 4H_2O = 2(NaAl(OH)_4)_{aq} + (CaCO_3)_s \quad (1)$$
$$(3CaO.Al_2O_3)_s + 3(Na_2CO_3)_{aq} + 6H_2O = 2(NaAl(OH)_4)_{aq} + 3(CaCO_3)_s + 4NaOH \quad (2)$$
$$(12CaO.7Al_2O_3)_s + 12(Na_2CO_3)_{aq} + 33H_2O = 14(NaAl(OH)_4)_{aq} + 12(CaCO_3)_s + 10NaOH \quad (3)$$
$$(2CaO.SiO_2)_s + 2(Na_2CO_3)_{aq} + H_2O = (Na_2SiO_3)_{aq} + 2(CaCO_3)_s + 2NaOH \quad (4)$$

Leaching presented a specific challenge that notably requires further investigation. Literature sources [10-13] contain the relevant published data, and finding the proper balance between temperature, concentration, and solid-to-liquid ratio is critical to reach high yields and low silica values. Comparing the data with that of the original Pedersen plant showed that the past approach in working with highly dilute solutions might also merit further investigation. The investigated case will undoubtedly involve larger, though not complex or cost-intensive, equipment; thus, further considerations at the next development level are notably needed. Figure 6 depicts the leaching reactor used in the pilot plant, and Figure 7 shows the gray mud produced within this reactor.

Figure 6. Leaching reactor at the MYTILINEOS pilot plant.

Figure 7. Gray mud produced in the MYTILINEOS pilot plant.

2.4 Precipitation

Precipitation tests were also conducted at laboratory and pilot scale, and the first challenge involved identifying the process conditions where $Al(OH)_3$ rather than dawsonite ($NaAlCO_3(OH)_2$)) precipitates [14]. High precipitation yields > 95% were possible, and, compared to the saturation-driven crystallization in the Bayer process, CO_2-driven chemical precipitation can be considered as fast process. The particle size generated within the first run was rather fine, but growing the particles by using the material produced from a previous run as seed for a subsequent one was also possible. Finally, metallurgical-grade material with a suitable particle size distribution was generated in the laboratory within seven cycles using the seeding technique. An additional outcome was the successful production of bayerite or gibbsite phases, which forms a critical aspect for easy implementation and good performance in existing plants. Figure 8 shows the test setup at the National Technical University of Athens (NTUA), and Figure 9 illustrates that regarding $Al(OH)_3$ precipitation, reproducing the laboratory results in a close-to-industrial environment is also possible.

Figure 8. Lab-Pilot setup at NTUA used for investigating the fundamentals and boundaries of the precipitation process.

Figure 9. Precipitated and filtered $Al(OH)_3$ at the MYTILINEOS pilot plant.

2.5 Utilization

Within the scale-up study for a 500-kt stand-alone alumina plant, equipment sizing was carried out, and the first economic figures were calculated based on different assumptions. A return-on-investment period of approximately seven years is considered a reasonable figure for a long-life strategic industrial plant.

3. Results

The process concept presented herein is a result of the executed work of the ENSUREAL project but could also be viewed as a next-generation approach. A priority process patent application was filed by MYTILINEOS and KON Chemical Solutions in Austria, and the evaluation process for the subsequent steps for commercializing the process are ongoing.

An evaluation of whether the Bayer or Pedersen process is the optimum production process seems to be the wrong approach. A side-by-side comparison does not sufficiently utilize the potential synergies between the two processes.

The process concept shown in Figure 10 merges the Bayer and Pedersen processes to optimize the strengths of each so that the input bauxite can be valorized at a significantly higher overall rate.

3.1 Process Concept Description and Explanation of Synergies

Figure 10 illustrates the concept of merging the Bayer and Pedersen processes to produce alumina and pig iron. As a starting point, the bauxite residue from the Bayer side, with a remaining moisture content of approximately 25 wt-%, is transferred to the Pedersen side. By having a direct connection between the two plants a reduced buffer capacity is required. Assuming that the Pedersen process is set up adjacent to an operating plant, the bauxite residue storage facilities could be located relatively nearby.

The material is milled and mixed with the required materials for the prereduction and smelting processes, namely, limestone, coke, and quicklime, for enhanced pelletizing properties. Depending on the final limestone source, milling and mixing can also be optimized in such a way that the proper moisture level for pelletizing is reached. Pelletizing is needed to avoid dust and supply material of a certain size to the SAF. The soft “green” pellets, produced on a pelletizing disc, are transferred to a Lepol rotary kiln setup where they are dried and hardened with the off-gas stream before prereduction; the hot material is then discharged for further treatment.

The off-gas released by the Lepol rotary kilns has an enriched CO_2 concentration compared to standard combustion processes. The off-gas is dedusted, cooled, and reused in the later $Al(OH)_3$-precipitation step.

The hot material is transferred directly into the SAF where smelting occurs. During this process, a specific Ca–Al slag and pig iron, are produced (pig iron is a valuable feed material in the foundry industry and a potential new product and revenue stream for the future alumina/iron industry). The generated slag is cast into molds and cooled following a specific procedure to attain self-disintegrating properties and become easily mill able.

The Ca–Al slag is then milled as required for processing reasons and transferred to the leaching circuit. A detailed leaching procedure must be put in place and optimized in an industrial-scale setup. In general, though, a two-stage approach is foreseen: the main leaching process takes place in the first stage, followed by retention and settling time allowing for self-desilication in the second. Gray mud – comprising mainly $CaCO_3$ (limestone) – generated within the leaching procedure is processed in a countercurrent washing procedure to reduce the loss of valuable process elements, such as Na and Al, to the greatest extent possible. The gray mud also constitutes a well-defined by-product with a small particle size distribution and could be further used in the fertilizer or cement industries or for the recovery of scandium.

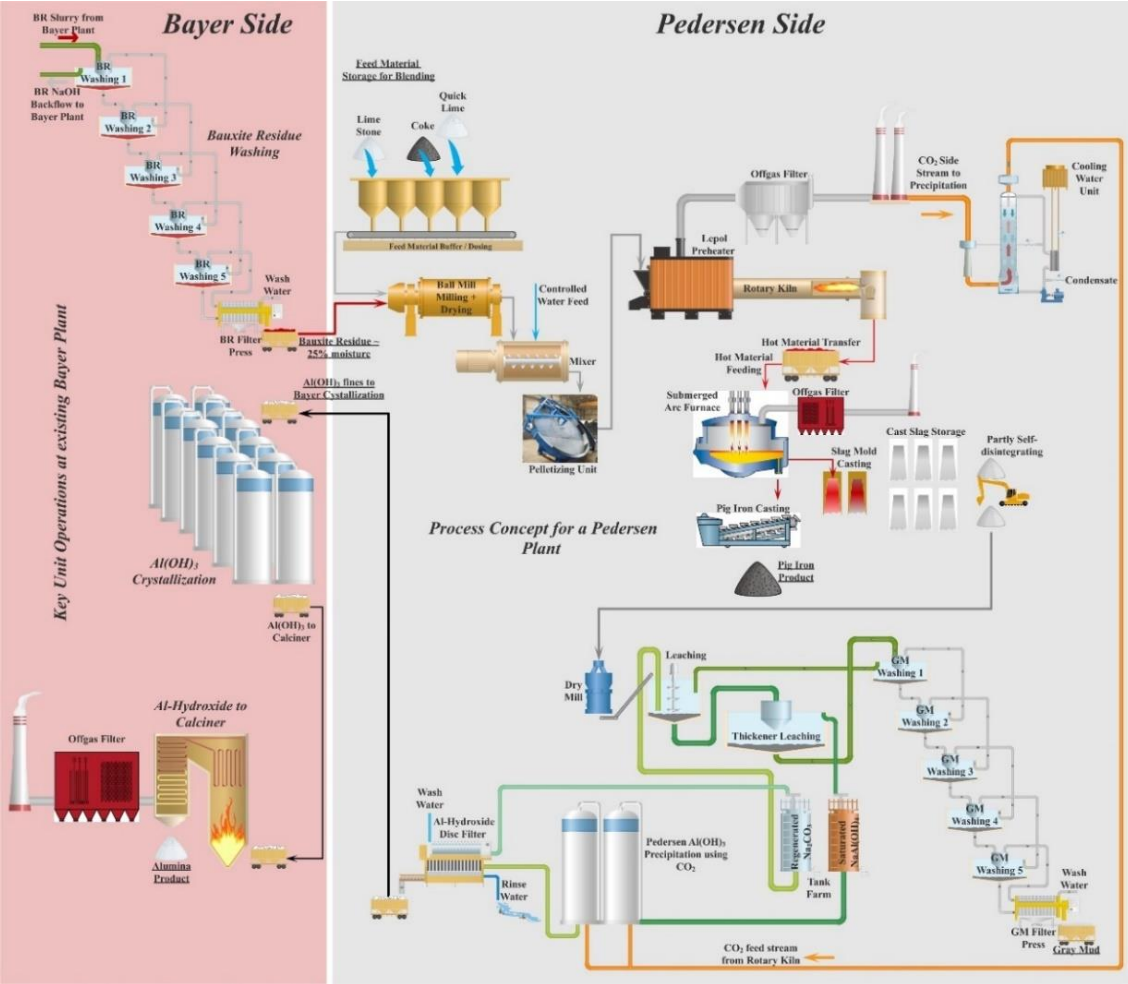


Figure 10. Process concept to utilize the benefits of the Bayer and Pedersen processes.

The generated Na–Al solution is buffered in a tank farm before being transferred to the precipitation stage. As shown in Figure 10, a fast one-stage precipitation process using CO_2 produces fine $Al(OH)_3$ particles and the recovered Na_2CO_3 solution. The solid fraction is separated from the recovered solution by filtration, and the solution is buffered in the tank farm for reuse in the leaching procedure. At the same time, the washed $Al(OH)_3$ is transferred to the Bayer plant's crystallization unit, where the seeding stream is increased and the particles can grow further to attain the well-defined and known properties for calcination and then electrolysis.

The possibility of jointly using existing equipment of operating plants and obtaining more products from the same input feed could lead to significant benefits. Furthermore, capacity increase of, for example, precipitation or calcination may be required but is expected to be much easier and more economically viable compared to using a greenfield approach.

3.2 Utilization of One Ton of Greek bauxite

The Sankey diagram in Figure 11 shows the effects on the utilization of Greek bauxite in the combined Bayer and Pedersen process. The data is obtained from a combination of literature and experimental data, which should reflect a reasonable first assumption for a later industrial approach. Using highly dilute solutions, the original Pedersen plant reportedly reached 95 % Al_2O_3 extraction [15], and within the ENSUREAL project, approximately 70 % Al_2O_3 extractions were achieved using concentrated solutions [12]. Thus, 85 % extraction for the Pedersen side in Figure 11 represents a reasonable intermediate value that should be achievable after some optimization.

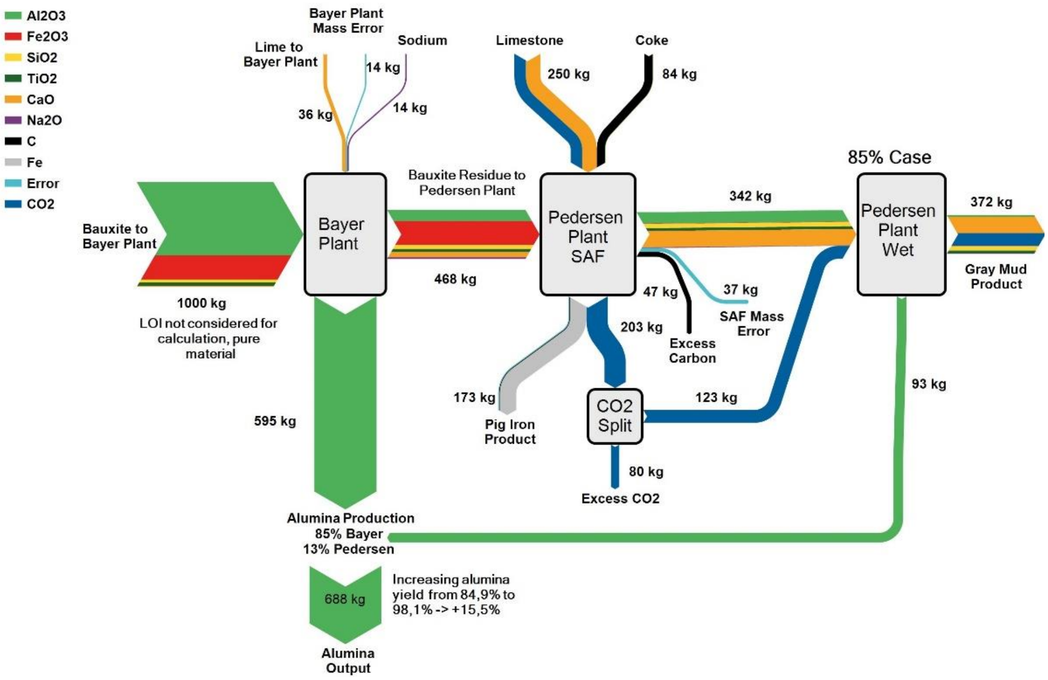


Figure 11. Preliminary mass streams for utilizing one ton of Greek bauxite in the combined Bayer and Pedersen process, assuming a Pedersen leaching yield of 85 %.

In summarizing the key findings from Figure 11, the following products and improvements can be achieved:

- Alumina extraction from the bauxite used can reach 98 %, which implies an additional 93 kg of Al_2O_3 per ton of treated bauxite, compared to 595 kg for the Bayer process alone.
- The generated 173 kg of pig iron is a significant by-product stream, which adds some diversification to alumina plant operations.
- The 372 kg of gray mud undoubtedly constitutes a lower-value side stream that would, based on its properties (mainly $CaCO_3$, less caustic), allow much easier valorization compared with bauxite residue.

4. Discussion

In the ENSUREAL project, the scope was to consider a greenfield project using tropical bauxite and a production capacity of 500 000 t/a of alumina. The evaluated case in this paper starts from a different perspective. Assuming a given bauxite residue and an existing Bayer plant with a production capacity of 800 000 t/a, the add-on Pedersen plant case was evaluated. In doing so, the process equipment considered in the ENSUREAL project can be significantly downsized, and synergies can occur by using the already available infrastructure and unit operations in the Bayer plant. An additional simplification is to assume sufficient spare capacity at existing Bayer precipitation and calcination facilities. By doing so, no additional CAPEX is required for these processing stages.

4.1 Summary of Case Evaluation

Following this approach, a base case was generated for a Pedersen plant installed adjacent to an existing and running Bayer unit to gain an initial impression of whet the business case would look like. In the considered case evaluation, the bauxite residue had the following composition: 38 wt-% Fe_2O_3, 18 wt-% Al_2O_3, 19 wt-% rest (SiO_2, TiO_2, CaO, trace elements) and 25 wt-% remaining moisture. The input and output streams are as follows.

Input streams:

• Bauxite residue	800 000 t/a
• Limestone	230 000 t/a
• Coke	40 000 t/a
• Sodium carbonate	29 000 t/a
• Quicklime	50 000 t/a

Output streams:

• Alumina as add-on to existing Bayer Plant	125 000 t/a
• Pig iron	210 000 t/a
• Gray mud (dry)	420 000 t/a

Figure 12 shows a layout concept for the Pedersen component of a combined Bayer and Pedersen process. This assumes that the Bayer component is an already existing plant and that land is available for the adjacent Pedersen plant. The assumed area is 410 x 180 m for the additionally required key process equipment.

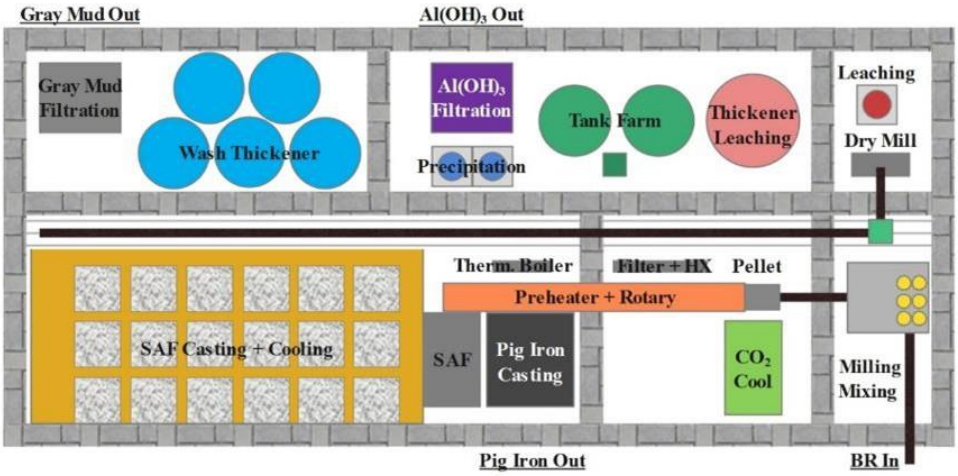


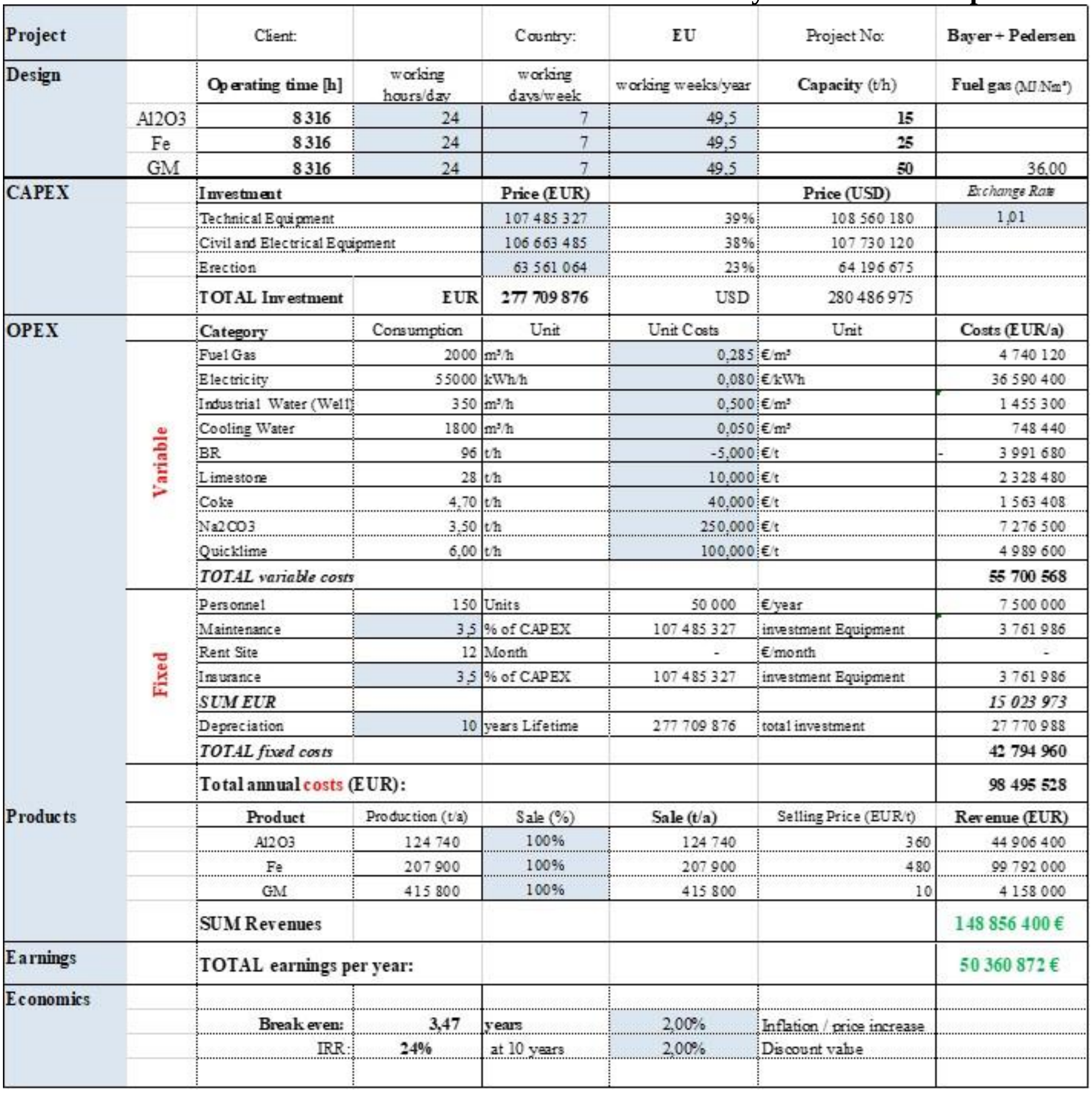
Figure 12. Layout concept for the Pedersen extension (410 x 180 m).

Based on the pricing of the available equipment in the mentioned greenfield case for a tropical bauxite ore, a downscaling and adjustment procedure was carried out for the relevant processing equipment. The equipment price estimation was conducted between Q3 2021 and the start of Q1 2022. Considering these price levels, the first CAPEX estimate for treating the abovementioned volumes with this new strategy is approximately €278 000 000.

We carried out an initial business case evaluation, as shown in Table 1. The assumed prices of alumina and pig iron are from 2021, which might be a fair consideration in light of the different global crises occurring in mid-2022. The gray mud was assumed to be fertilizer, and, considering a value of merely €10, one can view it as having a neutral or minor overall impact on the business case.

Regarding energy consumption, estimations given by the equipment suppliers were considered. At the current investigation level, this is a considered a reasonable and sufficiently detailed approach. For further process development, consumption figures of a continuously operating pilot plant and an initial optimization and detailed energy integration are essential. The energy prices are a first estimate and are different for each case and location.

Table 1. Initial business case evaluation of the combined Bayer and Pedersen process.



Project		Client:		Country:	**EU**	Project No:	**Bayer + Pedersen**
Design		**Operating time [h]**	working hours/day	working days/week	working weeks/year	**Capacity** (t/h)	**Fuel gas** (MJ/Nm³)
	Al2O3	**8 316**	24	7	49,5	**15**	
	Fe	**8 316**	24	7	49,5	**25**	
	GM	**8 316**	24	7	49,5	**50**	36,00
CAPEX		**Investment**		**Price (EUR)**		**Price (USD)**	*Exchange Rate*
		Technical Equipment		107 485 327	39%	108 560 180	1,01
		Civil and Electrical Equipment		106 663 485	38%	107 730 120	
		Erection		63 561 064	23%	64 196 675	
		TOTAL Investment	EUR	**277 709 876**	USD	280 486 975	
OPEX		**Category**	Consumption	Unit	Unit Costs	Unit	**Costs (EUR/a)**
	Variable	Fuel Gas	2000	m³/h	0,285	€/m³	4 740 120
		Electricity	55000	kWh/h	0,080	€/kWh	36 590 400
		Industrial Water (Well)	350	m³/h	0,500	€/m³	1 455 300
		Cooling Water	1800	m³/h	0,050	€/m³	748 440
		BR	96	t/h	-5,000	€/t	- 3 991 680
		Limestone	28	t/h	10,000	€/t	2 328 480
		Coke	4,70	t/h	40,000	€/t	1 563 408
		Na2CO3	3,50	t/h	250,000	€/t	7 276 500
		Quicklime	6,00	t/h	100,000	€/t	4 989 600
		TOTAL variable costs					**55 700 568**
	Fixed	Personnel	150	Units	50 000	€/year	7 500 000
		Maintenance	3,5	% of CAPEX	107 485 327	investment Equipment	3 761 986
		Rent Site	12	Month	-	€/month	-
		Insurance	3,5	% of CAPEX	107 485 327	investment Equipment	3 761 986
		SUM EUR					***15 023 973***
		Depreciation	10	years Lifetime	277 709 876	total investment	27 770 988
		TOTAL fixed costs					**42 794 960**
		Total annual costs (EUR):					**98 495 528**
Products		**Product**	Production (t/a)	Sale (%)	**Sale (t/a)**	Selling Price (EUR/t)	**Revenue (EUR)**
		Al2O3	124 740	100%	124 740	360	44 906 400
		Fe	207 900	100%	207 900	480	99 792 000
		GM	415 800	100%	415 800	10	4 158 000
		SUM Revenues					**148 856 400 €**
Earnings		**TOTAL earnings per year:**					**50 360 872 €**
Economics							
		Break even:	**3,47**	years	2,00%	Inflation / price increase	
		IRR:	**24%**	at 10 years	2,00%	Discount value	

Taking all these uncertainties into account, one should note that Figure 13 represents only an initial impression regarding a possible business case. Given these assumptions it can be concluded that in addition to the environmental benefits with the combined Pedersen-Bayer process, economic benefits are also achievable.

4.2 Future Directions

Generated expertise built and modified test infrastructure, and the skills developed for the Pedersen process by the involved teams are an ideal starting point for further development. Within the ENSUREAL project, initial batchwise tests revealed that the concept was functional and allowed to identify other important aspects of the process.

As with electromobility, which lay dormant for almost a century before it recaptured people's attention through its use in small sports cars, the Pedersen process requires a similar rebirth. The next logical step is a continuous working pilot and demonstration unit at an existing alumina plant, which would allow further and deeper investigation into matching the Bayer and Pedersen processes. Building and sustaining trust in the process and reaching several thousands of hours of continuous operation in demonstration plants are critical steps before implementing the process on an industrial scale. Notably, proof that the required specifications and performance of the product alumina can be reached is a mandatory precondition before a plant that solves the bauxite residue challenge can finally be built.

5. Conclusion

Overall, merging the Bayer and Pedersen processes can serve as a suitable approach to improve the valorization of mined and used bauxite. Generating a second value chain within an alumina plant with pig iron production would lead to some diversification. Iron production via alternative routes and using electricity has top priority within the EU's decarbonization agenda but also displays a high potential for global implementation within the following decades.

Gray mud can be used as a fertilizer or in the cement industry as a calcium source; thus, it is, in any case, sufficiently pure to be a product and not a waste stream. Also tests for the extraction of rare earth elements (REE) were executed as possible added value streams. Further investigation regarding the grade of mud as a product with a well-defined chemical composition and particle size distribution compared to natural sources must be included in subsequent development steps.

The possibility to reduce land use in the alumina industry and transform bauxite residue from waste into a product would be highly appreciated by all stakeholders.

Further efforts are undoubtedly needed before the process concept presented herein can be implemented. Both past and current expertise are available, and the project team would be very keen to accept the challenge of pushing the concept further forward to reach a more sustainable alumina industry.

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