Environmental Impact

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Green liquor dregs for carbon capture, utilization, and storage: initial LCA and economic analysis

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Abstract: Green liquor dregs (GLD) are one of the major inorganic residues generated during the production of pulp through the kraft process. Currently, GLD mostly end up in landfills. Research is underway to explore diverse alternative applications for GLD, and this study conducts an initial assessment to evaluate the feasibility of utilizing GLD as a material for carbon capture, utilization, and storage. The study aims to provide support for further experimental work in this area. Despite uncertainties, the carbonation of GLD demonstrates the potential for carbon capture, utilization, and storage. The results show that the environmental impact of the GLD carbonation process is predominantly influenced by the intended use of the final product (carbonates). Environmental benefits are estimated to range between 142 and 686 kg CO_2e/tonne of GLD, depending on the adopted scenario. The most environmentally advantageous option involves replacing materials with a high carbon footprint. However, options like using carbonates as construction materials, which necessitate energy-intensive drying, may only be economically viable with access to a low-cost energy source for drying.

Keywords: green liquor dregs; carbon capture; utilization; and storage (CCUS); life cycle assessment (LCA); economic analysis

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1 Introduction

Green liquor dregs (GLD) are one of the biggest inorganic residues of kraft pulp mills (Pöykiö et al. 2006). GLD is generated in the chemical recovery cycle of the pulping process during green liquor clarification (Ribeiro dos Santos et al. 2019). Presently, a substantial amount of these materials ends up in landfills (Kinnarinen et al. 2018), posing an environmental threat due to their elevated pH levels that surpass the tolerance range of aquatic life. Typically, the long-term pH tolerance range for aquatic life is between 6.5 and 9, whereas the pH levels of the GLD range from 10 to 13 (Bandarra et al. 2019).

The increasing costs associated with stricter environmental regulations and reduced space for disposal of GLD are driving the search for alternative ways of the GLD's utilization (Bandarra et al. 2019; Pöykiö et al. 2006). Some of the proposed applications encompass the usage of GLD as a neutralizing agent for acidic wastewaters (Pöykiö et al. 2006), a fertilizer (Mahmoudkhani et al. 2004), raw material for the ceramic industry (Ribeiro dos Santos et al. 2019), remedy for mine waste (Sirén et al. 2016), asphalt paving (Labart 1995) and a component for production of geopolymers (Sundqvist 2021).

Lately, there has been a growing interest in utilizing alkaline industrial residual flows for carbon capture, utilization, and storage (CCUS), driven by a range of favorable outcomes linked to this process (e.g., Ostovari et al. 2020; Sanna et al. 2014; Walker et al. 2024). In the current work, GLD is being explored for its application in carbon capture, utilization, and storage. The CCUS process facilitates the valorization of residual flows by reducing carbon dioxide emissions, cutting waste management expenses, and reusing the end products of carbonation, thereby decreasing the need for new raw materials. The process of carbonation can neutralize the high pH levels and alter the physical properties of the waste (Sanna et al. 2014). The CO_2 capture efficiency of the carbonation is lower compared to the frequently studied amine-based capture processes. Nevertheless, carbonation technology could offer more convenient localized carbon utilization and storage solutions, deriving benefits from the utilization of residual streams present at industrial sites. Furthermore, this process can operate at lower temperatures, reducing energy consumption and eliminating the requirement for hazardous supplementary chemicals when compared to amine-based carbon capture methods (Ostovari et al. 2020; Walker et al. 2024).

The main reaction for the carbon capture is a reaction between metal oxides and carbon dioxide towards the formation of carbonates. For the process to be utilized for carbon dioxide storage, the end products need to consist of insoluble and stable carbonates. To date, there are two main ways to carbonate the alkaline flows: direct and indirect carbonation (Walker et al. 2024). Direct carbonation (gas-solid or aqueous) takes place in a single step, wherein the alkaline feedstock reacts directly with CO_2 (Sanna et al. 2014). The indirect pathway occurs in multiple steps: first, reactive components (alkaline metals such as Mg, Ca) are extracted using solvents like acids or bases; then, these extracted components react with CO_2 in either an aqueous or gaseous phase. Compared to the direct approach, the indirect method is more complex and needs supplementary chemicals but generates purer products. The use of supplementary chemicals together with grinding of the feedstock, thermal activation, use of higher pressure and temperatures during carbonation showed increased CO_2 capture efficiency also in the direct route (Sanna et al. 2014). However, the employment of supplementary chemicals could potentially endanger human health and the environment (Shavalieva et al. 2021). The need for energy and supplementary chemicals leads to additional emissions that might outweigh the benefits of CO_2 emissions reduction. Thus, there is a need to evaluate the environmental viability of the carbonation process to ensure that the additional impact caused by the process does not negate the positive effect of the CO_2 capture.

In this work, an environmental and systems-level analysis of direct carbonation and product valorization options for GLD is presented in the form of a life cycle assessment. Additionally, a basic economic analysis is performed to review the potential operating costs of the carbonation process and revenue flows from selling the carbonates and CO_2 emissions rights for the sequestered carbon. The objective of the analyses is to ascertain that the GLD carbonation process results in overall carbon capture and to provide insight for subsequent research and development towards a potential scale-up.

The analyses are conducted within a research context that seeks to identify residual alkaline flows that could be useful for carbon capture. The present LCA study is a first study intended to help experimentalists understand what a future industrial, carbon capture system around GLD could look like. Understanding of where the main environmental impacts could occur are expected to facilitate the prioritization of continued research and development efforts towards reducing or even avoiding such impacts. More specifically, the analyses aim to provide decision-making support during the initial laboratory-scale development of the process (c.f., Leventaki 2023) and insights to the pulp and paper industry regarding the use of GLD for CO_2 emissions mitigation.

2 Materials and methods

To evaluate the feasibility of using GLD as a feedstock for CCUS, a comparison is made between the current practice of landfilling GLD and three GLD carbonation scenarios. The evaluation involves conducting a life cycle assessment (LCA) on an industry-scaled system, along with an economic analysis.

LCA is used to assess the environmental performance of the entire life cycle of the GLD carbonation process, from the production of GLD to its disposal or use. The LCA procedure consists of the following phases: goal and scope definition, inventory analysis, impact assessment, and interpretation of results (Baumann and Tillman 2004). The present LCA study is an example of LCA for emerging technologies and novel products which builds on a framework to scale laboratory data to industrial scale (Piccinno et al. 2016). Here, the scaling considers the optimization effects of lab data on carbonation of GLD provided by the earlier studies of Leventaki (2023) and to create a more realistic future representation of the developed industrial process system, we followed the framework suggested by Piccinno et al. (2016).

Several cases are considered in the comparative assessment. The base case consists of the business-as-usual scenario, where GLD is disposed of via landfilling. For the explored GLD carbonation scenarios, we look at various cases involving different utilization options of the carbonated material. With $CaCO_3$ (same as in limestone) as the main expected compound in carbonated GLD, we look towards the construction industry for utilization cases. These were chosen based on notions of material quality and CO_2 mitigation opportunity. To begin, we wanted to explore an option similar to the base case (landfilling) and two replacements options for long-lived products to ensure carbon storage, one with lower demands on material quality and one in a CO_2-intense application to get an idea of their respective significance for overall CO_2 reductions. These alternatives are best understood as types of utilizations and aimed to guide continued research. A schematic representation of the existing industrial system (business-as-usual), experimental

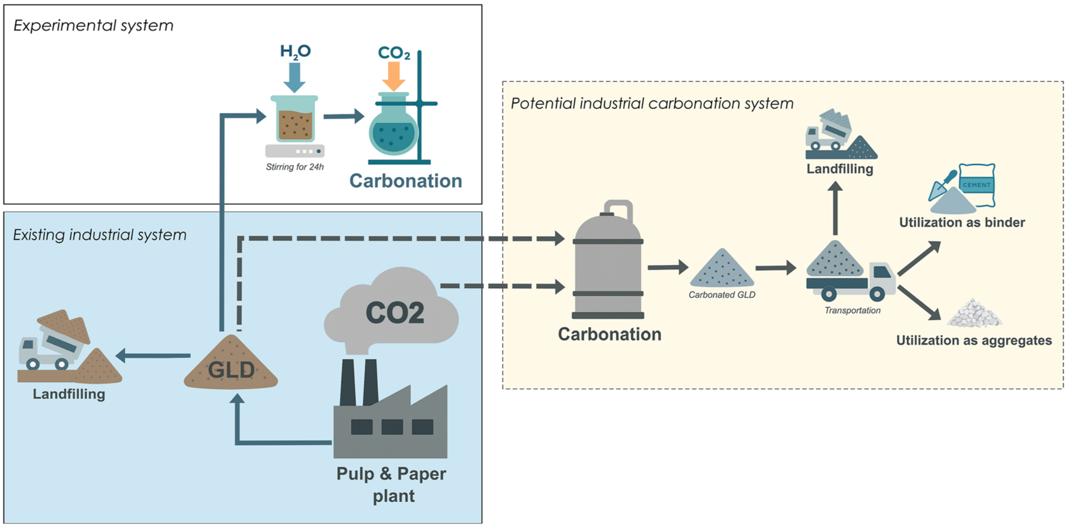

Figure 1: Schematic representation of the existing industrial system (business-as-usual), experimental lab system for obtaining basic reaction data for potential industrial system and potential industrial carbonation system for the explored scenarios (carbonates' utilization as binder and aggregates).

lab system and potential industrial carbonation systems (explored systems) for the considered scenarios are shown in Figure 1.

The first case is a carbonates landfill scenario, an option closest to current GLD management practices of landfilling. The other two cases involve the use of carbonates as construction materials, either as aggregates or as binders. In these cases, the carbonates are assumed to replace natural aggregates for road construction and ordinary Portland cement. The choice of carbonated GLD as fine aggregates in road construction is based in reported trials found in the literature (Quina and Pinheiro 2020; Sundqvist 2021) and patents (e.g., Labart 1995). Regarding cement, it has been found that substituting parts of Portland cement clinker with limestone has beneficial effects on cement hydration and that the utilization of carbonated alkaline wastes presents an opportunity towards sustainability of cements (Jin et al. 2024; Kang et al. 2024).

A point of departure in this study is laboratory data for the carbonation of GLD via the direct aqueous route provided from an earlier study by Leventaki (2023). To evaluate the feasibility of a future carbon capture and storage solution, additional stages of the system like post-processing and carbonates utilization and related uncertainties have been considered. Furthermore, the material and energy flows of the system have been scaled up. To accommodate the uncertainties associated with the scale-up and analyzed scenarios in the utilization cases, best- and worst-case scenarios are modeled. The calculations do not account for the potential atmospheric carbonation of GLD in landfills. However, this aspect is addressed in the Discussion.

The main aim of the basic economic analysis is to understand what the key costs or values related to carbonation of GLD could be as this outlines some boundary conditions for its economic viability. This involves the evaluation of potential operating costs and revenue streams related to the carbonation process (e.g., the sale of carbonates, and the CO_2 emissions rights for carbon sequestration). The analysis places a specific emphasis on the influence of electricity prices on these financial aspects. This is because electricity consumption plays a pivotal role in all stages of the process and constitutes a primary operational expense of the carbonation system. The considerable fluctuations in electricity prices observed in Sweden in recent years could have major implications on the cost-effectiveness of the carbonation system.

2.1 Materials

The analyzed GLD material is provided by a pulp mill located in northern Sweden, producing around one million tonnes pulp annually. The composition of the GLD varies depending on the type of feedstock, the process technology, operating conditions, and the properties of the pulp. Commonly, GLD consists of a diverse array of elements, such as calcium (Ca), sodium (Na), magnesium (Mg), sulphur (S), carbon (C), iron (Fe), manganese (Mn), potassium (K), phosporus (P), silicon (Si), aluminium (Al), and copper (Cu) (Kinnarinen et al. 2018). On average, Sweden produces approximately 12 million tonnes of pulp annually. Approximately 4–20 kg of GLD per tonne of pulp is produced in mills, depending on the size of the mill (Kinnarinen et al. 2018). These pulp mills typically also produce flue gas that can serve as a source of the CO_2 required for the carbonation reaction. The carbonation process leads to the formation of carbonated material that can potentially be utilized in a variety of ways depending on the properties of the final material.

The investigated options for carbonates' utilization are 1) landfilling, and their use as: 2) replacement of natural fine aggregates in road construction; and 3) replacement of a portion of ordinary Portland cement. By replacing part of these materials with carbonates, the pressure on natural resource sources could be reduced (National Academies of Sciences 2019). Currently, the primary source of aggregates consists of mined virgin materials such as sand, gravel, and crushed rock. Another possible utilization alternative is the use of carbonated GLD as a substitute for cement. The production of commonly used ordinary Portland cement has a significant impact on climate change due to its substantial release of CO_2 emissions (Turner and Collins 2013). Therefore, replacing a portion of ordinary Portland cement production with an alternative material might result in significant CO_2 emissions reduction. A study conducted by Batuecas et al. (2021) has shown that replacing 2 % of cement with nano $CaCO_3$ can result in a reduction of over 60 % in CO_2 emissions from cement plants. However, the effectiveness of these substitutions is strongly influenced by the properties of the carbonated GLD, and the performance of the carbonated material may be inferior compared to that of the substituted material (Benhelal et al. 2018; Ostovari et al. 2020). For example, concrete produced of cement with added calcium carbonate showed reduced compressive strength at the mature stage (Ali et al. 2015). Additionally, the substitution of the construction materials by the carbonated residual materials might be hindered by regulations and stakeholders' acceptance (Ramírez 2022).

2.2 LCA

2.2.1 Goal and scope definition

The analysis seeks to address the following question: Does utilizing GLD for carbon capture, storage, and utilization offer environmental benefits? The goal of the LCA is to assess use of GLD for carbon capture, storage, and utilization. The assessment compares landfilling of the GLD (base case) and GLD carbonation with three alternative utilization routes of the carbonated material: (1) CO_2 storage through landfilling of the carbonates; (2) CO_2 storage in carbonates utilized as construction aggregates, and (3) CO_2 storage in carbonates utilized as a cement substitution. The steps of the life cycle that influence the results the most are identified. Additionally, environmental performance related to the use of carbonated GLD as construction materials instead of being sent to landfills is explored.

In LCA, the environmental impact of the studied system is related to a reference unit representing the system's function, the functional unit (Baumann and Tillman 2004). However, the function of an industrial system can be defined differently depending on the needs and point of view of the main decision-maker. Since the current study supports experimental work on alternative utilization of GLD as a carbon capture, storage, and utilization material, the functional unit is selected to be 1 tonne of GLD entering the process.

The carbonation process is primarily focused on CO_2 reduction, consequently, the study's focus is on the impact category of climate change. IPCC 2013 GWP 100a was used as the life cycle impact assessment method. Other environmental aspects like depletion of natural resources and leaching of heavy metals are discussed qualitatively. The geographical scope is defined to be Sweden, with the underlying assumption that the dregs undergo carbonation at the pulp production site.

The modeled systems in the comparison are presented in Figure 2. Construction of the carbonation plant is not included in the calculations; however, the environmental impact of the plant's construction is described under uncertainties of the analysis in the Discussion. In cases when the final carbonated product is utilized as some form of construction material, the system is credited with the avoided environmental load of the averted production of natural aggregates and of cement binder for the construction sector.

For the calculations, experimental data for the GLD carbonation process are scaled up to be representative of a middle-size industrial plant. For scaling up the site-specific data for pulp and paper production plants in Sweden, and a scale-up framework for chemical processes presented by Piccinno et al. (2016) are used. For the background data, e.g., the impact of electricity fuels, and cement production average values are collected from the Ecoinvent 3.8 database (Ecoinvent 2024) and relevant literature.

2.2.2 Inventory analysis

The flowchart of the modeled systems in LCA is presented in Figure 2. The data used in the analysis is explained in the following and summarized in Table 1.

Base case: The current common practice is to landfill GLD. This has provided a base-case scenario which alternative carbonation and utilization scenarios are compared to. In the base case scenario, it is assumed that GLDs are landfilled close to the production site in the best case, and

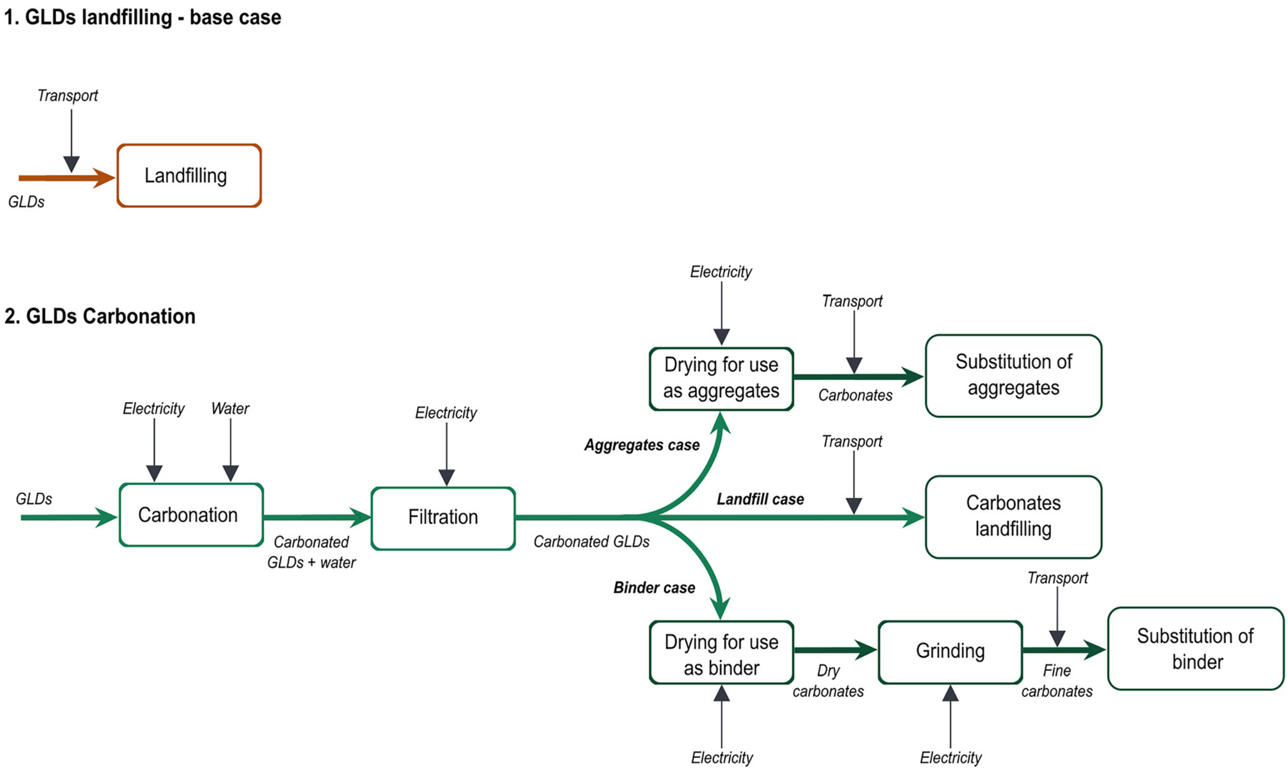

Figure 2: Life cycle of the GLD carbonation system with the compared options: (1) landfilling of GLD – base case; (2) GLD carbonation cases: the carbonates landfilling case; the aggregates case – use of the carbonated material to substitute natural aggregates; and the binder case – use of the carbonated material to substitute cementitious binder in construction.

100 km away in the worst case. The latter is a conservative assumption, reflecting the fact that landfilling might occur further off-site from a pulp mill in some cases (Olivegren 2024).

Carbonation: The inventory data for the GLD carbonation cases relies on both laboratory experiments and modeled data. The carbonation reaction of GLD requires a source of CO_2, alkaline material, and water (Leventaki 2023). The sample of GLD provided by SCA had 45 % total solids. Since GLD is a complex mixture of various minerals, salts and solubles, its composition is often expressed in oxides. Here, as a percentage of the total solids, it was 25 % CaO, 12.5 % MgO, 3.8 % Na_2O and 2.4 % MnO, as provided by the supplier. Thermogravimetric analysis (TGA) was used to estimate the proportion of Ca and Mg present as carbonates in the fresh GLD. The results indicated that most of the Ca (about 87 %) occurred as $CaCO_3$, with the remaining 13 % present as $Ca(OH)_2$. Similarly, around 77 % of the Mg was in the form of $MgCO_3$, leaving about 23 % as $Mg(OH)_2$. Due to the high moisture content, Na was expected to be present as a mixture of NaOH, Na_2CO_3, and $NaHCO_3$. Given that GLD also contain a small amount of Si, it is likely that some of the non-carbonated Na, Ca and Mg could be present in the form of silicates.

The sample was carbonated through an experimental procedure (Leventaki 2023). In this procedure, 5 g of GLD were dissolved in 100 ml of water and stirred for 24 h, consuming 1.29 Wh of energy during the stirring process. Subsequently, a gas mixture containing 30 % (vol) CO_2 and 70 % (vol) N_2, flowing at a rate of 200 ml/min, was sparged through the liquid to facilitate the carbonation process. The carbonation reactor was weighed throughout the experiment and monitored with FTIR and pH probes. Composition of GLD was analyzed before and after carbonation, with X-ray diffraction and scanning electron microscopy (Queiroz et al. 2025). More $CaCO_3$ was found in the carbonated GLD. The carbonation reaction lasted for 24 min, resulting in the capture of 0.177 g of CO_2 per gram of GLD material. Both stages of the experiment were conducted under ambient conditions. For more details on the experiments, see Leventaki (2023) and Queiroz et al. (2025).

Energy requirement for stirring energy was obtained from Leventaki (2023). For the scaling up of this data, Olivegren (2024) calculated the industrial stirring energy estimation using the equation proposed by Piccinno et al. (2016). This equation takes into consideration factors such as the type and diameter of the impeller, the rotational velocity of

Table 1: Key data used in the LCA calculations.

Base case		Unit	Data source
Transport to a landfilling site – best case	1	km	Assumption
Transport to a landfilling site – worst case	100	km	Assumption
Carbonation			
CO_2 captured	0.177	g/g GLD	Lab data (Leventaki 2023)
Gas flow	200.00	ml/min	Lab data (Leventaki 2023)
Electricity for flue gas delivery	8.20	MJ/s	Zhang et al. (2014)
Electricity for stirring	1.29	Wh	Lab data (Leventaki 2023)
Electricity for stirring (axial flow) – best case	4.28	kWh/ tonne material	Scaled-up lab data by Olivegren (2024) using the Piccinno et al. (2016) framework
Electricity for stirring (radial flow)	18.70	kWh/ tonne material	Scaled-up lab data by Olivegren (2024) using the Piccinno et al. (2016) framework
Electricity for stirring – worst case	84.80	kWh/ tonne material	Assuming 20 % of the linear scaling value based on reactor volume
Post-processing			
Electricity for filtration – best case	1.00	kWh/ tonne of dry material	Energy consumption of filters, lower value according to Piccinno et al. (2016) framework
Electricity for filtration – worst-case	10.00	kWh/ tonne of dry material	Energy consumption of filters, upper value according to Piccinno et al. (2016) framework
Utilization			
Transport to a landfilling site – best case	1	km	Assumption
Transport to a landfilling site – worst case	100	km	Assumption
Transport to a construction site	100	km	Assumption
Electricity for drying for usage as aggregates – best case	457.50	kWh/ tonne material	Estimation by Olivegren (2024) based on Piccinno et al. (2016) assuming 5 % moisture in the final product
Impact of natural aggregates, coarse – best case	1.34	kg CO_2e/ tonne of material	Dias et al. (2022)
Impact of natural aggregates, coarse – worst case	32.00	kg CO_2e/ tonne of material	Dias et al. (2022)
Electricity for drying for usage as binder – worst case	503.25	kWh/ tonne material	Estimation by Olivegren (2024) based on Piccinno et al. (2016) assuming 0 % moisture in the final product
Electricity for grinding – best case binder	30.00	kWh/ tonne material	Genç (2016)
Electricity for grinding – worst-case binder	50.80	kWh/ tonne material	Sousa and Bogas (2021)
Electricity usage by an electric truck	1.10	kWh/km	Volvo FH electric, Volvo Trucks (2022)
Mechanical properties of the cementitious binder with carbonates – best-case	100	% of cement	Assumption
Mechanical properties of the cementitious binder with carbonates – worst-case	60	% of cement	Assumption

stirring, the density of the reaction mixture, and the reaction time. For the best-case scenario, an axial flow reactor is assumed. In the worst-case scenario, energy consumption for stirring is modeled as 20 % of the linear scale-up of the energy consumption during the experiments. In reality, mixed axial and radial flows could perhaps be expected (Tajik et al. 2025.), thus, an intermediate value assuming the radial impeller type is also calculated (see Table 1).

Post-processing: For the post-processing stage, filtration of the carbonated material, inventory data is modeled based on literature data and calculations by Olivegren (2024) using the framework by Piccinno et al. (2016). The energy consumption of the filtration step largely depends on the particle size of the carbonated material and can vary between 1 and 10 kWh per tonne of dry material (Piccinno et al. 2016). Filtration of the smaller particles requires more energy, compared to the larger grains. For the best and worst scenarios, the lower and upper ends of the range put forward by Piccinno et al. (2016) are adopted. The average value is also computed for sensitivity analysis as recommended by Piccinno et al. (2016).

Utilization: In the carbonates landfilling case the carbonated GLD (stores CO_2) is transported to a landfilling place, same as in the base case. However, the transportation in this case will show the relative significance of CO_2 from transportations relative to the amount of absorbed CO_2

through carbonation. In case the carbonated material is utilized as a construction material, an extra drying step is modeled after the filtration. It is assumed that following filtration, the carbonated material contains 55 % moisture by mass, similar to the uncarbonated GLD material transported to the landfill. The energy consumed during the removal of the remaining moisture content was calculated as suggested by Piccinno et al. (2016). The assumption is that, in the best-case scenario for the aggregates, the final product contains 5 % moisture. In the worst case, 0 % moisture content is assumed. For the binder case, 0 % moisture is assumed for both scenarios. The distance of 100 km is assumed for the location of the construction site. For all the alternatives, the best-case scenario presumes an electric mode of transportation, while for the worst-case scenario, a diesel-fueled truck is assumed.

In the case of aggregates, the carbonated material substitutes gravel composed of crushed stone from open granite or basalt quarries or limestone. According to the review study performed by Dias et al. (2022), the environmental impact of coarse natural aggregates varies from 1.34 to 32 kg CO_2e per ton of material.

In the case of the binder, the carbonates replace cement whose particle size generally is in the range of 1–50 µm (Schumacher and Juniper 2023). Thus, an additional grinding step of the carbonates with a power consumption of 30 kWh/ton (Genç 2016) in the best case, and 50.8 kWh/ton (Sousa and Bogas 2021) for the worst case is modeled for the binder scenario. In the best-case scenario, carbonates can replace part of the cement, and no degradation of performance is assumed. In the event of reduced performance, the cementitious binder composed of carbonated GLD is assumed to have a performance level of 60 % compared to that of cement, which is considered the worst-case scenario.

2.3 Economic analysis

The economic analysis is made for a preliminary estimation of operational costs and revenues for the carbonation system in comparison to the GLD landfilling option. This is an initial analysis focusing on operational costs and largely excludes capital costs. Key data used for economic analysis are presented in Table 2.

Table 2: Key data used in the economic analysis.

Economic good	Price	Unit	Data source
Electricity – best case	0.02	EUR per kWh	Lowest value based on monthly electricity wholesale price in Sweden from January 2022 to November 2023 (Statista 2023b)
Electricity – intermediate case	0.09	EUR per kWh	Average value based on monthly electricity wholesale price in Sweden from January 2022 to November 2023 (Statista 2023b)
Electricity – worst case	0.23	EUR per kWh	Highest value based on monthly electricity wholesale price in Sweden from January 2022 to November 2023 (Statista 2023b)
Carbonates	10;30;60	EUR per tonne	Conservative assumptions based on European Commission (2017); Indexbox (2024)
Transportation	0.040	EUR per tonne/km	Estimate for 2023 assuming payload of 42 tonnes, based on report on the Swedish road freight sector by French National Road Committee (Comité national routier (CNR) 2021)
EU Carbon permit	87.59	EUR per tonne of CO_2	The average value for daily European Union Emission Trading System (EU-ETS) carbon pricing for the period Jan 2 – Oct 26, 2023 (Statista 2025)
Landfilling tax	0	EUR	Currently, GLD is exempted from the taxation (Skatteverket 2024)

The GLD landfilling scenario only includes the transportation of the GLD. The analysis of the carbonation options includes costs for electricity usage of the carbonation process, transportation costs, and potential revenue generated from the utilization of the carbonated material. In the case of carbonates landfilling, revenue is generated by selling surplus carbon permits, while in the aggregates and binder cases, revenue can potentially be generated from both selling emission rights and carbonates as a product. Transportation costs are estimated based on the 2021 report on the Swedish road freight sector by the French National Road Committee (Comité national routier (CNR), 2021).

While electricity usage plays a pivotal role in all process stages, the price of electricity can vary significantly due to factors such as supply and demand, fuel, and CO_2 prices, the output of renewable energy, and nuclear power (Hellström et al. 2012; Wang et al. 2022). These variations can significantly impact the economic performance of the process.

Consequently, the study explores the potential profitability and costs associated with different scenarios, considering a best, an intermediate, and a worst-case scenario. Regarding prices, the best, intermediate, and worst-case scenarios encompass fluctuations in electricity and carbonates prices (see Table 2). The potential profit is assessed by subtracting the operational costs of the carbonation process from the revenue at various electricity and carbonates selling prices. As for the process data, the same data used in the LCA process are applied for the best- and worst-case scenarios (see Table 1), while average values are utilized for the intermediate scenario.

3 Results and analysis

The results section describes the outcomes of the LCA and economic analysis of GLD carbonation compared to landfilling. The LCA indicates potential for carbon capture, utilization, and storage, particularly in the utilization case. The economic analysis underscores that achieving a viable economic outcome necessitates intermediate to best-case scenarios with lower electricity prices.

3.1 Life cycle assessment

The findings depicted in Figure 3 suggest that opting for carbonation of GLD instead of landfilling could be advantageous, especially if the carbonates can be utilized as construction materials. Depending on the scenario, the environmental benefit is estimated to fall within the range of 142–686 kg CO_2e/tonne of GLD. The substitution of construction materials by the carbonates may lead to additional benefits in the form of a substitution credit: up to 22 kg in the case of aggregates and up to 533 kg in the case of the binder. When carbonates are utilized as aggregates or cement substitutes, CO_2 is not only stored in the carbonates but also CO_2 and other greenhouse gas emissions are avoided following the substitution of natural aggregates and ordinary Portland cement.

The difference in environmental impact between the best and worst cases primarily emerges during the transportation and drying stages of the life cycle, with the latter being particularly significant for utilization scenarios. The environmental impact difference between the best and worst cases is relatively modest in comparison with the potential of emissions reductions due to the CO_2 capture and substitution of conventional products. Nevertheless, it remains important to limit greenhouse gas emissions from the added process steps.

Figure 4 illustrates CO_2e emissions from different stages of the carbonation process. It becomes clear that in the carbonation phase, under the worst-case scenario with the assumed linear scale-up of lab data, the majority of emissions can be attributed to the electricity demand for stirring. In the best-case scenario, using axial or radial flow impellers for stirring leads to significantly lower energy consumption. Energy consumption continues to be the primary contributor to the overall impact, extending into the subsequent post-processing and utilization steps. Notably, the filtration process exhibits a significantly higher impact in the worst-case scenario, where filtration of smaller carbonate particles is assumed. In all utilization cases, carbonates drying emerges as the main contributor to the impact. Lower levels of moisture in the final product result in higher energy consumption compared to values that permit higher moisture content in the carbonates. Transportation impact becomes notably significant under the worst-case scenario when fossil-based transportation modes are utilized, despite employing EURO6 diesel-powered lorries in these calculations. In contrast, Figure 4 shows that the electric mode of transportation in the best-case scenario with a carbon footprint around 60 times lower than that of the diesel lorry.

The assessment indicates that the most effective strategy for improving environmental impact involves utilizing carbonates as replacements for materials with a high carbon footprint, such as cement. However, in practice, substituting cement may prove more challenging due to stricter functional requirements placed on such a material compared to the replacement of aggregates. While replacing aggregates might result in a significantly lower environmental benefit, it could be easier to achieve. Further improvement in the carbon footprint of the process could be attained by adopting alternative energy sources, such as utilizing waste heat for drying carbonates and enhancing capturing yield, for example.

3.2 Economic analysis

The economic analysis suggests that the profitability of the CCUS process is influenced by specific conditions due to the additional process steps involved in various carbonation scenarios (see Figure 5). The profitability of GLD carbonation is largely determined by the electricity price and the amount of electricity utilized in the process. Landfilling of carbonated GLD may become a more attractive choice if electricity (or alternative energy source) prices for drying exceed 0.09 EUR per kWh, and the selling price of carbonates remains below 60 EUR. With lower electricity prices (best case scenario), selling the carbonates as construction material might

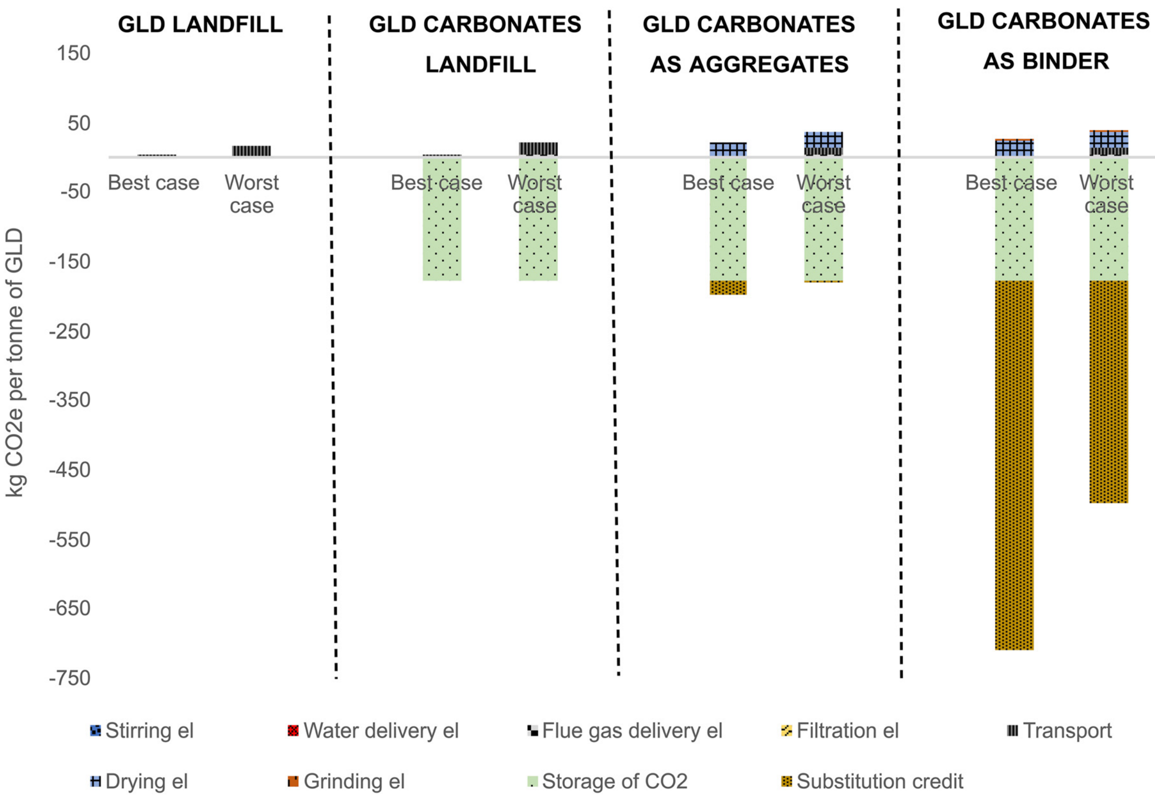

Figure 3: Environmental impact (climate change) of the compared scenarios.

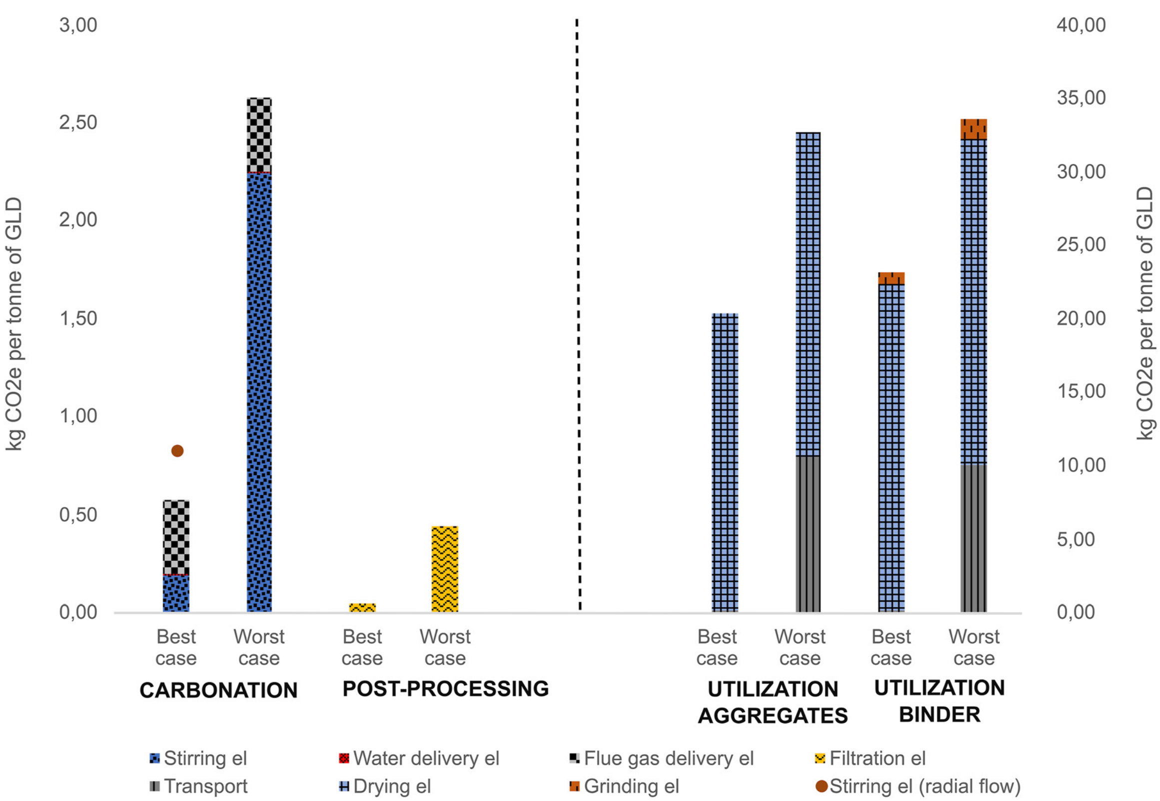

Figure 4: Greenhouse gas emissions in CO_2e from various stages of the carbonation system.

be profitable if the product prices are lower than 30 EUR per tonne of carbonates.

Figure 6 shows a breakdown of the costs associated with various steps of the GLD carbonation scenarios. In the scenario where carbonates are utilized as construction material, the sum of operational costs may surpass 100 EUR per tonne of GLD, primarily driven by the elevated energy demand of the drying process, especially when electricity prices are high, as observed in the worst-case.

Given the considerable costs associated with drying, it becomes important to search for more favorable alternatives, either opting for carbonates that require minimal drying or accomplish drying using an alternative, cost-effective energy source.

The costs of the equipment necessary for constructing the carbonation process have not been factored into the calculations. However, including them in the assessment could significantly reduce or even negate the benefits of the carbonation process. Therefore, to maximize its advantages, retrofitting previously constructed equipment is an option worth exploring.

Since electricity prices fluctuate, the production costs of GLD carbonates varies but these variations are easier to foresee than future selling prices of GLD carbonates since these are greatly affected by the properties and the demand for such carbonated materials. Uncertainties persist regarding its material properties and the anticipated demand for it, but the market for such materials is expected to grow (Rousseau et al. 2024). Currently, material standards, availability, and costs of production hinder the utilization of alternative cementitious materials (Rintala et al. 2021).

While GLD is currently exempt from landfill tax in Sweden (Skatteverket 2024), this may change in the future. Rising costs related to landfilling taxes may drive further development of alternatives for utilizing GLD and GLD-derived-carbonates. For the present analysis, data on operational landfilling costs was not available, but adding these to the tax-related landfill costs would likely further drive the exploration of utilization cases.

The effects of also considering the carbon credits obtained through the substitution of the cementitious material sold on the voluntary carbon market (VCM) could enhance the economic feasibility of the analyzed cases. Nevertheless, the potential effects remain uncertain. The price of the VCM credits is influenced by various factors, including the nature of the project, its scale, geographical location, associated co-benefits, and the year in which they were generated (Carbon Credits 2025; Streck 2021). At the time of writing this paper, the voluntary carbon offsetting market is small and not centralized, but the increasing need to reduce the greenhouse gas emissions of companies and individuals could drive the growth of the VCM significantly (Streck 2021).

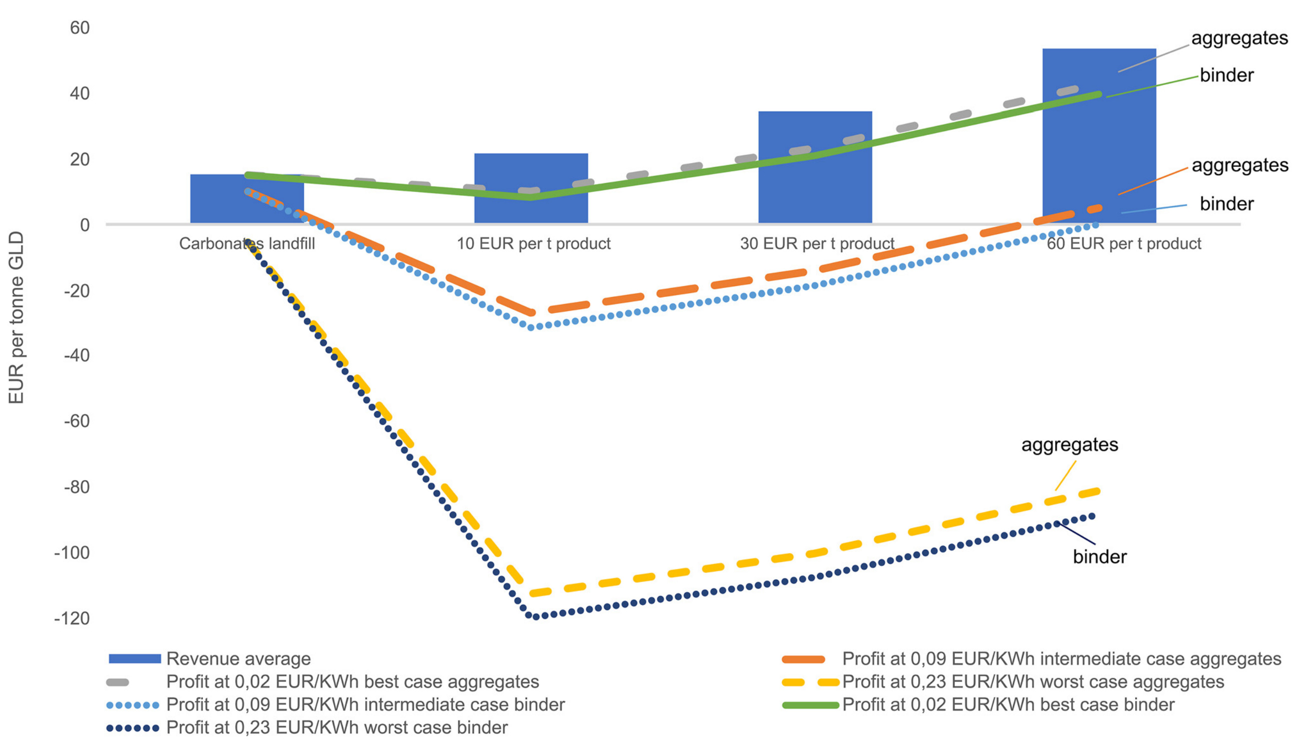

Figure 5: Revenue and profit flows of the carbonation process at different electricity and carbonates prices.

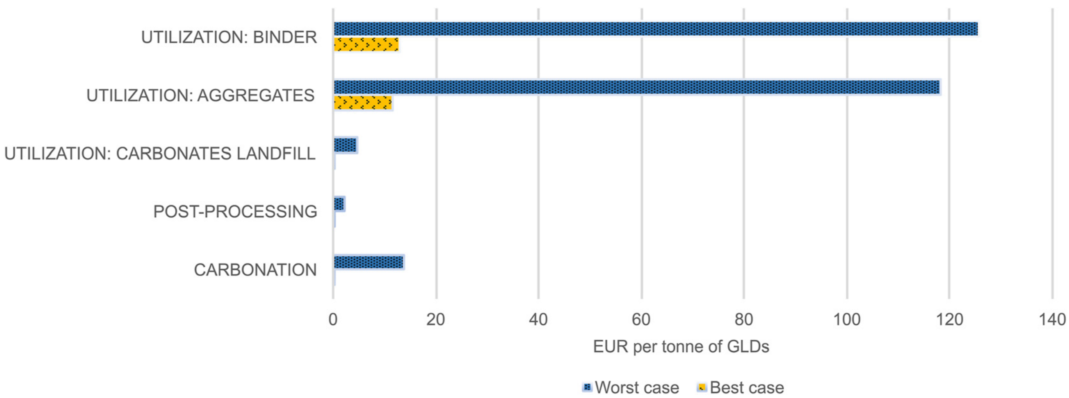

Figure 6: Comparison of the production costs of different steps of the GLD carbonation scenario.

4 Discussion

The current study is the first analysis of the use of GLD for carbon capture, storage, and utilization. Previous research into GLD utilization options included the application of GLD as partial clinker substitution, filler in geopolymers, material for road pavements, landfilling cover and sealing layer in mines, neutralizing agent for the treatment of acidic wastewater, mine waste, soil amendment, liming material (Quina and Pinheiro 2020) and pretreatment agent for lignocellulose (Sewsynker-Sukai et al. 2020). While there exists a variety of research on the utilization of GLD for diverse purposes, the option of CCUS has not been previously evaluated.

The current study demonstrates that using GLD for carbon storage and utilization could provide environmental benefits compared to the current practice of landfilling GLD. Sweden annually produces approximately 300,000 tonnes of GLD (Alakangas et al. 2014). The GLD carbonation and utilization of the carbonated product could help mitigate around 0.1–0.5 % of the country's carbon dioxide emissions (Statista 2023a) and reduce the amount of landfilling material, thus contributing to the UN SDGs on climate change and sustainable consumption and production. The most significant reduction of greenhouse gases occurs when GLD-derived carbonates can replace CO_2-intensive products, such as cement. However, achieving such a replacement may not always be environmentally and/or economically feasible.

4.1 Limitations and uncertainties

The findings reveal uncertainties due to the restricted availability of data across various system steps. The lab data utilized is specific to GLD from a particular site. While this can suffice to evaluate if pursuing CCUS using GLD can be worthwhile, the results should not be interpreted as definite. Since the composition of GLD, and consequently, the carbonation efficiency, may differ for different GLD samples and pulp mills. Assessing GLD samples from various pulp mills and establishing a carbon capture range for the material would be beneficial.

At present, GLD is classified as waste rather than a by-product, and, as per ISO 14044, no environmental impacts are currently assigned to it. However, this status may evolve in the future with the increased utilization of GLD across various applications, potentially leading to its reclassification as a by-product. The calculations employed an impact factor for the Swedish electricity mix, but the carbonation system may operate on renewable energy. Additional waste management might be required for the flows of the post-processing step if the filtered water is not suitable for recycling. No flue gas pretreatment is expected before the carbonation reaction; however, experimental work might be required to assess the behavior of the system when the industrial flue gas is supplied to the carbonator. Although there are data uncertainties, there are also improvement possibilities including energy sources for transportation and drying.

A more important factor that could determine the net carbon balance is related to the extent of atmospheric carbonation of landfilled GLD. The high moisture content of GLD may induce self-carbonation under atmospheric conditions. Laboratory samples demonstrated the capture of 0.115 g CO_2/g of GLD within one year of storage compared to 0.177 g CO_2/g of GLD in experiments. This suggests that carbonation of GLD could also occur during landfilling, albeit at a much slower rate and only in superficial layers (Jin et al. 2024). However, while forced carbonation of GLD accelerates the uptake of CO_2, it also holds the potential to impact the mechanical and chemical properties of the resulting carbonated material, which could be beneficial for its utilization. The reduction of pH and immobilization of heavy metals would take place before the material is landfilled or utilized for other purposes.

Research addressing the substitution of traditional fossil-based products in construction reveals that substitutability depends not only on the technical properties of the new materials but is also influenced by factors such as price, acceptance, regulations of building materials and codes, and knowledge among stakeholders (Arehart et al. 2021; Howard et al. 2021). Thus, other utilization cases (e.g., in geopolymers) could be of interest for further research than those analyzed here. According to Zink et al. (2016), the complete displacement of conventional products necessitates special conditions and that it is likely that low or even zero displacements could occur. In such a scenario, a carbonation-derived product can be introduced to the market without displacing any of the existing fossil-based products. However, solid carbonates are already used in construction, mainly in the production of cement and concrete (Jin et al. 2024). Furthermore, the massive need for construction materials in the future – global demand is estimated to exceed 60 billion tonnes by 2030 (Da and Le Billon 2022; Statista 2024) – might facilitate their use in the construction sector. The substitution of cement and natural aggregates with carbonates would significantly alleviate the pressure on natural resources and ecosystems.

4.2 Recommendations for further analysis

The analysis identified potential methods to enhance the environmental performance of the carbonation system, which require practical testing and evaluation. One such method has been tested with promising results: Queiroz et al. (2025) replaced the fresh water used in the experiments by Leventaki (2023) with alkaline wastewater from the pulp and paper industry which led to enhanced CO_2 absorption. Nevertheless, further research is essential to evaluate other ways to enhance the carbon capture and storage efficiency of the entire process, with minimal or no additional environmental impact. This may entail investigating access to low/no-cost surplus energy at pulp mills and other alkaline material flows within other industries towards utilizing the carbonation process. Surplus energy could be examined for enhancing, for instance, carbonation efficiency and drying the carbonated material. Further improving the process of leaching alkaline metals and filtering leached metals before the carbonation reaction (indirect carbonation) could result in a higher-purity material with a broader range of potential applications. Assessment of the attributes of the carbonated material could unveil potential applications and determine the feasibility of utilizing carbonates for specific purposes. Additionally, comprehending the maximum theoretical capture rate of CO_2 by the GLD under various conditions is crucial for evaluating the benefits of the process and optimizing the efficiency of GLD carbonation.

Strong alkalinity, presence of heavy metals and water, and limited knowledge of the long-term stability, technical performance, and environmental impact of GLD are possible reasons for a lack of industrial-scale applications of the material (Quina and Pinheiro 2020) beyond landfilling. Carbonation of the GLD could provide a better solution to some of these problems and thereby offer a viable alternative towards GLD utilization. For this, improved understanding of the composition of the carbonates is needed as this would help identify suitable utilization cases.

5 Conclusions

The current study represents the initial assessment of the environmental impacts of waste GLD for carbon capture, storage, and utilization. Also, a basic economic analysis has been conducted. Together the results suggest that the carbonation of GLD demonstrates the potential for carbon capture, utilization, and storage. However, both the environmental impact and the costs of the process are affected by the method of utilization of carbonated GLD. Substituting carbonates for materials such as cement, which emit a considerable amount of CO_2 emissions during production, can yield significant environmental benefits. From an economic perspective, the substitution option requiring extensive drying of the carbonates may only be economically viable if low-cost energy sources are readily available. Otherwise, alternatives such as CO_2 capture followed by landfilling or utilization as a product with minimal drying requirements might be more favorable.

Despite the limitations arising from the scarcity of available data and the need for scale-up estimations this study provides valuable insights that offers directions for future research and into some environmental considerations in the case of developing an industrial-scale GLD carbonation system. Future research that concentrates on improving the CO_2 capture yield and assessing the mechanical and chemical properties of the GLD-derived carbonates would contribute to a deeper understanding of the feasibility of the process at an industrial scale.

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