A Comparative Study of Conventional Lime Kilns and Plasma Calcination: Techno-Economic Assessment and Decarbonization Potential

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Abstract: Lime production is essential in the chemical recovery cycle of chemical pulping mills, typically relying on fuel combustion and thus contributing to greenhouse gas emissions. While Nordic pulp mills mainly use carbon-neutral biofuels, future biomass scarcity underscores the need for sustainable biomass management and alternative lime calcination methods. Electrification presents a promising solution, as CO_2 emissions depend on the carbon intensity of the electricity grid, which increasingly relies on renewable sources. Electrified solutions offer chemical pulp mills the opportunity to function as biorefineries and potentially produce higher-value biofuels in a constrained market. Plasma calcination provides benefits over conventional lime kilns, such as faster reaction times, reduced reactor volume, and lower shell losses. This work develops mathematical models for conventional kilns and plasma calcination to evaluate their techno-economic feasibility and decarbonization potential. A sensitivity analysis identifies influential parameters, and energetic requirements for both technologies under different fuel scenarios are assessed along with CO_2 emissions and economic factors. Results indicate that while plasma calcination's current decarbonization potential depends on the electricity grid's carbon intensity, future projections show its competitiveness over conventional kilns, with significantly lower CO_2 emissions across regions. The economic viability of plasma calcination is further influenced by projected carbon prices and process parameters, which impact its specific electricity consumption.

Keywords: Plasma calcination, Decarbonization, Electrification of heat, Lime kiln, Energy modelling, Pulp and paper

1. INTRODUCTION

The pulp and paper industry belongs to one of the most energy intensive industries in the world. Globally, it accounts for around 5% of total industrial energy consumption and is responsible for around 2% of emissions related to industry (IEA 2020). Despite rapid developments in digitalization, the pulp and paper industry is growing with a rate of 2% annually and is projected to continue rising due to several sectors, such as packaging or tissue production, more increasingly relying on paper-based products (IEA 2020; Summanen 2022). Currently, however, the pulp and paper industry is regarded as not being on track to reach net-zero emissions in 2050 which are obligated by the European Green Deal and legally binding through the European Climate Law (EC 2020; IEA 2020).More efforts need to be made in order to reduce carbon dioxide emissions by moving away from fossil fuels and adopting new technologies in the production process. Electrification is seen as a promising option for supplying the necessary heating demand currently covered by fuels as carbon dioxide emissions are ultimately subject to the carbon intensity of the electricity grid. With an increased development of renewable and low carbon sources, electrification is therefore a crucial strategy to reach ambitious climate goals and contribute to the decarbonization of energy supply chains (IEA 2024).

One of the main heat intensive process steps in the pulp and paper industry is the production of lime within the chemical recovery cycle for chemical pulp mills. This is conventionally achieved in lime kilns that rely on fuel combustion to meet thermal energy demand. Lime kilns are primarily powered by fossil fuels such as natural gas or coal which is directly linked to carbon dioxide emissions (Falcke et al. 2017).Alternatively, biofuels can be utilized as a renewable energy source in lime kilns which is mostly the case in Nordic pulp mills. In 2020, 90% of the total energy used in Swedish lime kilns was supplied by biofuels, mostly tall oil pitch (63%) and bark powder or sawdust (24%) (Berglin and Schenck 2022). In Finnish lime kilns, roughly 45% of the energy was supplied by biofuels, mostly through gasified bark (18%), tall oil pitch (13%), and lignin powder (8%) (Berglin and Schenck 2022). Even though biofuels are considered a renewable energy source due to their participation in the carbon cycle, carbon dioxide emissions from combustion still occur (Newell 2010). Simultaneously, biomass demand is projected to increase significantly in the future, potentially surpassing the available supply (Material Economics 2021). This supply bottleneck may be further exacerbated by the European Parliament's environmental committee, which advocates for restricting the use of primary woody biomass for energy purposes, arguing that an ecological limit on biomass harvesting must be considered (Material Economics 2021; Svebio 2022). This would conclusively result in a significantly tighter market for biofuels and an urgent need to consider how to best use biomass in Europe. Pulp mills therefore can play a significant role in becoming bio-refineries by converting available biomass residues into higher-value biofuels while at the same time opening the door for electrified calcination solutions.

Plasma calcination is seen as a promising, electrically driven solution for lime production (Madeddu et al. 2020). It can offer several advantages over conventional lime production through lime kilns such as faster reaction times, reduced reactor volume and decreased shell losses (Andersson and Skogström 2020). It is, however, not yet commercially available for the pulp and paper industry and comparative studies to conventional lime kilns showcasing the potential of plasma calcination have not been conducted. The key question, therefore, is to evaluate the comparative benefits of plasma technology versus lime kilns under both current and projected future conditions.

This works aims to conduct a comparative techno-economic assessment between the plasma calcination technology and conventionally used lime kilns. Mathematical models for lime kilns and plasma calcination are developed to analyze the energetic requirements for both technologies under the investigation of different fuels. Corresponding carbon dioxide emissions are further calculated to assess the decarbonization potential of the plasma calcination technology and an economic assessment including operational and capital expenditures is performed.

2. PROCESS DESCRIPTION

Two technologies are being considered and compared for the production of lime, those being conventional lime kilns and plasma calcination. Energy models for both technologies are developed to estimate the energy and fuel consumption as well as production related CO_2-emissions under consideration of the relevant process steps and corresponding energy and mass balances.

Long rotary kilns are conventionally used in the pulp and paper industry for the production of lime. Fuel is combusted at the discharge of the lime kiln and its flue gases move counter-current to the flow of the lime mud, providing the necessary energy to cover the heating demand for lime production. In its core, a lime kiln can be divided into four heating zones as seen in Fig. 1. First, wet lime mud enters the kiln and is dried to remove all water. After that, the dried lime mud is heated up until it reaches the calcination temperature. The calcination reaction itself is endothermic and requires additional heat to occur. In a last step, the produced lime sintered by further raising its temperature and agglomerating smaller particles to bigger ones. The hot lime is then discharged and cooled while preheating a secondary air stream to reduce the fuel consumption of the kiln (Bajpai 2018).

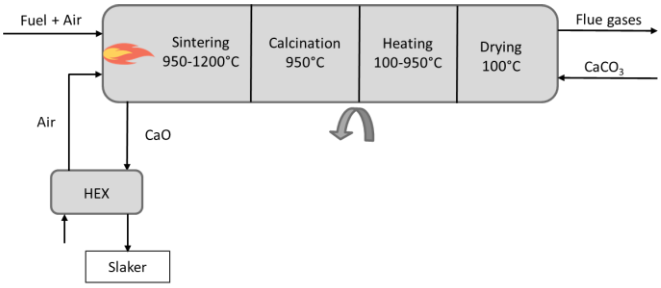

Fig. 1. Illustration of lime kiln with corresponding heating zones.

The plasma calcination technology aims to replace fuel driven lime kilns by providing the necessary heat for lime production with an ionised gas stream, also referred to as plasma, in a calcination reactor. The plasma stream itself is generated from an electric arc through which the gas stream passes, partially ionises and leaves as a gas-plasma mixture reaching temperatures of up to 5000 °C. The hot gas-plasma stream can then drive the calcination reaction (Andersson and Skogström 2020).

Figure 2 shows a simplified process schematic of the electric plasma calcination technology. Lime mud needs to be dried before entering the plasma reactor to reduce its electricity consumption as the evaporation enthalpy of water is comparatively high. For the scope of this work, a heat exchange network design is modelled in which the heat content of the hot discharge gases is used to dry the lime mud partially or fully. In the plasma generator, CO_2 is heated when getting in touch the electrically generated arc and leaves the plasma generator as a gas-plasma mixture to drive the calcination reaction in the plasma reactor together with the dried lime mud. The CO_2 stream after the plasma reactor needs to be cooled before the CO_2 stream is separated into two streams due to the additional CO_2 created during the calcination reaction. The first stream is compressed and recycled into the process while the second CO_2 stream is emitted and thus leaving the system boundaries. CO_2 created during plasma calcination is, however, of high purity meaning that the non-recycled CO_2 stream can be used in other process steps within the pulp production process, such as pulp washing (Bjotveit et al. 2003).

Plasma calcination can offer significant benefits compared to conventional lime kilns. Lime mud reburning with plasma can significantly reduce the process time of lime production from several hours in lime kilns to only a few seconds in a plasma reactor offering improvements in process control and reduced start and stop times. Additionally, equipment size is significantly smaller with a plasma reactor only having 1% of the volume of a lime kiln and no moving parts allowing for reduced operating costs and decreased shell losses (Andersson and Skogström 2020).

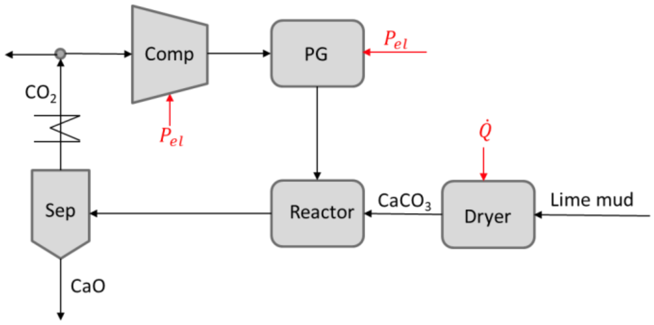

Fig. 2. Simplified process diagram for plasma calcination, adapted from (Andersson and Skogström 2020).

In order to quantify the energy demand of the conventional lime kilns and the plasma calcination technology, the two thermodynamic principles of energy and mass conservation are applied for every relevant component. Temperature dependant heat capacities are being considered as well as additional ionization enthalpy when modelling a phase change from gaseous to plasma state within the plasma calcination energy model. For the lime kiln model, different fuels can be investigated based on their molecular composition. Aside from the energetic requirements, both energy models further allow to estimate fuel and electricity consumption as well as corresponding CO_2-emissions to further assess the decarbonization potential of the plasma calcination technology and its economic competitiveness.

3. RESULTS

The developed energy models for conventionally used lime kilns and the novel plasma technology allow for analysis of energetic requirements as well as corresponding fuel consumption and CO_2-emissions. Both models were developed in Python 3.12.2. A list of relevant process parameters can be found in table 1.

Table 1. Process parameters

Parameter	Value	Unit	Source
Cooling temperature	200	°C	(Bajpai 2018)
Flue gas temperature	200	°C	(Lundqvist 2009)
Solids content	0.75	-	(Vainikainen 2021)
Sintering temperature	1100	°C	(Gulbrandsen and Stenqvist 2016)
Loss factor shell	0.1	-	(Lundqvist 2009)
Plasma temperature	3600	°C	(Andersson and Skogström 2020; Blackman 2024)
Reactor efficiency	1	-	(Bjotveit et al. 2003)

Figure 3 shows the sensitivity analysis results for the energetic analysis of both technologies. Key influential process parameters for the lime kiln model on its heat rate can be identified as the solids content of the lime mud entering the kiln, the flue gas temperature of the combustion products leaving the kiln and the loss factor accounting for heat losses through the shell of the kiln. In the plasma calcination model, partially ionized CO2 provides the necessary heat for the calcination process. Results indicated that the temperature range of the ionized CO2 has a significant impact on the electricity demand of the plasma generator, with a steep increase for plasma temperatures lower than 3000 °C. Additionally, variations in the sintering temperature have a higher impact on the specific electricity consumption of the plasma generator than on the heat rate of the lime kiln.

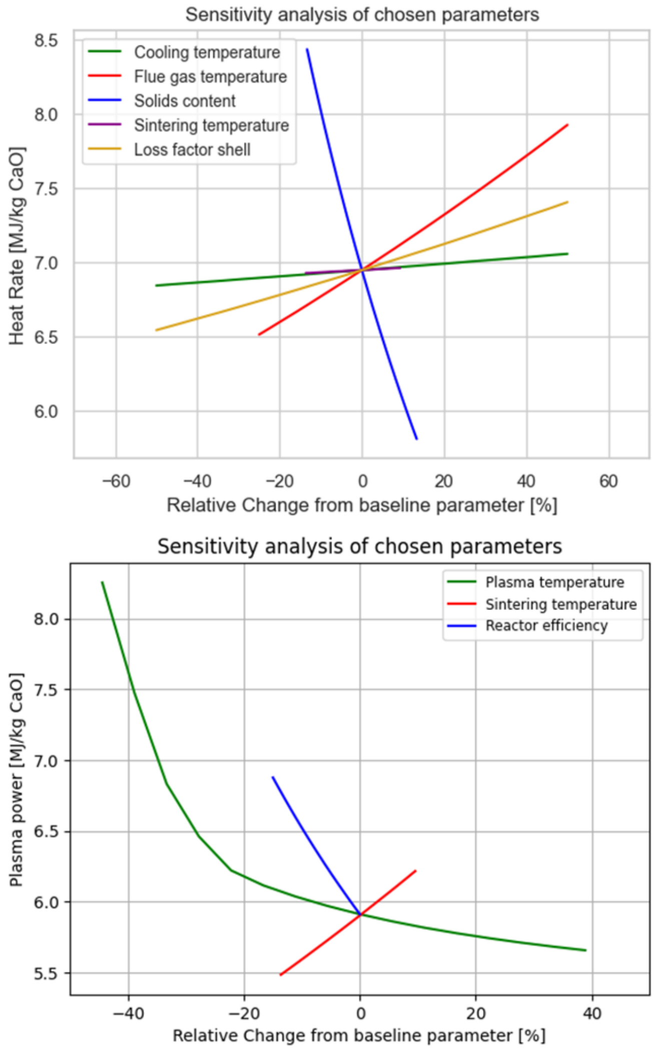

Fig. 3: Sensitivity analysis of chosen process parameters for the lime kiln model (top) and plasma calcination model (bottom). The values in the brackets of the legends refer to the corresponding baseline parameter.

The decarbonization potential of plasma calcination can be estimated with the modelling results generated by an in-depth sensitivity analysis in which the parameters described above have been cross varied. The conducted emissions analysis takes into account direct CO2-emissions from fuel combustion in lime kilns and indirect emissions for the electricity supply of lime production via plasma technology. For the lime kiln, three fuels have been analyzed those being natural gas (NG), tall oil pitch (TOP) and bituminous coal (BC). CO2-emissions from plasma calcination are ultimately subject to the carbon intensity of the electricity grid. As carbon intensities vary, three regions have been selected to assess the decarbonization potential of the plasma calcination technology those being the EU, China and Sweden. Current as well as projected carbon intensities for 2030 and 2050 are taken into consideration for the analysis. Figure 4 illustrates the decarbonization potential of the plasma calcination technology. The boxplots show the potential range of specific CO2-emissions based on the multi-way sensitivity analysis for the given regions and time spans. The rectangles across the figure represent the interquartile range of CO2-emissions for the analyzed lime kiln fuels. It can be concluded that the current decarbonization potential of plasma calcination is heavily dependent on the carbon intensity of the corresponding electricity grid. Countries with a higher carbon intensity, such as China, cause higher CO2-emissions from plasma calcination in comparison to the analyzed lime kiln fuels. Countries with a low carbon intensity, such as Sweden, however, cause plasma calcination to outperform conventional lime kilns from a decarbonization perspective. Future trends regarding carbon intensity further reveal the competitiveness of plasma calcination and lead to lower specific CO2-emissions for all analyzed regions in comparison to the analyzed lime kiln fuels in 2050.

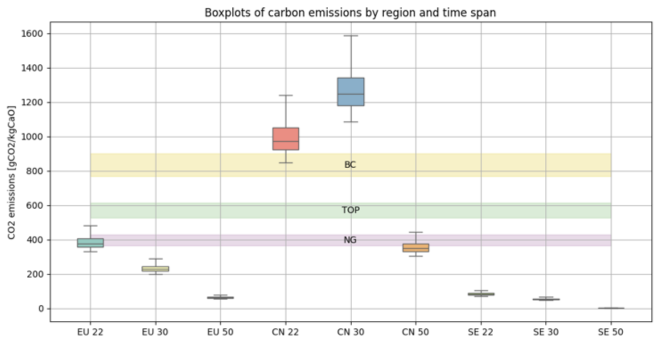

Fig. 4: Decarbonization potential of plasma calcination technology for different regions and time spans.

For the economic analysis, both operational and capital expenditure are being considered using the Net Present Value (NPV). Operational expenditures consider current and projected fuel and industrial electricity prices until 2050. Additionally, carbon prices are applied based on the EU ETS framework. For the capital expenditure, a 15 MW plasma generation system is considered with total investment cost for the plasma generator itself as well as auxiliary equipment. Based on the in-depth sensitivity analysis, this system would yield a lime production capacity of 121 – 245 t/day. This difference creates an upper and lower bound for the total investment costs of a lime kiln with equivalent production capacity. Figure 5 illustrates the NPV over a time span until 2050 as a function of the lime production capacity for two scenarios. The first scenario uses a high carbon price of 500 €/t in 2050 and the second one applies a more conservative carbon price of 200 €/t in 2050. Lower production capacities imply a lower electrical efficiency of the 15 MW plasma generation system while higher production capacities imply an increased electrical efficiency. For the high carbon price scenario both natural gas and bituminous coal can be outperformed by the plasma calcination technology even though higher electrical efficiencies of the plasma calcination system are required to result in a higher NPV than lime kilns fueled with natural gas. A lower carbon price requires higher electrical efficiencies of the plasma calcination system to be economically competitive with lime kilns fueled by bituminous coal while natural gas fired lime kilns always result in a higher NPV.

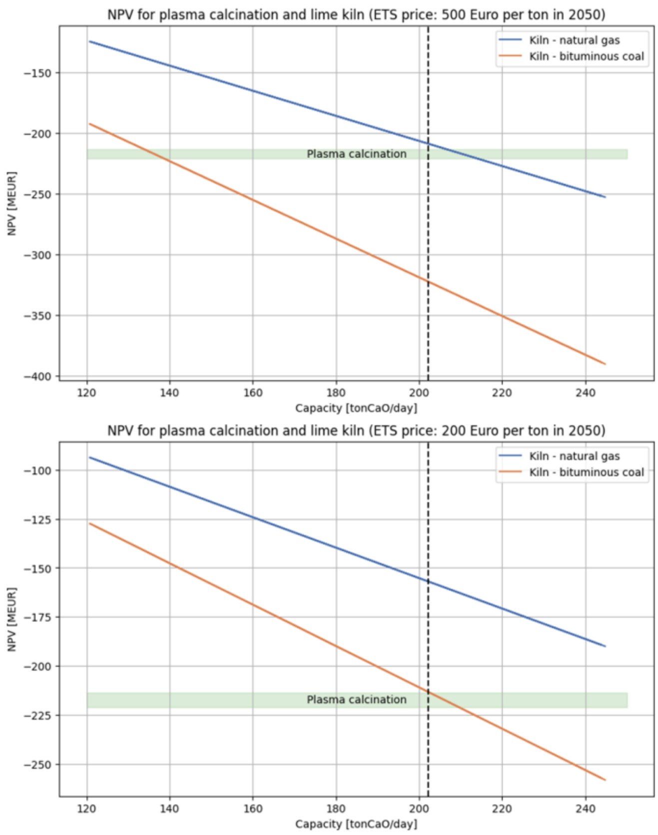

Fig. 5: NPV for plasma calcination system and lime kilns fueled by natural gas and bituminous coal for a carbon price of 500 €/t in 2050 (top) and 200 €/t in 2050 (bottom). The vertical line shows the median value obtained from the sensitivity analysis.

It can be concluded that the economic competitiveness of the plasma calcination technology is highly dependent on the projected EU carbon prices and the time of investment as a general increase of carbon prices is forecasted with a rather high uncertainty regarding the precise extend of this increase.

4. DISCUSSION

The primary objective of this work was to explore the potential of plasma calcination as a sustainable alternative to conventional lime kilns for lime production within the pulp and paper industry. This investigation was motivated by the industry's need to reduce greenhouse gas emissions and align with European and global decarbonization goals. The study examined the technological, economic, and environmental aspects of plasma calcination and compared those to conventionally used lime kilns. The generated results for the energetic requirements of both models are aligned with literature findings even though a data bottleneck for the plasma calcination technology has been identified. Plasma calcination for lime production in the pulp and paper industry is not yet applied on a commercial level and public availability of process flow sheets or process parameters is very limited causing the range of certain process, such as the plasma temperature, to be most likely wider than for a scenario in which this technology is well established in the industry. The decarbonization potential of plasma calcination is currently heavily dependent on the carbon intensity of the electricity grid. While the current decarbonization potential is geographically limited, the results show that future trends indicate plasma calcination to clearly outperform lime kilns from a decarbonization perspective. The economic competitiveness of plasma calcination is not only governed by the investment costs which are dependent on the selected process parameters but also on the operational expenses. The latter mostly depend on the projected carbon prices and corresponding policies such as the carbon leakage status of the pulp and paper industry or a potential inclusion into the Carbon Border Adjustment Mechanism.

Future work should explore enhancements and additions to the plasma calcination model developed in this work. The results generated in this work are based on simulation models. Especially for the plasma calcination model, for which a data bottleneck has been identified, experimental validation of the modeling results and potential adjustments would be desirable. Based on that, the plasma calcination model can further be enhanced by modelling the plasma generator and calcination reactor component in greater detail under consideration of dynamic heat transfer and different plasma torches. One major advantage of the plasma calcination system are fast ramping times which could be applied to fluctuating electricity prices to optimize lime production from an economic perspective. Additional gases other than carbon dioxide under different thermodynamic properties can further be investigated to increase the performance of the plasma calcination system.

5. CONCLUSIONS

This study demonstrates the potential of plasma calcination as a sustainable and efficient alternative to conventional lime kilns for decarbonizing lime production in the pulp and paper industry. While current adoption depends on the carbon intensity of the electricity grid, future projections indicate significant CO_2 emission reductions and competitive economic performance under anticipated carbon pricing scenarios. Plasma calcination offers technological advantages, including faster process times, reduced equipment size, and integration potential within biorefinery frameworks. Future efforts should focus on experimental validation, enhanced modeling, and dynamic optimization to fully realize the benefits of this promising technology.

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