

BR10 - Flowsheets that Can Turn BR into Building Materials and Save CO₂

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Abstract

Within the EU-funded ReActiv project (GA No. 958208), significant efforts have been undertaken to convert bauxite residue BR into valuable building materials for the construction industry. In this study, the environmental implications of different BR conversion projects, focusing on co-calcined BR, Vitrified BR and iron-free BR slag by smelting were investigated. The CO₂ emissions per ton of product for each processing route were estimated. Additionally, to provide a basis for comparison, the current CO₂ emissions associated with the production of 100% cement clinker, which serves as the conventional material in the cement industry is presented. The advantage of this approach is that the BR contains only a very low content of chemically-fixed carbonates that will be released during its thermal treatment procedure, as is the case with limestone during the clinker production process. Consequently, replacement of standard clinker by a ReActiv SCM (Supplementary Cementitious Material) has the potential for a direct reduction of the overall CO₂ footprint in the cement industry. The paper in hand shows flowsheet concepts for the three BR processing technologies and discusses mass and energy requirements. It also shows the available state-of-the-art equipment such as multiple hearth furnaces, rotary kilns, submerged arc furnaces and treatable mass streams. An indicative SWOT analysis for each technology together with a business concept summary will also be presented. The aim is also to show a possible path in how two or even three currently parallel-acting industrial sectors, i.e., alumina, cement and steel could reduce their overall environmental footprint through smart cooperation.

Keywords: Valorization of bauxite residues, flow sheet, cement substitute, SCM, pig iron coproduction, industrial symbiosis.

1. Introduction

Advancements in sustainable practices within the construction industry are of the utmost importance in the quest to mitigate environmental impacts. 30 billion tons of concrete used each year are responsible for at least 7 to 8% of global CO₂ emissions [1,2]. In this context, the EU-funded ReActiv project (GA No. 958208) has made significant strides towards transforming bauxite residue (BR) into valuable building materials and helping to reduce the CO₂ footprint of the building industry. Additionally, the valorization of BR represents a significant improvement in the alumina industry footprint, since over 160 million tons of BR are being produced every year globally. This paper presents an overview of some of the current project's key findings and technologies developed, with a particular focus on the potential to reduce the carbon footprint of the cement industry.

Traditionally, the addition of small amounts of BR to the clinker production process, as alumina and iron oxide source, has been a common practice, with approximately < 3% weight utilized. However, through the innovative approaches developed by the ReActiv project, BR can be transformed into a Supplementary Cementitious Material (SCM) and added as a clinker substitute in concrete. Clinker replacement levels of up to 30% and beyond can be achieved with the BR SCMs, depending on the specific system and the properties to reach.

One crucial advantage of this approach lies in the absence of chemically-fixed carbonates within bauxite residues. Typically, a significant amount of CO₂ is released from limestone during the thermal treatment process within standard Portland cement production. The current values of CO₂ emissions per kilogram of cement clinker produced vary depending on several factors, including the specific production processes and technologies used. On average, the production of 1 ton of cement clinker emits approximately 0.84 tons of CO₂ [3]. As a result, every ton of standard clinker replaced by a ReActiv binder translates to a direct reduction in the overall CO₂ footprint within the cement industry.

The ReActiv initiative explores three distinct technologies to harness the potential of bauxite residue:

- Co-calcination of BR with kaolin at approx. 700 °C
- Vitrification of a mix containing at least 70% of BR and corrective components such as C, SiO₂ and/or CaO above 1250°C
- Smelting and granulation of a mix containing BR, 15-20% CaO and 15-20% C to reduce iron above 1600 °C and recover an iron-free amorphous material.

In addition to the technical aspects, the paper concludes with an indicative SWOT analysis for each technology and a summary of the business concept, emphasizing the potential for collaboration among parallel-acting industry sectors to collectively reduce their environmental footprint. Through this work, a viable pathway is presented towards a more sustainable future, where mutually beneficial cooperation leads to tangible environmental gains in multiple industries.

1.1 Co-calcination of Bauxite Residue Introduction

The chemical basics of co-calcination of BR together with kaolinite were developed by Danner et al. [4] and in previous ReActiv work [5] showing that low-grade kaolin and its use as a SCM was presented at the ICSOBA 2022 in Athens [6]. Work is ongoing and reaching out to process

optimization. The chemical challenges solved at the laboratory scale are now replaced by practical process issues. The Mytilineos team is executing a pilot test at a 500 kg/h scale, intending to reach an industrial scale level; in this work an indirect heated rotary kiln is used.

In the co-calcination process, a mixture of BR with kaolin is heated up between 700 to 750°C, forming reactive phases which lead to an SCM [5] without emitting any direct CO₂ from the processed materials. The product generated can have, depending on the process atmosphere, a red or black colour that also remains in the finally generated mortar and final construction product. Also considering marketing perspectives it can become an advantage if these colours are perceived by the user as a more environmentally friendly building material.

Mortar tests were performed, based on a mixture containing 50% BR and 50% kaolin (on a dry basis). 30% of clinker was replaced by this material and a strength of 46-52 MPa could be reached after 28 days. Especially the strength of 26-32 MPa after 2 days shows that the unique selling proposition of co-calcined BR towards other SCMs is mostly exhibited at an early age. These values are a promising first step for future industrial implementation.

1.2 Vitriified BR (VBR)

The vitrification technology at a temperature range of about 1200 to 1300 °C with common additives allows the full use of 'as-obtained' bauxite residue in the production of novel supplementary cementitious materials (SCM). Published reports by Yiannis Pontikes and coworkers demonstrate [7,8,9] the activity of the VBR as a SCM

At the Bauxite Residue Valorisation and Best Practices Conference 2022, an overview of BR vitrification was presented including the successful pilot scale production (2 t in total treated material during the tests), as well as the performances of binders that were produced with VBR [10]. At this point, the focus lies on the optimization of the mix designs (for firing as well as for mortar recipes) as well as on alternative up-scaling approaches (e.g., retrofitting of a cement rotary kiln).

In summary, the press-filtered bauxite residue from Mytilineos is mixed with coke (or any other source rich in carbon, such as biochar), limestone and a silica source. A pelletizing step ensures uniformity and avoids dusting at the vitrification step. The pelletized material is then partially molten at a temperature of 1200 – 1300 °C. The execution of the melting step in a top-blown rotary converter (total production of about 2 t at RWTH Aachen) has already been demonstrated under the H2020 REMOVAL project (n° 776469); this process step uses a rotary kiln. The partly molten material is subsequently quenched to transform the melt into an amorphous glass phase (content > 70 wt%), which after size reduction, serves as a novel SCM with a reactivity superior to calcareous fly ashes and close to ground-granulated blast furnace slag.

Direct use as a SCM together with cement clinker has been demonstrated and a strong synergy with limestone has been observed, similar to limestone calcined clay cements. This allows high replacements of clinker (e.g., a strength class of 42.5N was reached with only 40 wt% of cement clinker in the mix design). VBR is black in colour due to its chemical composition and iron oxidation state (one crystalline spinel solid solution, e.g., magnetite Fe₃O₄ is present). This characteristic also serves as a visual criterion for identifying this type of material. Cement produced with VBR has thus a darker grey appearance than other cement systems.

1.3 Iron-free SCMs by BR smelting

This approach necessitates substantial investments in terms of process equipment requirements and operates at the highest temperature compared to the other methodologies investigated.

Nevertheless, it yields pig iron as a valuable co-product. The advantage of pig iron lies in its standardized nature, offering the potential to compensate for the additional production costs. By utilizing the produced pig iron and fluxing the slag to produce a composition suitable for the production of an SCM, the full utilization of the BR feed in the process is achieved. The fluxed added to the smelting process (CaO and SiO₂) were studied to produce an SCM with a composition close to that of the blast furnace slag, which is already used as an SCM in concrete production. Figure 1 below, shows different BR slag compositions tested on a laboratory scale at NTUA. The S5 formulation has achieved the most promising results when substituting 20% clinker in cement mortars. The present paper aims to provide insights into the potential realization of an industrial process configuration for generating iron-free SCMs from BR and pig iron.

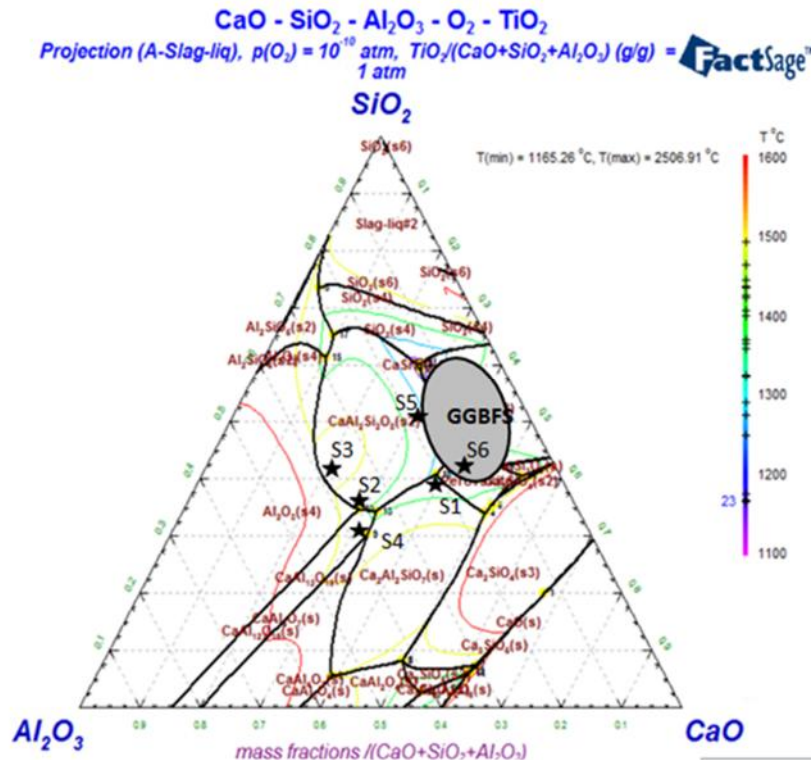


Figure 1. BR slag systems tested concerning the respective GGBFS (ground blast furnace slag) compositional range.

1.4 Perspective of Utilization

As a general comment, Holcim sees applications of these different materials as a substitute to current SCM such as fly ash, and blast-furnace slag which are expected to decline in availability in the near future. Construction cements are of course the first target due to the large market volume. However, the exact types of cement that could be developed with these materials are still under investigation and the durability work that will be performed at the University of Aachen will provide more insights on that question.

2. Technology Evaluation

2.1 Co-calcination of Bauxite Residue Technology

Figure 2 shows the simplified process flow diagram of the BR co-calcination approach. At the takeover point filter pressed BR and kaolin are brought to the system.

Filter-pressed BR has typically a moisture of about 25 %wt. Due to this fact, it is most likely that the plant will be installed in the immediate vicinity of the Bayer production facility. The unnecessary transport of water contained in the BR would have a negative effect on the economic and the ecological balance. The water brought to the system has a direct effect on the overall energy balance and there will be further room in the future for energy integration and optimization on that issue.

BR is already a fine material (median particle diameter typically between 2 to 6 μm) and the kaolin has to have a particle size distribution PSD also in the mentioned range. The process flow diagram shown assumes that a kaolin having the mentioned PSD is received and just mixed and no milling is required. If only coarse material is available, a milling procedure has to be added before mixing is executed.

For the calcination and activation, it is currently assumed that the thermal treatment is done in a Multiple Heard Furnace (MHF). The MHF compared to a direct-fired rotary kiln allows for better temperature control and also for the treatment of dusty material. The counter-current operation ensures a kiln's internal pre-heating of the material and pre-evaporation of included moisture together with lower off-gas temperatures. The material itself is moved by an air-cooled rack through the kiln which mixes and moves the material gently through the unit. The excellent control of the temperature profile is an additional benefit. The cooling air of the rack which is used for moving the material in the MHF is used as preheated combustion air.

The generated co-calcined BR leaves the MHF at the bottom and after cooling is ready for purpose. If needed, further mechanical treatment can be applied on-site or also in the cement plant. Generated dust emissions are separated in a dust filter and directly returned to the MHF.

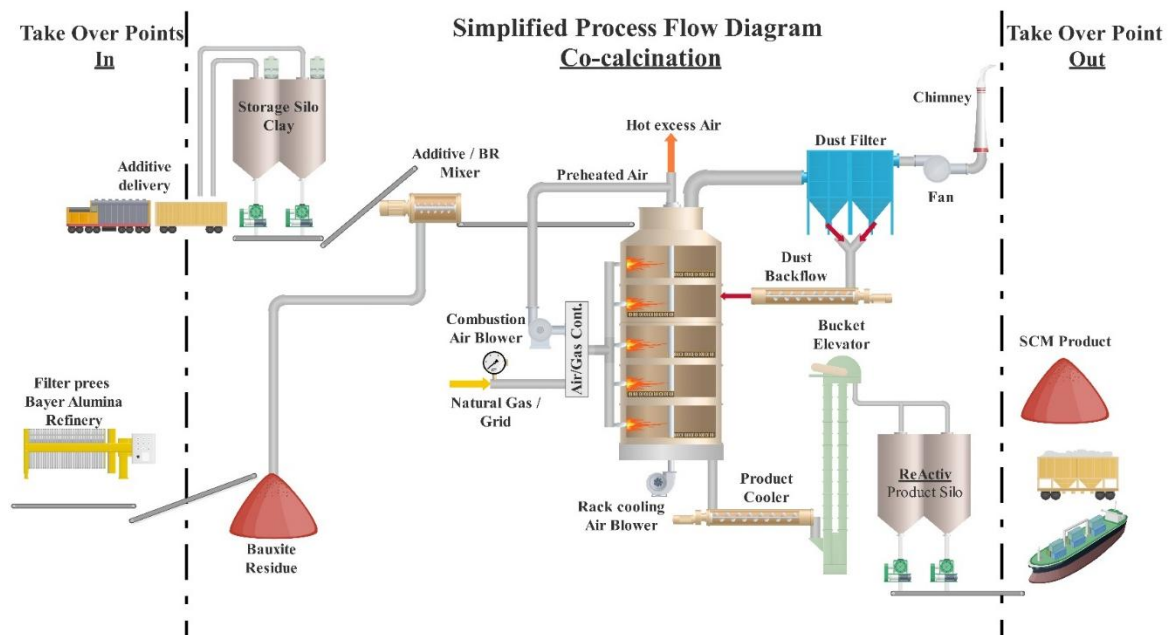


Figure 2. Process Flow Diagram BR Co-calcination.

The simplified mass balance shown gives a first idea regarding the thermal energy consumption of the co-calcination process. The approx. 38 t/h product reflects the by FLSmidth indicated max. throughput manageable by a state-of-the-art standard MHF system of 900 t/day. This assumption has to be verified for each project and for each BR used by running pilot scale tests. The use of a MHF allows a flexible and accurate temperature profile to be achieved for the product but e.g.

input moisture and specific feed material properties may lead to the requirement of a fine-tuning procedure.

Considering an annual uptime of the system of 8200 h per year leads to the following input and output figures:

BR wet consumption ~ 213 kt per kiln
 Kaolin consumption ~ 160 kt per kiln
 Product ~ 312 kt per kiln

The process required thermal energy was estimated at 1072 MJ/t of the product (25% BR moisture, dry kaolin).

The preliminary assumed natural gas consumption for the shown balance case results in a CO₂ output of approx. 30 Nm³/t of product or approx. 60 kg/t of product.

Considering a simplified case evaluation besides the thermal heating, all other electrical consumers in the process can be operated with CO₂-neutral electricity a significant CO₂ reduction is reached.

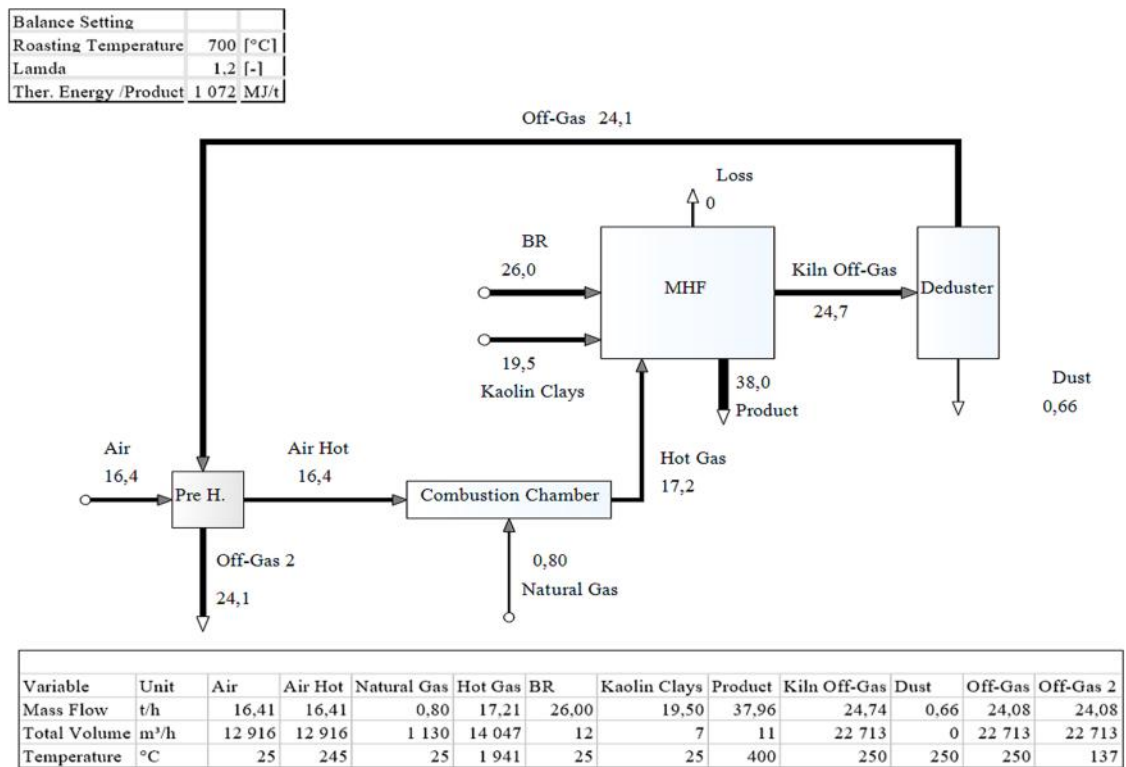


Figure 3. Mass balance assumption for largest standard MHF unit available ~ 900 t/day product.

Within the balancing procedure also a parameter variation regarding the moisture content of the BR was executed. It was shown that the water content has a significant impact on the overall energy consumption of the co-calcined material. Regardless of whether the moisture is introduced by the BR or the kaolin, it does not have a significant impact on the system. Due to lower temperature levels and less possibility of internal energy recovery, it is more relevant for the co-calcination case but similar effects are seen for all of the three cases.

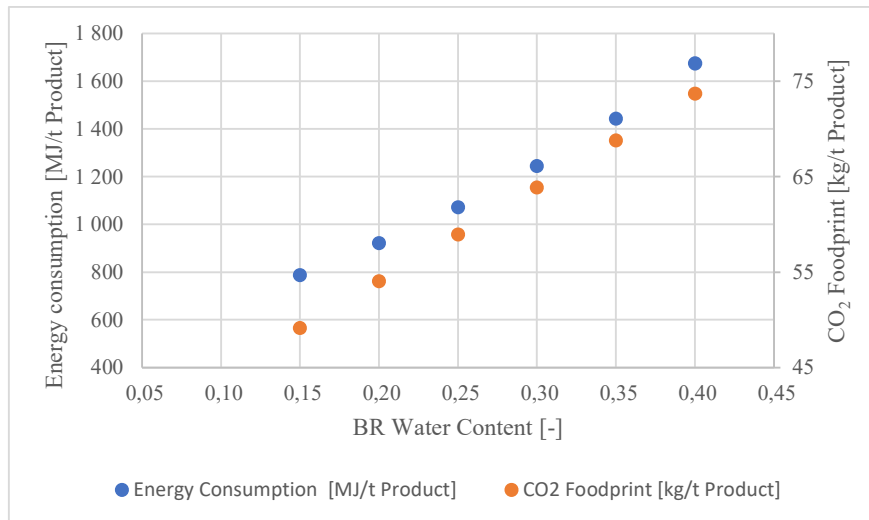


Figure 4. Energy Consumption and CO₂ Footprint.

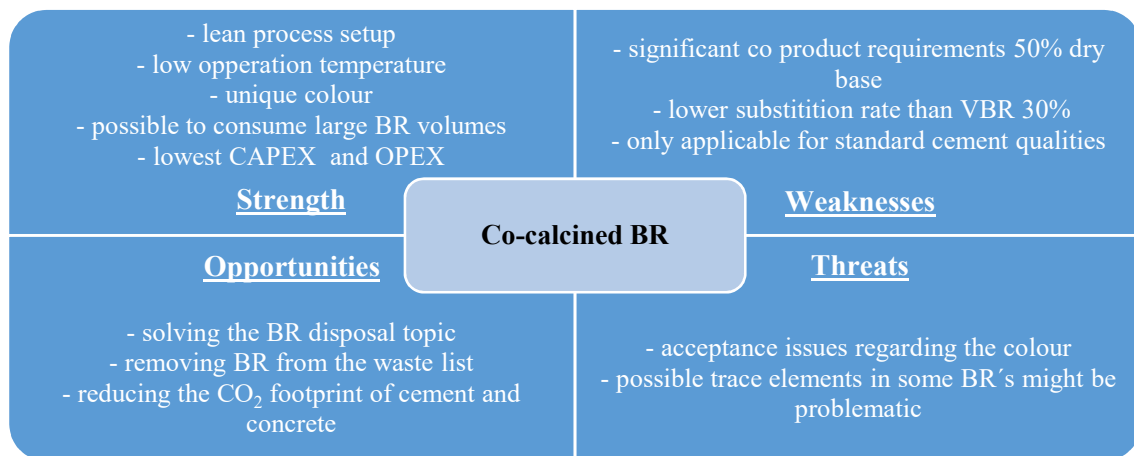


Figure 5. SWOT BR Co-calcination Case.

2.2 Vitrification of Bauxite Residue Technology

The process flow concept for the production of vitrified BR is shown in Figure 6. Filter-pressed BR is mixed with a silica source and milled limestone and coke. The moisture content of the final mixture must allow a smooth pelleting procedure. The addition of some binder in combination with a moisture adjustment also could be necessary. The specific mix design has to be found for each BR during some piloting tests for plant design. It is important to note that the process can work for any iron-rich (> 40 wt% Fe₂O₃) BR with appropriate fluxing. For the case shown, a simplified and general approach was chosen.

The pellets are transferred to a direct-fired rotary kiln using a Lepol preheater as the first production step. The main reason for using this old preheater type is to reduce the dust formation in the system when drying the pellets and to reach some mechanical strength before the material enters the rotary kiln. Within the rotary kiln, the mixture is partly to fully molten at a slightly reducing atmosphere and a temperature level of 1200 to 1350°C. The Lepol preheater was originally developed for the cement industry so a revival in the industry might be easily possible. The off-gases are connected to a gas-cleaning system. It would be possible to use an air quench/granulation system to rapidly reduce the temperature of the molten material in order to generate glassy and reactive phases. Also heat recovery for drying of the input material (BR) is

an option to utilize by quenching generated hot off-gases. After quenching the product is directly milled on-site or later in the cement mill.

Depending on the binder mix design and required strength class, it is possible to use the vitrified BR in different contents in alkali-activated binders (inorganic polymers) (100 wt% of VBR), in hybrid cement (50-70 wt% of VBR) or in a blended cement system (20-50 wt% of VBR) The resulting product colour depends on the content of VBR and ranges from dark grey to black.

In blended cement, very promising strength activity indices of >70 % after 2 d and >80% after 28 d were reached, in a mix consisting of only 53 wt% cement, 29 wt% of VBR, 15 wt% limestone and 3 wt% of gypsum [11,12].

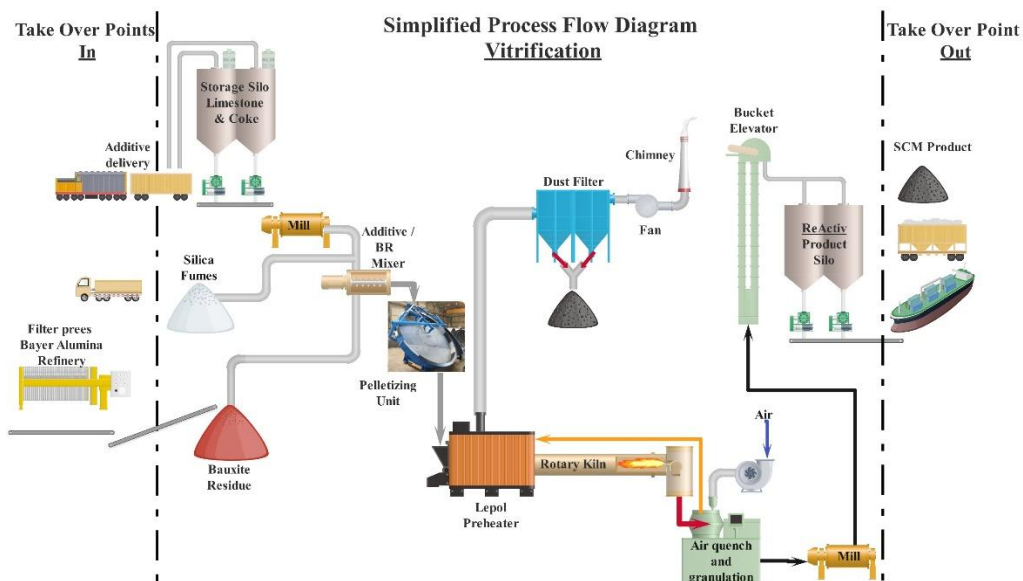


Figure 6. Process Flow Diagram BR vitrification.

The BR vitrification case evaluation has fewer throughput restrictions than the MHF approach. The equipment used is similarly in operation in the steel and cement industry and is already tuned for large throughputs. The balancing is still based on a large number of estimations that need to be finetuned during a possible implementation of the process in the industry. The paper aims to provide an overview of this very promising process concept for turning BR into a low CO₂ SCM.

Figure 6 shows an indicative mass and energy balance not considering the phase change energies. The assumed input for this case is 100 t/h wet (25% moisture) BR. To reach the required properties the additives limestone, silica and carbon are required. The total solid input feed of ~ 128 t/h wet base becomes ~94 t/h SCM product.

Considering again an uptime of 8200 h per year the following volumes will be reached:

BR consumption	820 kt
Coke consumption	17 kt
Limestone Consumption	110 kt
Silica Consumption	101 kt
Product	770 kt

The process thermal energy requirement was estimated at approx. 2730 MJ/t of product.

The preliminary assumed natural gas consumption for the shown balance case plus the CO₂ from Limestone and coke results in a CO₂ output of approx. 132 Nm³/t of product or approx. 261 kg/t of product. It is again assumed that the electrical consumers in the process are operated with CO₂-neutral energy and there will be no contribution to the CO₂ footprint. Even when the assumed value is higher compared to the previous case, it's still significantly lower than the emissions from OPC. Additionally, the grinding energy needed is close to Ordinary Portland cement and thus lower than granulated blast furnace slag when VBR is milled with grinding aid. Considering higher substitution rates the overall CO₂ comparison of those SCM / OPC systems has to be made when the recipes are finalized.

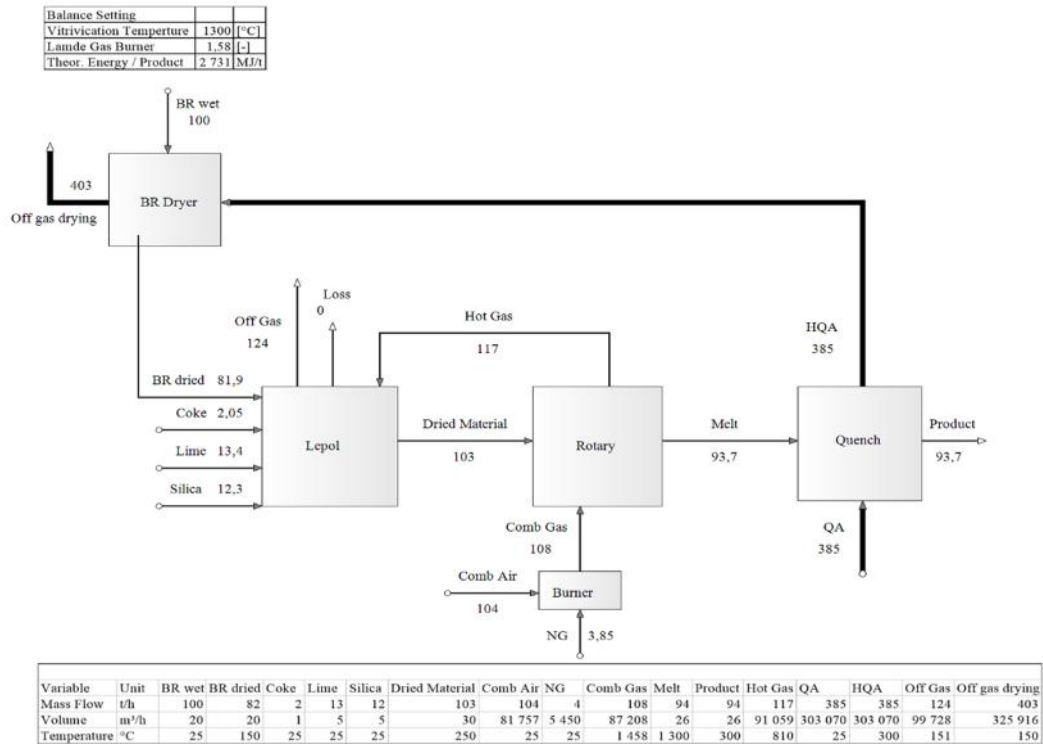


Figure 7. Mass balance assumption BR vitrification case.

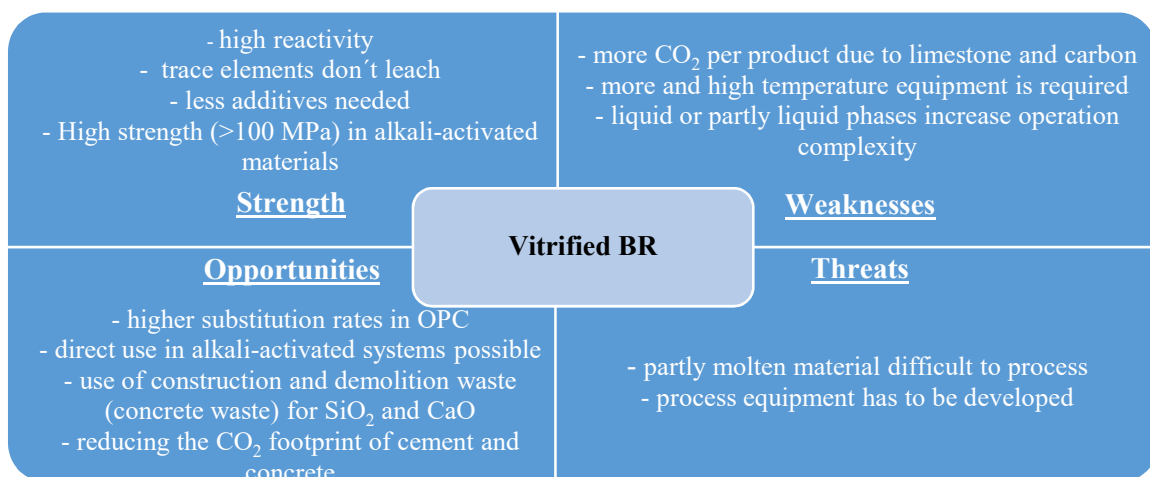


Figure 8. SWOT BR vitrification case.

2.3 Iron-free SCM from BR Smelting Technology

The iron-free SCM is produced through carbothermic smelting of BR to produce pig iron and slag. The process flow diagram presents similarities with BR vitrification. The main additional equipment is the required submerged arc furnace following the rotary kiln. There are also overlaps with the hot section of the Pedersen process [13] investigated during the EU-funded ENSUREAL project (GA No. 767533) [14,15] considering a slightly different slag design as well as fast instead of slow cooling.

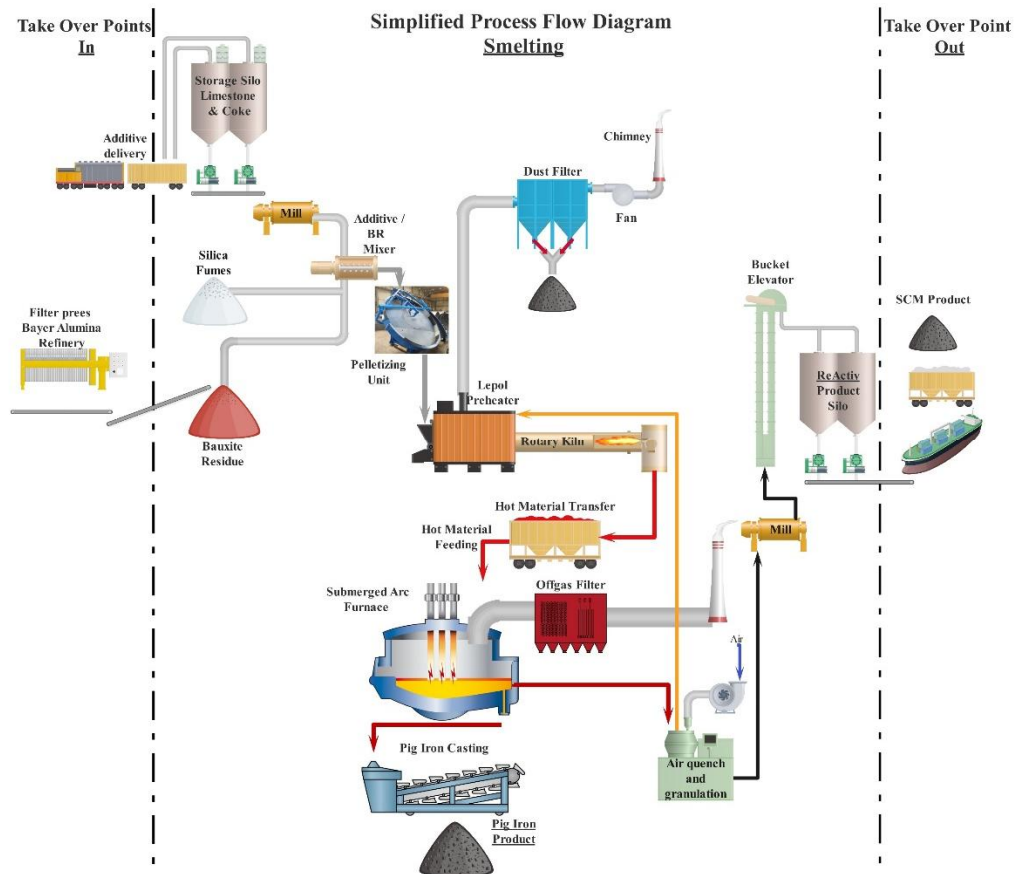


Figure 9. Process Flow Diagram BR smelting for iron-free SCM.

Within this process, shown in Figure 9, a mixture of BR, limestone, silica and coke is generated and again pelletized to reduce dust formation in the rotary kiln and the SAF. The pellets are dried and hardened in the Lepol preheater using the hot off-gases coming from the rotary kiln. In the rotary kiln a further preheating of the material together with an iron pre-reduction utilizing the added coke takes place. The hot material is afterwards transferred to the SAF where the final melting and phase separation (iron and slag) takes place. The iron phase is tapped off and poured off as a coproduct in a market-applicable form. The slag is treated with a fast air-cooling process to generate the required glassy and reactive aggregates. The final milling will lead to an SCM required particle size and product properties.

To be able to have qualitative comparable processes, the vitrification mass balance was extended by an SAF fully electrically operated. The recipes together with the process temperatures were adjusted together with the chemical reactions considering that e.g. a full reduction of the hematite to metallic iron takes place

The assumed input for this case is 100 t/h wet BR (25% moisture). To reach the required properties the additives limestone and carbon are required. The total solid input feed of ~ 149 t/h wet base becomes ~ 72 t/h SCM product and ~ 24 t/h pig iron. The increased input-to-product ratio in this case compared to the vitrification is mainly driven by increased flux addition. In other cases e.g. for Ca-Si enriched SCMs possibly can reach better results regarding this ratio as well as for the CO₂ by switching from limestone to CaSi-Minerals or mixtures thereof. Considering again an uptime of 8200 h per year the following volumes will be reached:

BR consumption	820 kt
Coke consumption	80 kt
Limestone Consumption	135 kt
Product SCM	590 kt
Product pig iron	198 kt

The average thermal fossil energy requirement for the production process was estimated to be ~ 3400 MJ per ton of product, considering both pig iron and SCM. Additionally, an extra ~ 870 MJ per ton were estimated for the electrical energy required to raise the temperature to 1580°C in the SAF.

Considering the preliminary assumed natural gas consumption for the specific balance case, along with the CO₂ emissions from limestone and coke, the approximate CO₂ output is calculated to be 243 Nm³ per ton of product or approximately 482 kg per ton of average product. The main energy source in the investigated case is coke (assumed as dry carbon) and only 15% of the fossil energy is provided by natural gas. It is important to note that the electrical consumers in the process are assumed to operate using CO₂-neutral energy, thereby making no contribution to the CO₂ footprint. However, it is necessary to account for the CO₂ generated in the production of metallic iron when making a fair comparison of the different SCMs investigated. This ensures an accurate assessment of their CO₂ requirements.

Exploring alternative fuels, and reduction agents, as well as CCS (Carbon Capture and Storage) and CCU (Carbon Capture and Utilization), are undoubtedly viable strategies for further reducing the environmental footprint. While these strategies are promising for industrial implementation, it is recommended to adopt a stepwise approach. Initially, SCM products need to be introduced to and accepted by the market. Once this milestone is achieved, further process optimization and reduction of CO₂ emissions can be pursued.

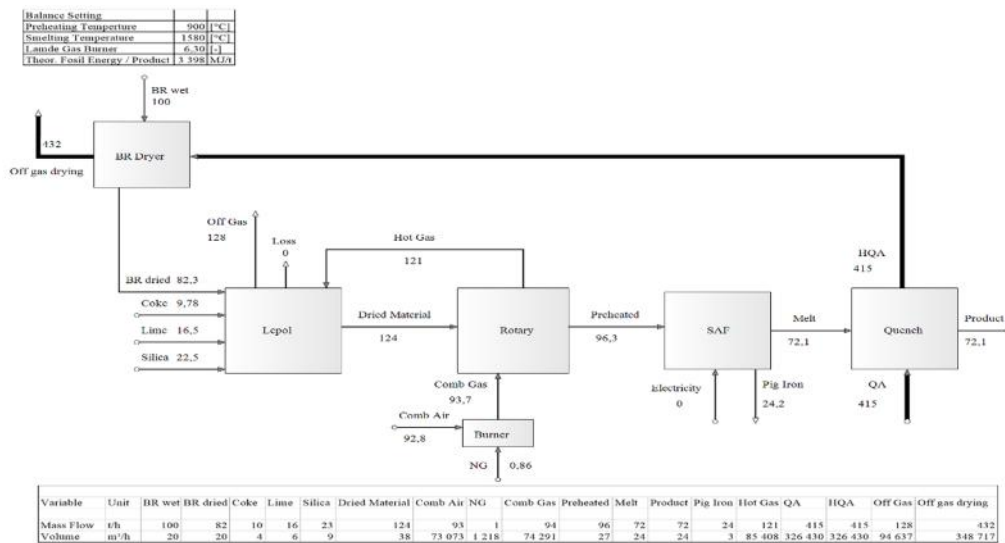


Figure 10. Mass balance assumption iron-free SCM by BR smelting case.

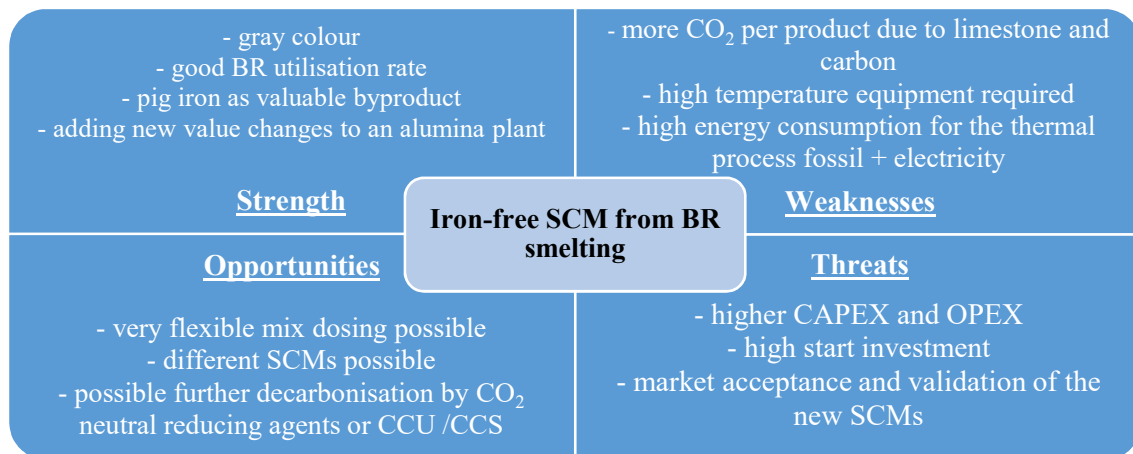


Figure 11. SWOT iron-free SCM by BR smelting.

3. Implications of CO₂ Emissions on Final Cement Product

Cement production is a significant contributor to global CO₂ emissions, primarily due to the calcination of limestone and the high-temperature processing of cement clinker. By substituting cement clinker with BR-derived slags, it is possible to reduce the CO₂ emissions associated with cement production. The different technologies considered in this study offer varying levels of emissions reduction.

It is shown for the co-calcination and the vitrification case that CO₂ emissions per kilogram of product are lower than those of conventional cement clinker. This implies that by incorporating these slags into cement, clinker content is reduced, and the overall CO₂ emissions of the final product can potentially be lower than that of 100% cement clinker. These projects hold promise for achieving significant environmental benefits in the cement industry. Utilizing the BR is an advantage for the alumina industry which allows a win-win situation.

In the iron-free SCM case, the separation of CO₂ emissions for slag and pig iron production requires a more detailed analysis. The CO₂ emissions associated with slag production are significantly lower than those of cement clinker, suggesting a potential for emissions reduction when using slag as a substitute. However, pig iron production is associated with higher CO₂ emissions compared to cement clinker. It is fair to allocate some of the process emissions to pig iron and compare these emissions with the current pig iron production and also with future projections for its production. This requires a more detailed environmental analysis. The overall impact of the iron-free SCM from BR smelting Technology on the final cement product's emissions would depend on the ratio of slag to pig iron used.

4. Business case

The business case development must be done case by case. Different BR, different availability of required additives, and different markets for the products make it impossible to give a general recipe for how these new technologies are brought in the best way to the industry. Also entering established production sites with some new Products / SCM requires significant efforts to convince the people in charge. In safety-related applications, e.g. SCMs in concrete, homologation and licensing time can take up to five years.

In any case, some challenges need to be taken into consideration:

- Due to the low price of the products, logistics must be optimized. Avoidance of BR transportation is important. A satellite or adjacent conversion plant to the alumina refinery seems to be the most proper location. However, proximity to the end markets is significant. In India, the proximity of alumina refineries with big cement plants is the key driver for achieving the high BR valorization rate.
- Profitability is sensitive to critical revenues and cost factors (CAPEX and OPEX). Long-term planning must be done carefully with a good understanding of factors' possible fluctuation range.

Therefore, one attractive strategy might be a joint venture approach where the benefiting groups from alumina, cement and even the steel industry join forces to have a new and independent corporation. For such a case the business would be based on leveraging BR disposal avoidance costs along with profits from SCM products to an attractive margin contribution with a reasonable payback period. Furthermore, if existing equipment (e.g. rotary kilns) in alumina or cement plants can be retrofitted for the developed process, OPEX and CAPEX may be reduced further.

5. Benefits to Society

The utilization of SCMs for substituting cement clinker is one of the most efficient ways to reduce GHG emissions. However, the most voluminous of those, e.g. GGBFS (ground granulated blast furnace slags) and Fly-Ash from blast furnace iron production and coal thermal power stations respectively, are becoming less available due to structural changes in steel and energy industries.

The utilization of BR-derived slags as cement clinker substitutes partially remedies some of the above challenge and indeed offers several potential benefits to society. Firstly, by reducing the reliance on traditional cement clinker, these projects can contribute to a substantial reduction in overall CO₂ emissions from the cement industry. This aligns with global efforts to mitigate climate change and transition towards a more sustainable future.

Furthermore, the implementation of these projects promotes circular economy principles by transforming BR, which are typically considered waste, into valuable construction materials. This not only reduces the need for raw materials extraction but also minimizes the environmental footprint associated with the disposal of BR.

Among the approaches examined, the Vitrified Project and the co-calcination Project demonstrate the most significant potential for benefiting society in terms of CO₂ emissions reduction and the utilization of BR. These projects offer the possibility of significant reductions in CO₂ emissions in the cement industry while utilizing a considerable proportion of BR.

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