



# Efficient and Complete Exploitation of the Bauxite Residue (Red Mud) Produced in the Bayer Process

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## Abstract

The Bayer Process for the production of alumina from Bauxite ore is characterized by low exergy efficiency and it results in the production of significant amounts of dust-like, high alkalinity bauxite residues known as red mud. Currently red mud is produced almost at 1 to 1 mass ratio to metallurgical alumina and is disposed into sealed or unsealed artificial impoundments (landfills), leading to important environmental issues. A patent-pending energy and exergy efficient process has been recently developed by the Advanced Mineral Recovery Technologies (AMRT, Ltd.) and NTUA's Laboratory of Metallurgy, for the direct transformation of red mud into valuable products, such as pig iron and mineral wool. The novel process utilizes an innovative electric arc furnace (EAF) technology to achieve the carbothermic reduction of the red mud waste without any pretreatment, producing pig iron and viscous slag suitable for direct mineral wool production. Thus, the environmental footprint of the Bayer Process is reduced substantially, as the initial bauxite ore is exploited in full and no solid wastes are produced. The overall exergy efficiency of the new bauxite exploitation schema increases from 3 % in the conventional Bayer Process to 6 to 9 % depending on the method used to produce the electricity needed to power the Electric Arc Furnace. Additionally, as the novel process enables the single step co-production of two highly valuable by-products (pig iron and mineral wool), it has the potential to significantly increase the versatility and profit margin of the alumina producing industry. In this paper a thermodynamic study and preliminary experimental results of the red mud treatment process are presented, along with an overall energy, exergy and economic analysis of the new bauxite exploitation schema.



# 1 Introduction

Today primary aluminium is produced exclusively from bauxite ore through a common industrial production practice consisting of two distinct stages: (i) the production of high grade metallurgical alumina ( $\text{Al}_2\text{O}_3$ ) from bauxite that is performed according to the Bayer Process and (ii) the electrolytic reduction of alumina to aluminium, which is performed according to the Hall-Héroult process. The Bayer Process [1], patented in 1888 by Karl Joseph Bayer, is essentially a cyclic chemical process where at first (digestion stage) alumina is extracted from bauxite ore with sodium hydroxide solution, under high pressure (3.5 MPa) and elevated temperature (140 °C to 300 °C). The pregnant solution after clarification in thickeners (liquor clarification stage), is cooled down to 60 °C and seeded with gibbsite in order to achieve the precipitation of trihydrate alumina (precipitation stage). The spent caustic soda solution is regenerated usually through evaporation (liquor surge stage), while the trihydrate alumina is calcinated at 1100 °C to produce powdered, high grade metallurgical alumina (calcination stage). A previously published study of this cyclic process [2], shows that it is characterized by a very low exergy efficiency (3 %) as it spends large amounts of heat (12.77 MJ/kg of  $\text{Al}_2\text{O}_3$ ), usually generated through heavy fuel burning (leading to 0.83 kg of  $\text{CO}_2$ /kg  $\text{Al}_2\text{O}_3$  including limestone calcination). On average the Bayer Process requires 2.65 kg of bauxite ore to produce 1 kg of alumina, while the slurry containing the remaining bauxite ore, which is removed from the thickeners during the liquor clarification stage, is by far its greatest environmental problem.

This byproduct, called bauxite residue or “red mud”, on a dry basis is produced in almost a 1 to 1 kg ratio to alumina and consists from various metal oxides of Fe, Al, Ti, Si, K, Na, V, Ga (depending on the initial chemical composition of the bauxite ore) along with inclusions of unwashed sodium aluminate solution. Mineralogically, in red mud, iron content is found as hematite ( $\text{Fe}_2\text{O}_3$ ) or goethite ( $\text{FeOOH}$ ), while aluminum content is found as gibbsite ( $\text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O}$ ), diaspore ( $\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$ ), cancrinite ( $\text{Na}_6\text{Ca}_2(\text{Al}_6\text{Si}_6\text{O}_{24})(\text{CO}_3)_2 \cdot 2\text{H}_2\text{O}$ ) and katoite ( $\text{Ca}_3\text{Al}_2(\text{SiO}_4)(\text{OH})_8$ ) [3]. A red mud chemical analysis used in the present work is given in Table 1.

Table 1: Red mud sample

Weight used	1000 kg
Chemical species	in % wt./wt.
$\text{Al}_2\text{O}_3$	18.76
$\text{Fe}_2\text{O}_3$	44.38
CaO	11.88
$\text{SiO}_2$	6.13
$\text{TiO}_2$	5.54
$\text{Na}_2\text{O}$	2.20
$\text{H}_2\text{O}_{(\text{cry})}$	9.77
Moisture	1.00
Total	99.66



Although red mud is classified by EC as a non hazardous waste [4], its small particle size (dust-like, mean particle size 0.49  $\mu\text{m}$  [3]), high alkalinity and large amounts (30 to 35 million tonnes per year on a dry basis worldwide [5]) makes its disposal a significant problem. Today, red mud is disposed into sealed or unsealed artificial impoundments, leading to important environmental issues (e.g. groundwater pH change, leakage, overflow, air pollution by dust) and substantial land use (and thus substantial costs for the alumina producing industry) [5, 6]. The catastrophic red mud spill in Hungary in October 2010 is indicative of the magnitude of the red mud waste disposal problem.

As the production of red mud in the Bayer Process is inevitable, the primary aluminium industry has always focused on discovering potential applications for it. As red mud is generally characterized by high iron content many attempts have been made to produce pig iron from red mud, but so far no economically viable solution has been found. The extremely fine particles of red mud require agglomeration prior to feeding in conventional reactors; their high alkaline nature is unsuitable for blast furnace reductive smelting and the low, when compared to iron ores, content in iron oxides makes the production of pig iron a cost ineffective process [7, 8]. Valuable metals like Ti, Ga and V contained in the red mud can be extracted [9, 10], but again with no economical benefits [7]. Other proposed uses for red mud include the production of construction or ceramic materials [5], as well as utilizing red mud in waste waters or polluted soils treatments [5, 11, 12]. Till this day, due mainly to high costs and low yields, no industrial application of red mud reuse is in effect.

## 2 The novel Red Mud Treatment process

### 2.1 General description

A patent-pending process has been recently developed by the Advanced Mineral Recovery Technologies (AMRT, Ltd.) and the NTUA's Laboratory of Metallurgy, for the direct transformation of red mud into valuable products and is currently being optimized under the EC-funded "Novel technologies for enhanced energy and exergy efficiencies in primary aluminium production industry (ENEXAL)" collaborative research project [13]. This treatment is based on the innovative AMRT-Electric Arc Furnace (AMRT-EAF) which has the capability of processing finely sized materials, notably below 1 mm in particle size (dust like), without any dusty material loss in the off-gas stream [14]. As seen in Figure 1, the AMRT-EAF innovative feeder technology can deliver the dusty raw materials directly into the "arc zones" of each of its three electrodes, where flash smelting takes place. This feeder is below the off-gas suction tube thereby preventing loss of material, and it feeds the reactor with small batches at appropriate time intervals in order to allow enough time for ventilation of the off-gases produced from the previous batch. The operation of the AMRT-EAF is controlled by a unique patented PLC control system [15], which, based on system-dependent programmable parameters predicted accurately by a proprietary thermodynamic model, can control impedance of the electric arc produced by the electrodes through the control of the positioning of the electrodes within the furnace influencing the energy input, the arc stability, the solid charge melting pattern and the electrode consumption. Therefore, this innovative EAF technology is ideal



for processing the dust-like red mud without any pretreatment or substantial energy losses, thus providing the proposed process with a significant industrial advantage.

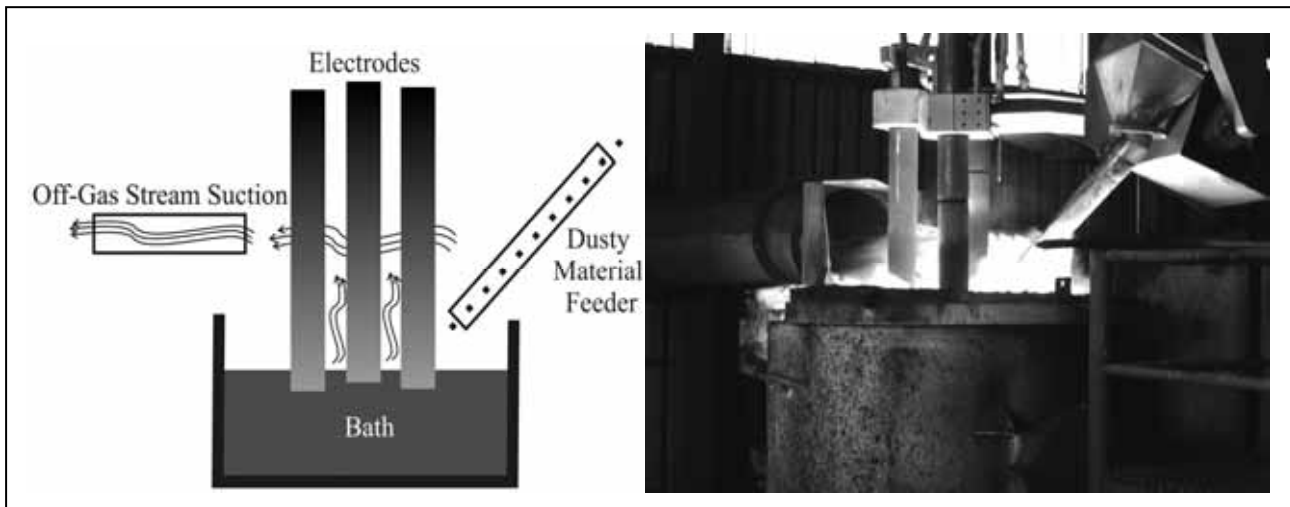


Figure 1: Schematic diagram and photograph of the innovative AMRT-EAF melt reduction furnace in operation

Using carbon as a reducing agent, the red mud can undergo reductive smelting in the AMRT-EAF producing pig iron and a viscous slag. The pig iron produced in this way would amount to approximately to 35 % of the initial red mud charge, therefore pig iron production alone would not solve the industry's waste disposal, as 65 % of the red mud would still have to be disposed as EAF slag, while the expected turnover from selling such amounts of pig iron would not suffice to make for an economically viable process.

The novel red mud treatment process, however, aims at turning all of the red mud waste into valuable products and this includes transforming the EAF slag into inorganic fibres suitable for the production of a variety of products commonly known as mineral (or slag, rock) wool products. Mineral wool products due to their light weight, low thermal coefficient, incombustibility and high temperature melting points ( $> 1000\text{ }^{\circ}\text{C}$ ) are widely used as refractory, thermal and acoustic insulation or even light weight construction materials [16]. These broad range of applications guarantees a large market for the red mud treatment EAF slag. Indicatively, in 2003 mineral and glass wool products accounted for 60 % of the thermoinsulation market in Europe [17]. Taking into consideration that the EAF slag can be fiberized in situ, therefore avoiding the expensive melting phase of conventional mineral wool production (which accounts for up to 70 % of the total mineral wool production energy [18]), then one can expect that the proposed red mud treatment process will be economically viable.

The proposed red mud treatment comprises of four stages as shown schematically in Figure 2. The first stage is the red mud drying stage, as even red mud dewatered in filter presses contains significant amounts of moisture (up 25 % wt./wt.). This stage can take place in a rotary kiln, utilizing the heat content of the hot off-gases from the EAF. In the next stage of the process the material feed of the EAF is prepared by mixing the dry red mud, coke fines and appropriate fluxes like silica sand to



adjust the properties of the produced slag. This mixture is feed into the AMRT-EAF where the raw materials undergo reductive smelting and are transformed in three distinct fluid phases: liquid slag, liquid pig iron and off-gases. The off-gases after heat exchange in the red mud dryer are sent in a bag-house unit to remove dust particles prior to realising them to the atmosphere. The dust collected is recycled in the feed material. The liquid pig iron and slag phases are separated by sequential pouring (or by tapping in a continuous process) and the slag is driven directly to the final stage of the process, where the liquid slag is fiberized to produce inorganic fibres and mineral wool.

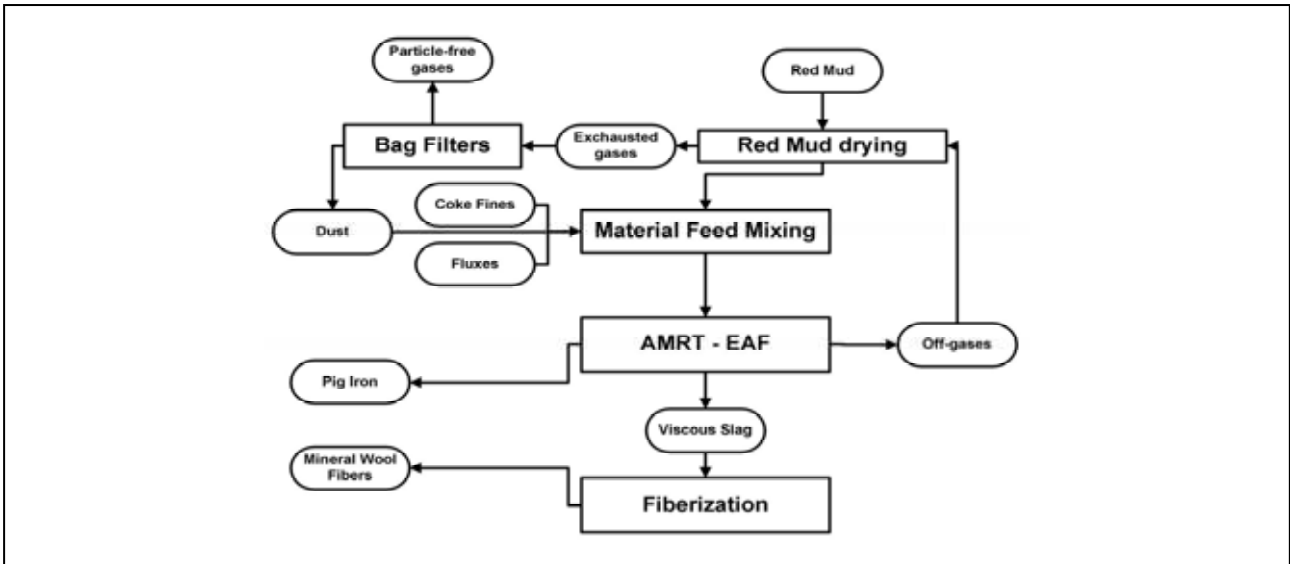


Figure 2: Schematic diagram of the red mud treatment process

## 2.2 Thermodynamic analysis

The chemical analysis of a red mud sample is given in Table 1. In order to design the process the following operational parameters have to be established: melt temperature, required amount of coke and required amount of fluxes. As shown in the Ellingham diagram presented in Figure 3, the oxidation of carbon to  $\text{CO}_{(g)}$  can reduce  $\text{H}_2\text{O}$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{Na}_2\text{O}$ ,  $\text{K}_2\text{O}$  at temperatures higher than  $1000\text{ }^\circ\text{C}$ , it can reduce  $\text{SiO}_2$  and  $\text{TiO}_2$  at temperatures higher than  $1700\text{ }^\circ\text{C}$ ,  $\text{MgO}$  at temperatures higher than  $1800\text{ }^\circ\text{C}$ , while  $\text{Al}_2\text{O}_3$  and  $\text{CaO}$  cannot be reduced at temperatures lower than  $2000\text{ }^\circ\text{C}$ . As the melting point of pure iron is  $1537\text{ }^\circ\text{C}$  the furnace operational temperature should be in the range of  $1500\text{ }^\circ\text{C} < T < 1700\text{ }^\circ\text{C}$  in order to effectively reduce the iron in the red mud while avoiding silicon and titanium reductions. The furnace operational temperature for this study is therefore set at  $1600\text{ }^\circ\text{C}$  and the reactions that are thermodynamically expected to take place and consume carbon are



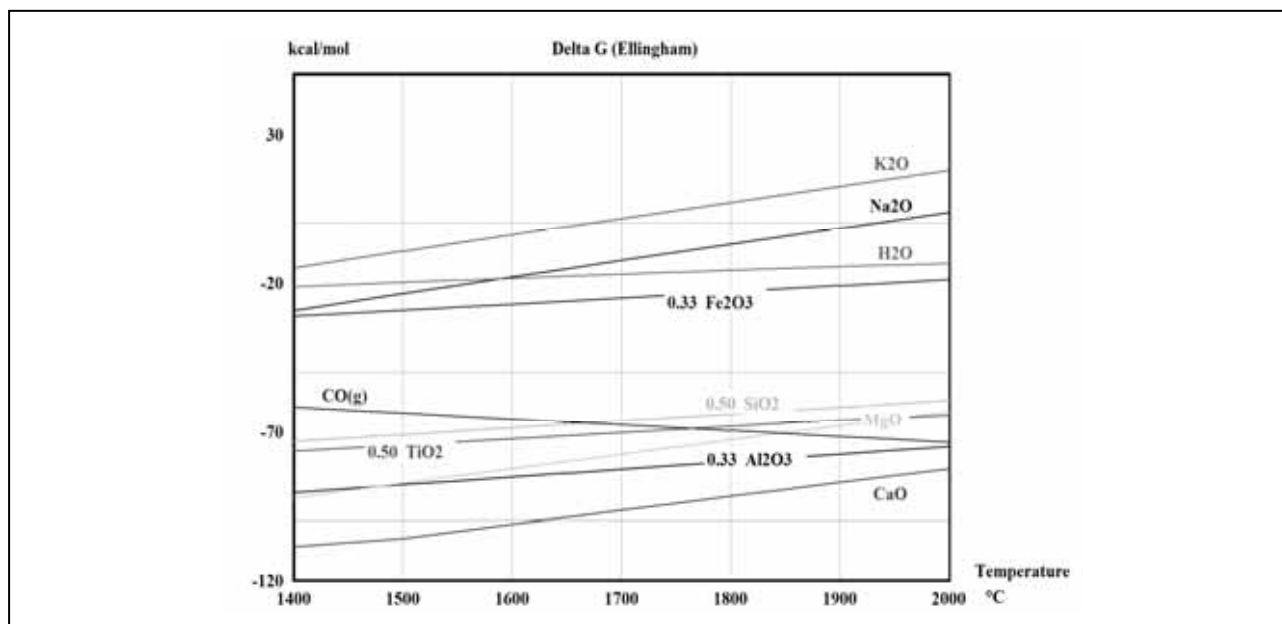
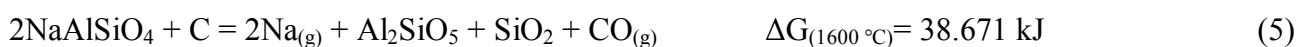


Figure 3: Ellingham Diagram calculated from HSC Chemistry 6 software

The evolution of  $\text{Na}_{(g)}$  and  $\text{K}_{(g)}$  according to Equations (3) and (4) are highly unlikely as these oxides are in reality part of aluminosilicates complexes which are not as easily reduced. For instance if sodium in red mud is found as nepheline (a logical assumption as nepheline is a part of cancrinite), then its reduction is thermodynamically unlikely



Therefore according to Equations (1) and (2) and the red mud chemical analysis given in Table 1 the amount of carbon required to reduce all iron and hydrogen content found in 1000 kg of red mud is 172.24 kg. To account for all possible side reactions an excess of 10 % carbon will be used in the process (189.46 kg/1000 kg red mud). As mentioned earlier, this carbon will come from coke fines the chemical analysis of which is given in Table 2.

Table 2: Coke fines

Weight used	249.29 kg
Chemical species	in % wt./wt.
C	76.00
S	0.20
P	0.40
Fe	1.20
$\text{SiO}_2$	11.87
CaO	0.74
$\text{Al}_2\text{O}_3$	5.19
Volatiles	3.20
$\text{H}_2\text{O}$	1.2
Total	100.00



According to the chemical analysis of the red mud the mass ratio of alkaline oxides (CaO + MgO) to acidic ones (SiO<sub>2</sub>), known as basicity ratio, is 1.95 which indicates that the slag phase produced would be characterized by low viscosity and low electrical resistivity, as SiO<sub>2</sub> acts both as a network former and an electric insulator (by “capturing” in silica networks possible current carriers like sodium and titanium). To achieve good EAF operation and an efficient slag fiberization an acidic slag is required and therefore the addition of silica fluxes is necessary. In this preliminary work an ad-hoc addition of 150 kg of silica sand was used, in order to lower the basicity ratio of the produced slag to 0.58 and achieve more favourable conditions both for EAF operation and for subsequent slag fiberization.

Using as input the red mud, coke fines and silica sand described in Tables 1 to 3, the thermodynamic equilibrium composition of the system at 1600 °C was calculated using FactSage<sup>TM</sup> 6.2 Equilib module and the results are presented in Tables 4 to 6. According to these results 99.9 % Fe recovery can be achieved in the pig iron phase. It is worth noticing that part of silicon and titanium is reduced in the process, as their dissolution in the pig iron phase changes their activities and therefore makes these reductions possible. Traces of sodium gas (0.3 vol.-%) are found in the predicted gas phase.

Table 3: Chemical Analysis of silica sand

Weight used	150 kg
Chemical species	in % wt./wt.
SiO <sub>2</sub>	95.0
Fe <sub>2</sub> O <sub>3</sub>	0.7
Al <sub>2</sub> O <sub>3</sub>	2.2
Na <sub>2</sub> O	0.4
K <sub>2</sub> O	0.9
Total	99.1

### 2.3 The complete bauxite exploitation schema

The electrical energy requirement for the operation of the AMRT-EAF furnace at the conditions described in the section 2.2 is calculated at 6621.2 MJ/t of red mud processed. Based on this calculation and the results of the thermodynamic modelling the mass and energy diagram of the proposed red mud treatment process is shown in Figure 4. The exergy efficiency of the red mud process is 8 % when the EAF is powered with coal produced electricity (which from exergy point of view is the worst case scenario for electricity generation [2]). If the EAF was powered from electricity produced at no additional exergy cost (e.g. from hydroelectric dams or renewable sources) the exergy efficiency of the process would be 14 %.

The new process for the complete bauxite exploitation (for alumina, pig iron and mineral wool production) will increase the exergy efficiency from 3 % in the conventional Bayer Process [2] to 6 % when the EAF is powered with coal produced electricity or 9 % when the EAF is powered with





hydroelectrically produced electricity. This increase in efficiency is due to the fact that the solid waste of the Bayer Process (red mud) with chemical exergy of 0.32 MJ/kg Al is now replaced by pig iron and mineral wool products, with total chemical exergy 4.4 MJ/kg Al. From an economic perspective a solid waste with costly disposal, is replaced, in a single step process, by two valuable byproducts thereby significantly increasing the versatility and profit margin of the industry.

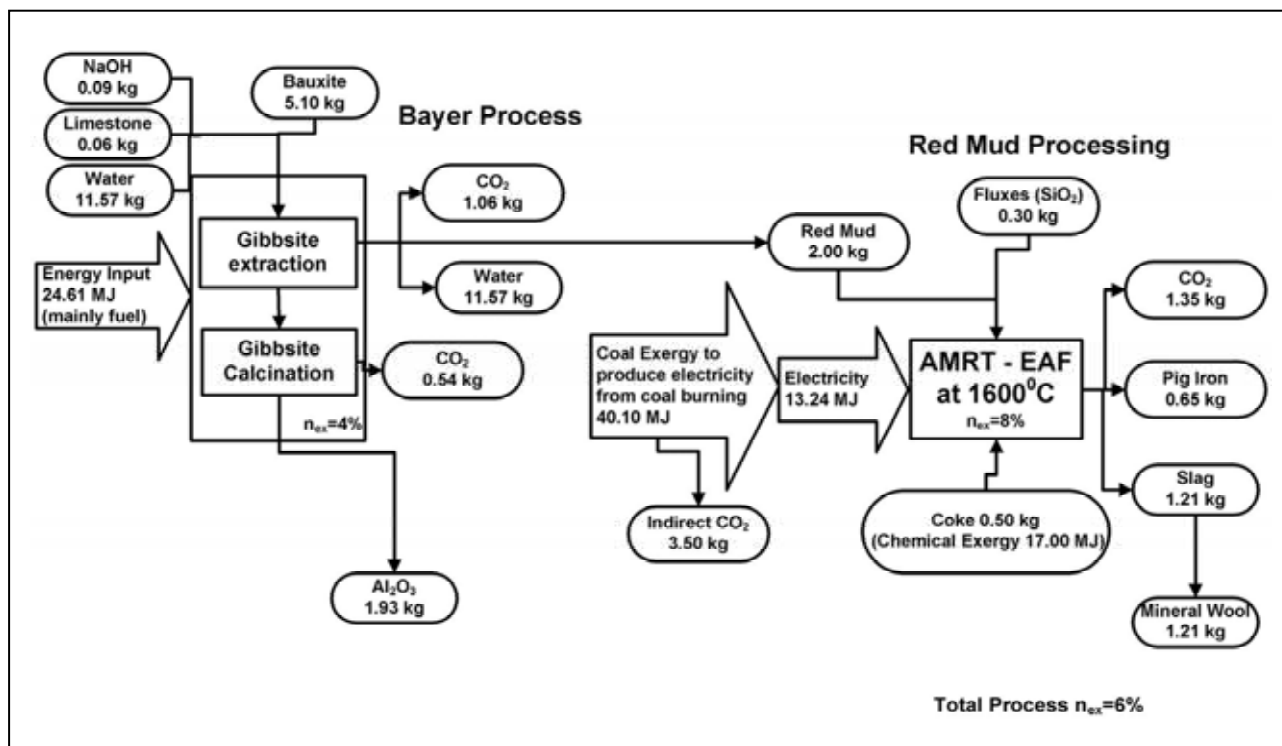


Figure 4: The complete bauxite exploitation schema. The hot off-gases from the red mud treatment process ( $\text{CO}$ ,  $\text{H}_2$ ) are assumed to be burned to  $\text{CO}_2$  and steam during the red mud drying stage

### 3 Preliminary Experiments

In order to achieve of proof-of-concept, using the materials described in Tables 1 to 3 a preliminary batch experiment of the red mud treatment process was conducted in a 750 kVA AMRT-EAF located in South Africa. The melt temperature during operation was measured at 1608 °C and at the end of the experiment two distinct liquid phases were formed and poured sequentially. Part of the slag phase was fiberized using a high speed air/water jet.

Due to the operational conditions of the plant, the sampling of the produced material was limited. Pig iron samples were analyzed in atomic adsorption spectroscopy after fusion of pig iron with dilithium tetraborate ( $\text{Li}_2\text{B}_4\text{O}_7$ ) and dissolution in hydrochloric acid, while carbon and sulfur content was determined in LECO equipment and the results of these analysis are presented in Table 5. Samples from the slag phases produced were chemically analyzed using X-rays fluorescence and





the results of this analysis are presented in Table 4. During this preliminary experiment no sampling of the gaseous phase was possible.

Table 4: Slag phase, weight: 603.83 kg

	Thermodynamic prediction	Experimental result
Chemical Species	in % wt./wt.	in % wt./wt.
Al <sub>2</sub> O <sub>3</sub>	33.31	33.18
FeO	0.12	0.65
CaO	19.68	20.23
SiO <sub>2</sub>	34.39	36.15
TiO <sub>2</sub>	8.90	3.89
Na <sub>2</sub> O	3.37	1.21
MgO	0.16	0.15
K <sub>2</sub> O	0.22	0.14
S	0.13	–
Total	100.00	95.60

Table 5: Pig iron, weight: 325.82 kg

	Thermodynamic prediction	Experimental result
Chemical Species	in % wt./wt.	in % wt./wt.
Fe	95.18	90.455
Si	3.29	5.145
Ti	0.15	–
S	0.03	0.032
C	1.04	4.368
P	0.30	–
Total	100.00	100.00

Table 6: Off-gases, weight: 443.76 kg

	Thermodynamic prediction
Chemical Species	in % vol.
CO	71.28
H <sub>2</sub>	28.24
Other gases	0.48
Total	100.00

The results of the pig iron analysis show large concentrations of carbon and silicon in the pig iron phase. The presence of carbon should be attributed to the extra carbon introduced in the system during furnace start-up (coarse coke was used), while the presence of silicon could be the effect of partial dissolution of the refractories used in the furnace bowl. The results of the slag phase analysis show small deviations (except in the case of the TiO<sub>2</sub> oxide) from the model results. However, any



conclusions would be premature as the sampling of the slag produced was limited and the method of chemical analysis used, was intended for qualitative and not quantitative analysis.



Figure 5: Photograph of inorganic fibres produced from the red mud treatment process slag

The inorganic fibres produced from the slag (Figure 5) were examined with scanning electron microscopy, in order to assess the physical qualities of the fibres (Figure 6 and 7). As seen, fibres with diameters less than  $20\ \mu\text{m}$  were mostly formed, along with some substantially thicker fibres ending in oval shaped slag inclusions.

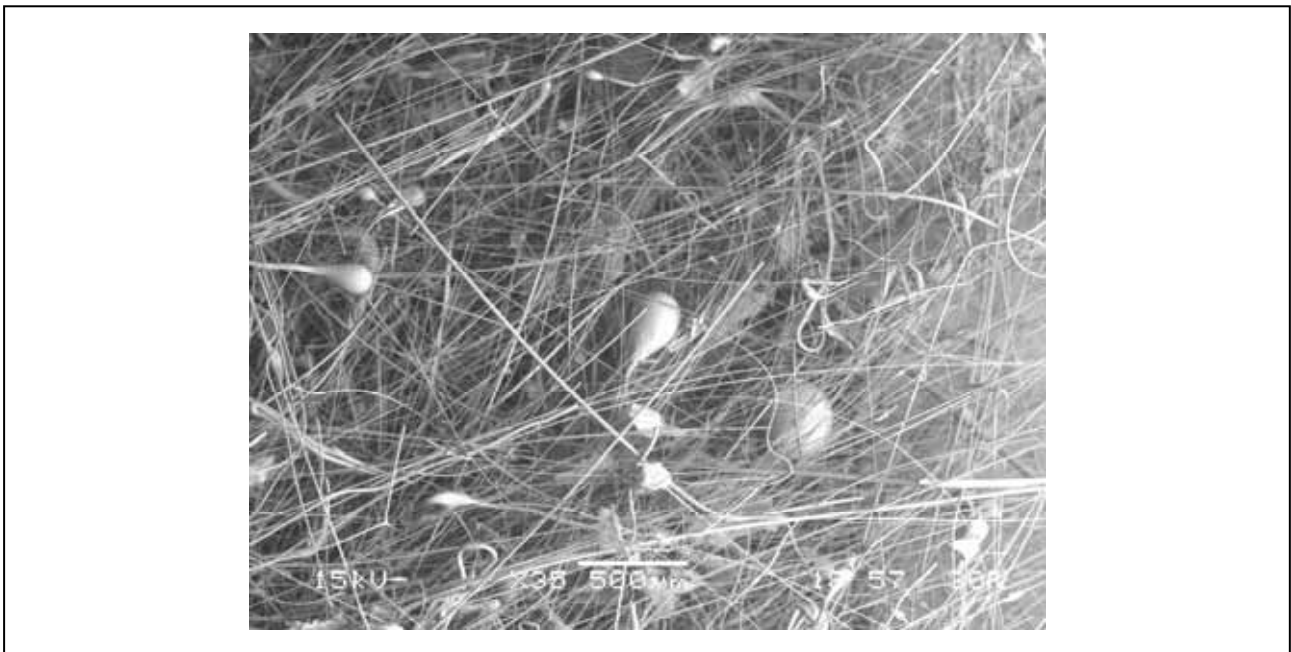


Figure 6: SEM photograph of inorganic fibres produced from the red mud treatment process slag, magnification  $35\times$



Figure 7: SEM photograph of inorganic fibres produced from the red mud treatment process slag, magnification 150×

Such imperfections, caused by the slag freezing prior to the completion of the fibre formation, can be attributed to the lack of an automated system during this preliminary experiment. In general, it is apparent that the slag from the red mud treatment process can be fiberized.

## 4 Conclusions and future work

Thermodynamic analysis and preliminary experiments have proven that the reductive smelting of red mud at 1600 °C in an AMRT-EAF, using carbon as reducing agent and appropriate fluxes to regulate the composition of the generated slag, can produce pig iron and a viscous slag that can be converted into glassy fibres suitable for mineral wool production.

From a resource management point of view the exergy analysis of the process shows that even though it is based on the utilization of an energy intensive EAF, it has the potential to increase the exergy efficiency of an alumina plant by exploiting in full the bauxite ore. No solid or liquid wastes are generated in the process, while the direct CO<sub>2</sub> emissions are relatively low (0.67 kg/kg of red mud).

For the primary aluminium industry the proposed Bauxite exploitation schema is expected to be highly profitable as it will replace the costly disposal of the red mud waste with two valuable products. Key economical aspect of the process is the production of the mineral wool, which is a versatile product with multiple applications and though this novel process is produced at a 70 % lower energy cost, thereby providing the industry with a significant commercial advantage.



However, before this process can be industrially implemented, further research and experiments are needed in order to refine the qualities of the produced products. The chemical and physical properties of the produced inorganic fibres, which depend greatly on the fluxes used in the reductive smelting, have to be optimized in order to meet market standards. Additionally, the composition of pig iron, especially with respect to silicon content as well as the utilization of the off-gases in the red mud drying stage have to be further studied.

The optimization and industrial demonstration of this process is one of the key goals of FP7 ENEXAL project which is currently underway.

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The preliminary experiment described in this work, were conducted in the AMRT facilities in South Africa in co-operation with Aluminion S.A. of Greece.

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