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Life Cycle Assessment of Waste Glass Geopolymerization for the Production of Sustainable Construction Materials

Georgios Manthos [1], Dimitris Zagklis [2], Christos Georgopoulos [3], Constantina Zafiri [4] and Michael Kornaros [5,*]

[1] Department of Environmental and Resource Engineering, Quantitative Sustainability Assessment Section, Technical University of Denmark, Bygningstorvet, Building 115, DK-2800 Kongens Lyngby, Denmark; gema@dtu.dk
[2] Department of Industrial Engineering and Management, School of Engineering, International Hellenic University (IHU), 57400 Thessaloniki, Greece; zagklis@ihu.gr
[3] Department of R & D, Enalos Research and Development PC, 51 Metamorfoseos Str., 15234 Chalandri, Greece; georgopoulos@enalos.com
[4] Green Technologies Ltd., 5 Ellinos Stratiotou Str., 26223 Patras, Greece; nzafeiri@tee.gr
[5] Laboratory of Biochemical Engineering & Environmental Technology (LBEET), Department of Chemical Engineering, University of Patras, 1 Karatheodori Str., University Campus, 26504 Patras, Greece
* Correspondence: kornaros@chemeng.upatras.gr

Abstract: Replacing conventional materials with new recycled materials is one of the goals of sustainable development, as it promotes the creation of environmentally friendly products while reducing the amount of waste to be treated. A common recyclable waste stream associated with urban living is waste glass, which typically comes from packaging or product containers. Although most of this stream can be reused and/or recycled, it is worth exploring alternative uses, especially for areas with high fluctuations in waste glass production. An example would be the sudden increase in waste glass in tourist areas during the high season. To this end, the present work presents the results from the life cycle assessment of waste glass geopolymerization for the production of cement tiles. The methodology includes the estimation of mass and energy balances by dividing the whole process into several sub-processes (NaOH addition, energy consumption, etc.). The NaOH addition was found to be the most burden-intensive process, with a total damage of 9×10^{-5} DALY per ton of waste glass in the human health category, while a minor contribution in all damage categories was attributed to process electricity demands (7.7 to 19.4%). By comparing the geopolymerization process with conventional recycling, an environmental benefit of 20 mPt and 26 kg CO_2 per ton of waste glass was demonstrated, indicating the process's expediency. The present study is a valuable tool for the up-scaling of processes towards a circular economy.

Keywords: geopolymerization; waste glass; life cycle assessment; recycling; construction materials

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1. Introduction

In the last decades, there has been a growing need for a transition to more sustainable systems regarding the three different aspects of sustainability: economic feasibility, environmental sustainability, and societal expectations [1,2]. The first goal is related to the economic challenges between a proposed process/product and its environment (market instabilities, investor decisions, etc.). The environmental sustainability goal can be achieved by minimizing the environmental footprint of a process/product compared to the existing

regime. Social sustainability is the most overlooked part of a sustainability assessment, due to the lack of a clear methodology [3]. However, as the methodology matures and expands, it is necessary to include it when possible.

Waste glass constitutes a substantial portion of urban solid waste due to its large production volumes. Although this type of waste is easily recyclable, it tends to accumulate in landfills and recycling facilities [4]. The most common form of glass in the municipal waste stream is food and beverage containers. Specifically, in the European Union, about 19 million tons were generated in 2015, and the recyclable fraction of this amount reached a percentage of 86% [5]. Several factors can be crucial for glass reuse, such as glass dimensions and its concentration of heavy metals [6]. Also, the color of broken and mixed waste glass can affect the performance of the recycling process and the properties of the new material [7]. In most municipalities in Greece, recyclable packaging is collected as a separate stream and transported to sorting and recycling centers. Seasonal variations, especially in tourist areas during high tourist seasons, can result in quantities of glass exceeding the capacity of such recycling facilities.

Geopolymerization is a promising technology that facilitates the recycling of hazardous wastes (such as slag, dust, etc.), with the goal of creating new products [8]. The process mechanism involves the dissolution of any pozzolanic compound in an alkaline solution to initiate the formation of an aluminum, oxygen, and silica structure [9]. The underlined process is characterized by low energy requirements, low carbon footprint values, and low installed equipment costs. One of the main factors influencing geopolymerization is the nature of raw material. Calcined materials (such as waste glass) have shown faster dissolution and improved mechanical properties for this type of treatment [10]. Regarding the economic feasibility of geopolymerization, the addition of NaOH has been reported as the main factor affecting the sustainability of the process [11]. At the same time, the energy cost of geopolymer brick production is found to be much lower than that of conventional brick production, as the conventional process requires temperatures in the range of 1100 °C to 1400 °C [12].

Life cycle assessment (LCA) is a method that measures the resources consumed and emissions produced throughout the life cycle of a material or process and evaluates its environmental impact. This type of analysis is a tool increasingly considered necessary for research and policy-making purposes by institutions and companies. The core concept of this assessment is to connect the environmental impact of a process to the materials and energy required for product creation [13]. In addition, this analysis can be used to identify the stages of the process with the highest environmental impact in order to achieve further targeted optimization at these specified process stages [14].

The objective of this study was to evaluate the environmental impact of the geopolymerization of waste glass for the production of sustainable building materials, namely pavement tiles, and to compare this process with the recycling of waste glass. The general methodology followed was the estimation of mass and energy balances according to experimental data and the literature, and the implementation of these results in the LCA software (SimaPro 8). Several assumptions were made regarding the system boundaries and the yields of the sub-processes. The geopolymer production process has been studied in the literature with respect to its environmental impact [15,16]. In the work of Petrillo et al. [17], the production of tiles using geopolymerization showed the environmental benefits of the process. Additionally, the work of Salas et al. [18] addressed the up-scaling of geopolymer concrete production from the laboratory to industrial scale. The analysis showed that the NaOH addition was the step with the highest environmental impact. Although geopolymerization has been studied in the existing literature, data on a direct comparison between waste glass recycling and this technique are lacking. Moreover, this

assessment offers insights into the geopolymerization technique, identifying the process's hotspots and providing direction to material engineers and researchers toward a sustainable society's transition. The present study aims to highlight the use of environmentally friendly techniques for new product formation using solid waste streams in synergy with established municipal recycling processes.

2. Materials and Methods

2.1. Goal and Scope

The mandatory steps to be followed for a life cycle assessment of a process are as follows: (1) goal and scope definition, (2) life cycle inventory analysis, (3) life cycle impact assessment (LCIA), and (4) life cycle interpretation [19]. The goal of the analysis was not to completely replace conventional recycling technology, but to propose an alternative sustainable solution for the residual waste that can be processed in recycling facilities. Thus, a functional unit (FU) of 1000 kg of waste glass was used as the basis for the analysis; so, all results presented here refer to the treatment of this amount of waste. This functional unit was selected considering that approximately 2.7 tons of waste per capita are generated annually and a percentage of 60% of this waste is recycled (1.7 ton per capita) [20].

The life cycle inventory (LCI) data were derived using SimaPro 8 software [21] and the Ecoinvent 3.5 database [22]. Background processes covering the whole region of Europe was selected in order to assure a wide applicability of the assessment. The environmental impact of the processes was calculated using the IMPACT 2002+ v2.15 method, which groups the LCI data into 15 midpoint categories, which are then aggregated into 4 damage categories, as shown in Figure 1. The graph shows that elementary flow results with comparable impact pathways are combined into midpoint impact categories. The differentiation between midpoint and endpoint categories reflects the distribution of impacts at an intermediate stage between inventory results and environmental quality changes [23]. All midpoint categories use units as reference compounds that can be converted to the units of the damage categories.

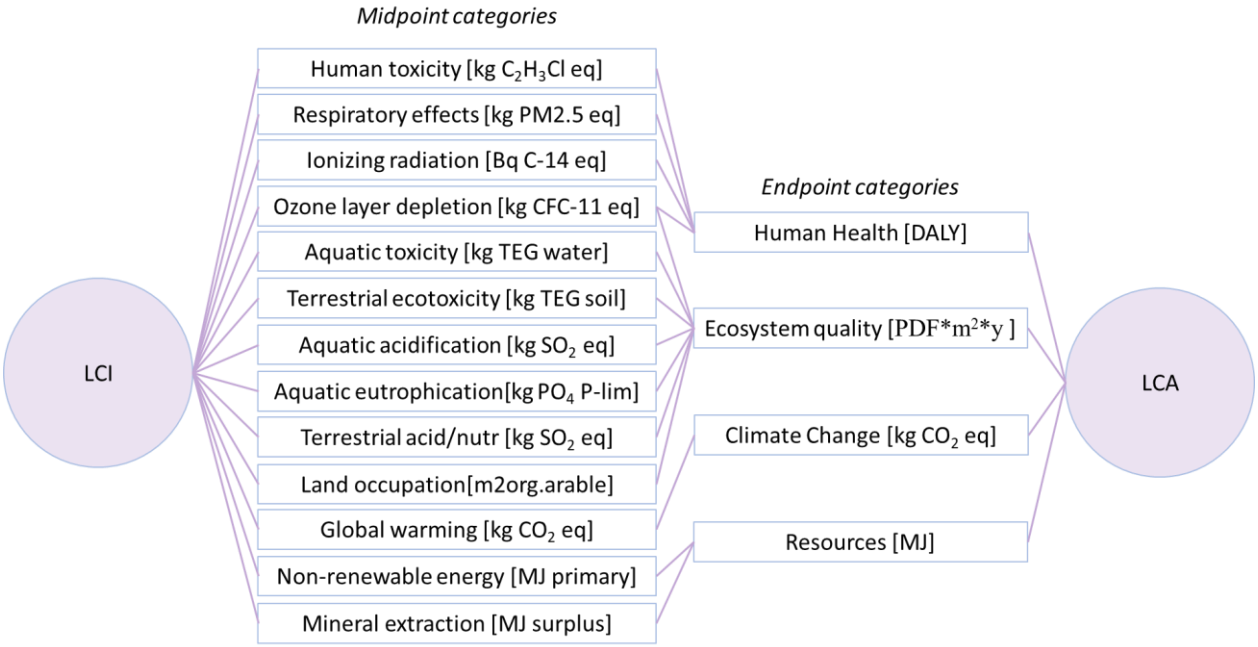


Figure 1. Scheme of the IMPACT 2002+ based on the work of Humbert et al. [24].

An LCA can stop at the LCI generation step. However, grouping the LCI results into impact categories is usually necessary to facilitate the understanding of the results, their

interpretation, and comparison with the literature data. The midpoint damage categories aggregate the LCI results into categories that are not easily understood, such as land occupation or water extraction, but with fewer assumptions. On the other hand, endpoint damage categories are easier to understand, with damage categories such as climate change and resources used, but with more assumptions in aggregating the results. Several studies present results only in midpoint or endpoint damage categories. In this study, the midpoint damage categorization was first performed using the results from the LCI, and then a second grouping into endpoint categories was performed, allowing results to be presented at both levels.

For the final normalization of the endpoint categories, the exposure of an average European citizen (Pt) during one year was used. The normalization values from the IM-PACT 2002+ method were 0.0071 DALY/Pt, 13,700 PDF*m^2*y/Pt, 152,000 MJ/Pt, and 9950 kg CO_2/Pt.

2.2. Mass Balances and Assumptions

The process stages included in the analysis were divided into those directly affected by the process design (foreground) and those indirectly affected through mass and energy balances (background). The processes included in the analysis for both the geopolymerization method and conventional recycling up to the crushed glass production stage are shown in Figure 2. More specifically, the geopolymerization process includes the collection of waste glass, its crushing, the addition of reagents and mixing, a curing stage, and finally, the production of tiles. The modeling of the recycling process was based on data from the work of Blengini et al. [25] and includes the transport of waste glass, the washing stage using oxygen to avoid anaerobic conditions, the drying and grinding stage, and the production of crushed glass. In the case of geopolymerization, the final product displaces the production of concrete tiles, while conventional recycling displaces the production of silica sand [25]. The construction materials of the process equipment are not included in the analysis, which is quite common in such analyses, because the environmental impact of constructing the equipment is considered minimal compared to the impact of its operation, due to the equipment's long lifespan [14].

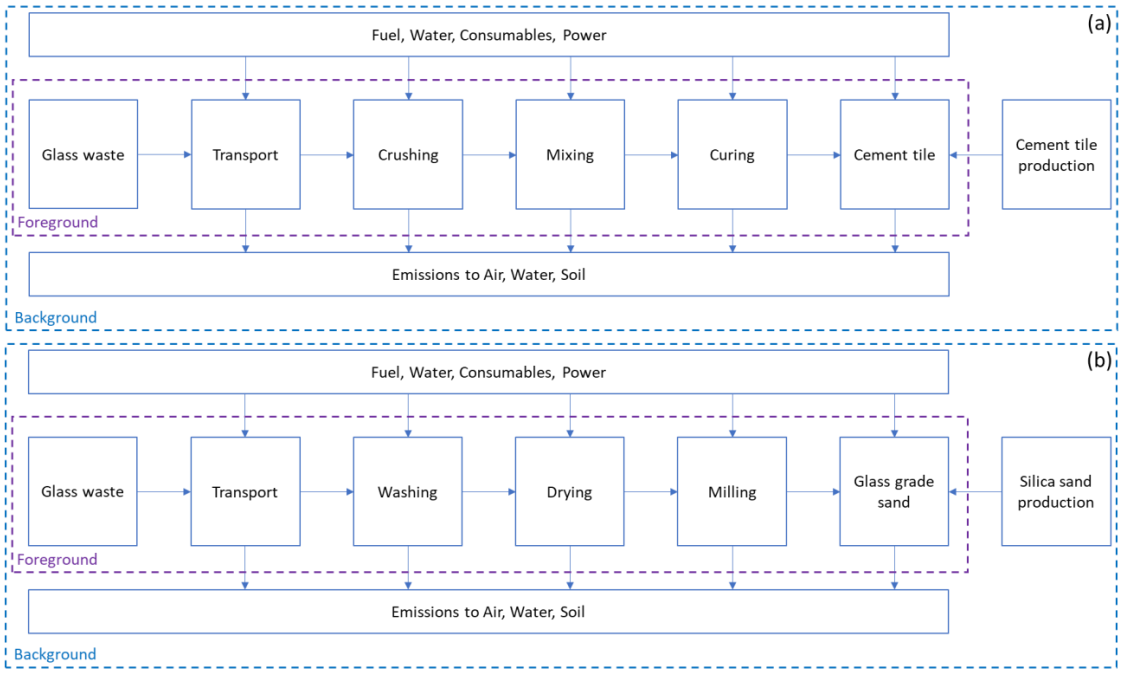


Figure 2. Process stages for the different treatment methods of waste glass ((**a**) geopolymerization, (**b**) recycling).

2.3. Geopolymerization Process Modeling

The geopolymerization process was modeled according to the data presented in Table 1. The processed product displaces the production of cement tiles, while the raw materials used in the process are sodium hydroxide and water. Since the scope of the present assessment is the environmental study of a small plant capable of processing seasonal waste glass that cannot be recycled, it was assumed that the plant would be located within the geographic boundaries of a city; therefore, a small transport distance of 5 km was assumed. This was the only transport burden considered in this analysis, as the handling of material during the processing and transport of NaOH to the pilot geopolymerization unit are expected to have a minimal impact on the overall process. It was also assumed that the cement tiles produced would be used to pave the local sidewalks. Typically, the impact of transporting raw materials and products is a small part of the total impact (shown in the following analysis). In this analysis, a total transportation impact of 5 tkm (ton-kilometers) was assumed.

Table 1. Input data for LCA of the geopolymerization process of waste glass.

Avoided products		
Cement tile	1068	kg
Materials/fuels		
Sodium hydroxide	56	kg
Tap water	200	kg
Transport, freight, lorry	5	tkm
Electricity/heat		
Electricity, medium voltage	87.5	MJ

The Municipality of Megara, Greece has a separate glass collection system utilizing bell-shaped containers. Part of this stream was supplied for pilot operation and obtaining the data used in this study. No separate glass types were investigated as the provided waste glass stream was a mixture of all the available glass containers.

The input data for the geopolymerization process were based on experimental data obtained during the operation of a pilot geopolymerization unit (0.5 t/h capacity) in the Municipality of Megara, Greece, for the treatment of excess waste glass during the high tourist season.

The product of the geopolymerization process was modeled by displacing cement tiles. This was based on the literature data that highlight the physical properties of this product, comparable to conventional construction materials [26,27].

Background processes were modeled using the Ecoinvent 3.5 database. More specifically, data representing Europe, excluding Switzerland, were utilized. If European data were unavailable, global data were used instead.

2.4. Recycling Process Modeling

Although the geopolymerization process will not be used in competition with the recycling process, as it is recommended for periods when the quantities of waste glass exceed the capacity of the recycling plant, the process is presented here for comparison purposes. The analysis of this process is based on the literature [25]. The input data for SimaPro are shown in Table 2. The process was studied up to the stage of crushed glass production, which displaces silica sand production. Several raw materials were included in the analysis in order to perform the necessary washing (water and oxygen to avoid the

growth of anaerobic microorganisms), drying of the material (thermal energy), and finally, its crushing (electric energy).

Table 2. Input data for LCA of the recycling process of waste glass.

Avoided products		
Silica sand	850	kg
Materials/fuels		
Oxygen, liquid	0.49	kg
Tap water	830	kg
Transport, freight, lorry	20	tkm
Diesel	0.425	kg
Electricity/heat		
Heat, district or industrial, natural gas	103.59	MJ
Electricity, medium voltage	25	kWh

3. Results and Discussion

3.1. Characterization

The mass and energy balances of the processes were aligned with raw materials extracted from the environment and emissions of pollutants/compounds released into the environment, using SimaPro 8.0 and the Ecoinvent 3.5 database to compile the life cycle inventory. The IMPACT 2002+ v2.15 methodology was used to aggregate the LCI into 15 midpoint impact categories. To identify the hot spot in the process, the analysis was performed by dividing it into five different sub-processes. The NaOH sub-process included the amount of NaOH required for the production of cement tiles, while the avoided product (displacement of cement tiles) was included in the product sub-process. To assess the results of various impact categories, a percentage-based analysis was conducted by dividing the value of each impact category by the damage in the scenario with the highest absolute value for that category. The normalization values are provided in Table 3, while the comparative results are presented in Figure 3.

Table 3. Normalization values for midpoint damage categories.

Damage Category	Unit	Maximum Absolute Value
Mineral extraction	MJ surplus	0.20
Non-renewable energy	MJ primary	14×10^2
Global warming	kg CO_2 eq	27
Aquatic eutrophication	kg PO_4 P-lim	13×10^{-3}
Aquatic acidification	kg SO_2 eq	0.7
Land occupation	m^2 org.arable	1.4
Terrestrial acid/nutr	kg SO_2 eq	3.3
Terrestrial ecotoxicity	kg TEG soil	36×10^2
Aquatic ecotoxicity	kg TEG water	11×10^2
Respiratory organics	kg C_2H_4 eq	3.2×10^{-2}
Ozone layer depletion	kg CFC-11 eq	4.5×10^{-5}
Ionizing radiation	Bq C-14 eq	10×10^2
Respiratory inorganics	kg $PM_{2.5}$ eq	16×10^{-2}
Non-carcinogens	kg C_2H_3Cl eq	3.0
Carcinogens	kg C_2H_3Cl eq	1.3

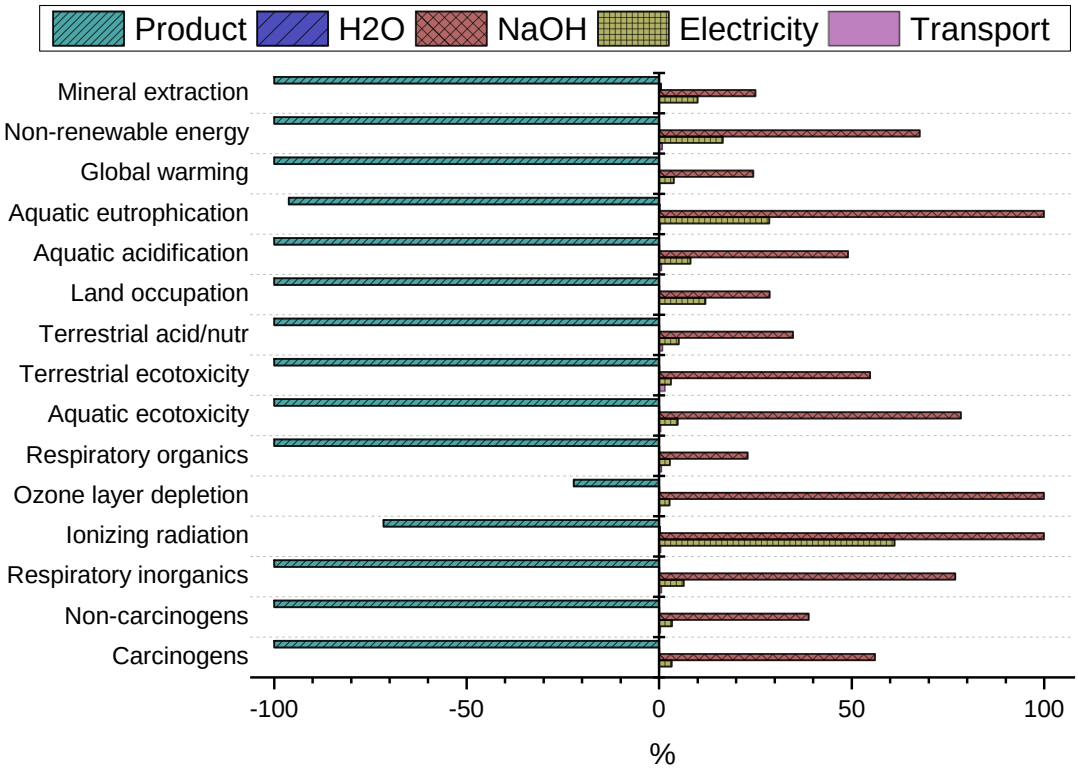


Figure 3. Comparative results per 1000 kg of waste glass for the different parts of the geopolymerization process in the midpoint damage categories.

In all of the midpoint impact categories, the process product had the highest absolute value, and in all cases, it was expressed as a negative harm (or environmental benefit). A negative impact value is strongly connected with the reduction in emissions to the ecosphere implementing the proposed methodology, avoiding the production of a conventional material [28]. In these midpoint categories, the addition of NaOH had the second highest absolute value, always expressed as damage (environmental impact). The environmental burden of NaOH addition outweighed the environmental benefit of the final product in three midpoint categories, namely aquatic eutrophication, ozone layer depletion, and ionizing radiation. The significance of these three midpoint categories will be clarified after their comparison and aggregation with the other relevant categories at the endpoint level. The third most significant stage of the process was the consumption of electric energy, especially in the ionizing radiation category. Conversely, water consumption and transportation had a negligible impact on the overall results.

3.2. Damage Assessment

The results of the LCI midpoint category aggregation were further aggregated into four different endpoint categories to provide a comprehensive interpretation of the results. The endpoint category results for the different sub-processes are shown in Table 4.

Table 4. Aggregated damage assessment results per 1000 kg of waste glass for the different sub-processes of geopolymerization.

Damage Category	Unit	Transport	Electricity	NaOH	H_2O	Product	Total
Human health	DALY	7.08×10^{-7}	7.52×10^{-6}	8.97×10^{-5}	3.67×10^{-8}	-1.2×10^{-4}	-2.2×10^{-5}
Ecosystem quality	PDF*m^2*yr	0.5	1.3	17.7	0.01	−34	−14.5
Climate change	kg CO_2 eq	0.7	10	65	0.1	−265	−189
Resources	MJ primary	11	235	962	1	−1421	−212

The aggregation of damages into endpoint categories reiterates the significant contributions of the end product and NaOH addition to the overall impact of the process. At this level, the environmental benefits of the product outweigh the impact of NaOH addition in all four endpoint damage categories. Electricity consumption is again the third largest contributor, while transportation and water consumption have very small contributions to the overall results.

3.3. Normalization

A final normalization step was performed to aggregate the results in terms of the environmental impact of an average European citizen. The results of this step for the different sub-processes are shown in Figure 4. The addition of NaOH was identified as the most environmentally damaging process, as it has the highest impact in all categories of endpoint impact. On the other hand, the prevention of cement tile production was the process with the most positive impact (negative damage). Specifically, in the case of the human health damage category, the negative damage of the avoided product outweighs the positive damage of the chemical addition in the process.

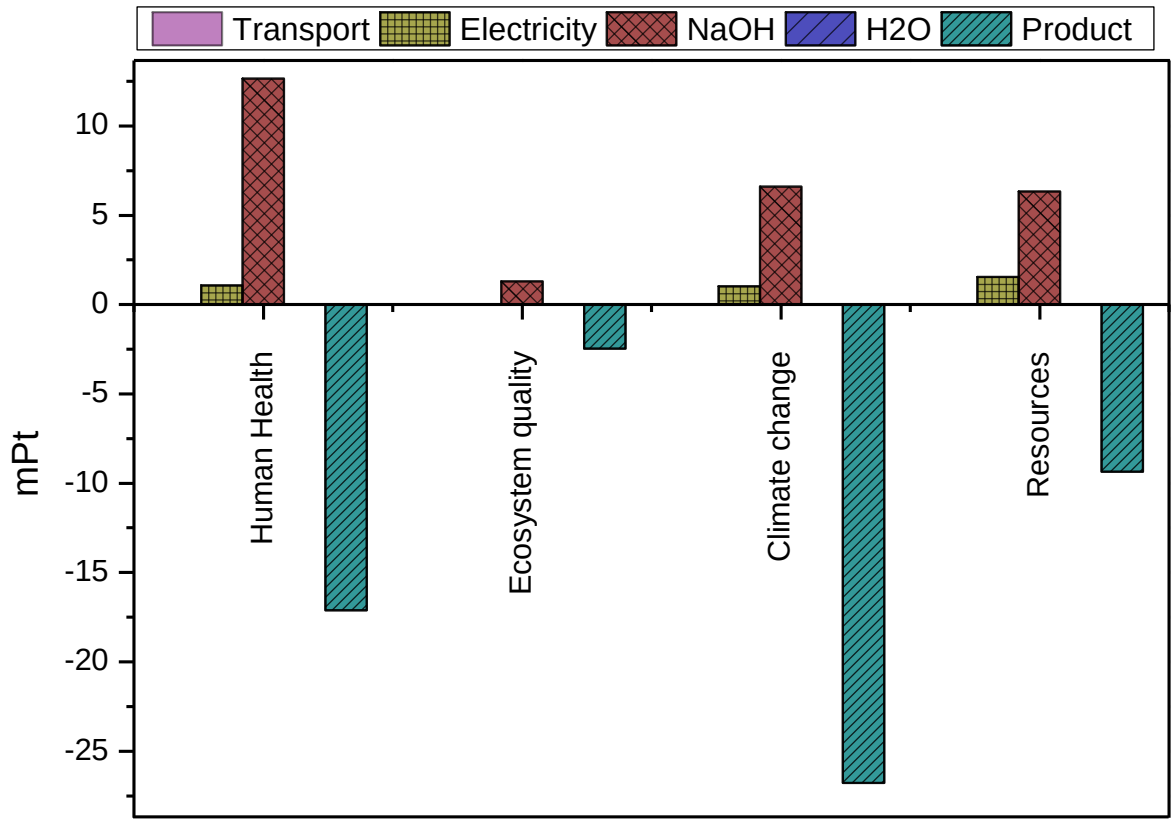


Figure 4. Normalized damage per 1000 kg of waste glass for the different parts of the geopolymerization process in the endpoint damage categories.

The proposed process was compared to the baseline scenario process of glass recycling (Figure 5). The geopolymerization process shows similar impacts to the recycling process in the human health and ecosystem quality damage categories, but there is a significant advantage of the geopolymerization process (about seven times less impact compared to the recycling process) in the climate change category. This difference between the two processes in the climate change category is due to the displacement of the production of cement tiles, as their production is very energy intensive. For recycling, the low impact in the Resource Damage category is related to the abundance of silica sand in the ecosphere, the

displacement of which has little impact on the process. Also, the energy used to dry the raw material after washing may contribute to the increased impact in this impact category.

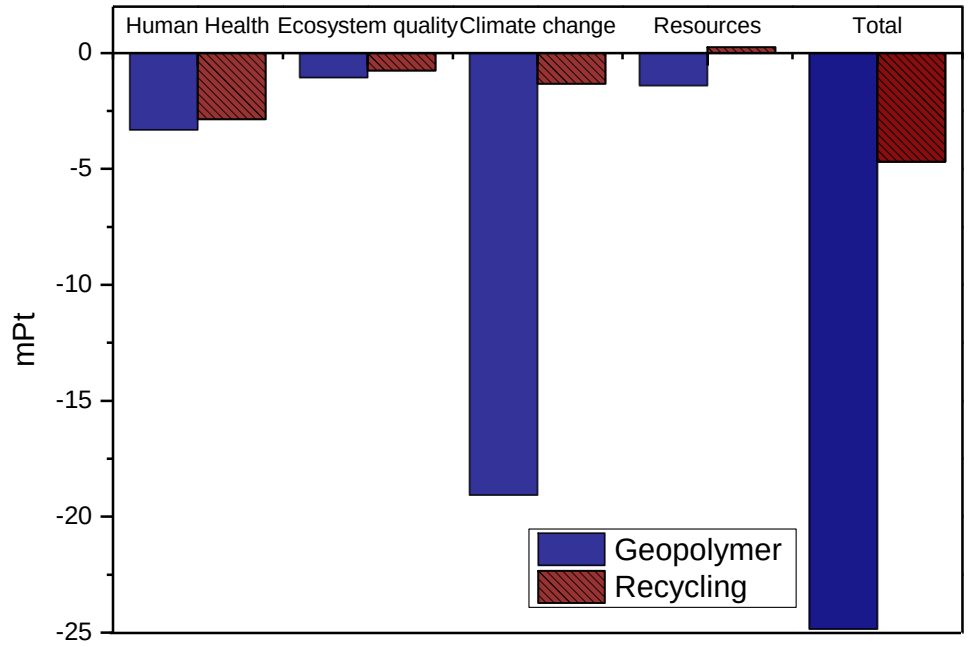


Figure 5. Comparison of geopolymerization and recycling process per 1000 kg of waste glass for the endpoint categories.

4. Interpretation

According to the evaluation of the results of the midpoint categories, the negative damage caused by the prevention of cement tile production proved to be satisfactory, since it can compensate for the environmental impact of the remaining process steps. High impact values were observed for the NaOH addition, which is consistent with similar studies in the existing literature [18].

The NaOH production process consists of several sub-processes (well drilling, brine extraction, etc.) that consume significant amounts of energy and fuel. The efficiency in energy and fuel consumption has been reported as a crucial parameter affecting the environmental impact of NaOH [29]. Therefore, NaOH production using a renewable energy mix can further reduce the geopolymerization environmental burden in the climate change impact category. Another alternative for impact reduction during the alkaline treatment is the use of combinations of different activators. Sodium silicate and fly ash and slag can be used in different concentrations alongside NaOH and KOH in order to optimize the process' environmental impact and product's characteristics [30]. According to the Ecoinvent database, sodium silicate (Na_2SiO_3) has a lower environmental impact across all categories compared to sodium hydroxide (NaOH) (5.27×10^{-6} DALY kg^{-1} Na_2SiO_3 instead of 1.41×10^{-5} DALY kg^{-1} NaOH and 4.05 PDF m^2 year kg^{-1} Na_2SiO_3 instead of 23.5 PDF m^2 year kg^{-1} NaOH). Thus, any admixture of this component to the alkali treatment can lead to less impacts in alkaline treatment.

A significant burden was attributed to the electricity needed for the geopolymerization process, while the water needed, and the transportation of waste glass do not significantly affect the process. This impact can also be reduced by using electricity from renewable energy sources. The use of this type of energy could decarbonize up to 5% of the total process impact in the climate change category [31]. To reduce the impact of the NaOH addition, several authors suggest replacing conventional NaOH with NaOH produced from solar salt. This can lead to an 18% reduction in the global warming category [32]. In

another study by Mir et al. [33], the impact assessment of recycled ceramic tile and recycled brick waste-based geopolymers was addressed. The larger impacts in this process were also observed by the NaOH addition and electricity consumption (about 60% of the total impact). The same observation has been made by other authors in their analysis of the environmental impacts of alkali-activated concrete produced with a silicate activator derived from waste glass [34]. By converting the midpoint categories to endpoint damage categories, the three impact categories that show environmental impact (aquatic eutrophication, ozone layer depletion, and ionizing radiation) do not affect the overall endpoint result because the environmental damage avoided by the product outweighs the damage of these categories. In all four endpoint categories, the benefit of the product outweighs the burden of the rest of the process. The endpoint categories were reported as the final step in linking the LCA to the Sustainable Development Goals (SDGs) of Good Health and Well-being, Clean Water and Sanitation, Affordable and Clean Energy, Sustainable Cities, Climate Action, and Life on Land and Water [35].

Regarding the comparison of the geopolymerization process with the conventional recycling process, the impacts had similar values regarding the endpoint categories of ecosystem quality, human health, and resources. However, the geopolymerization process showed a significantly reduced impact in the category of climate change. This can be attributed to the avoided products of each process. Conventional cement tiles have a higher production impact than silica sand (avoided product of the recycling process) because the raw material (cement) has several environmental impacts from its production. It is clear that the geopolymerization process offers environmental benefits compared to conventional cement tile production. In the work of Samuel et al. [36], a sustainability comparison was made between geopolymerization and lime treatment, considering the environmental, economic, and social index. The geopolymerization process showed a reduced sustainability index of 10% compared to lime treatment, so there are other alternatives worth pursuing and investigating. Nevertheless, the geopolymerization of waste glass for the production of cement tiles during the periods of excessive waste glass generation seems to be a promising alternative to recycling. Moreover, feedstock material (solid precursors) can be a crucial factor for efficient geopolymerization in terms of raw materials consumption and energy accumulation. While Si-rich precursors require more energy but less chemical treatment compared to clayey materials (like kaolin) in the geopolymerization process, this study reveals that chemical treatment remains a significant impact factor for Si-rich materials [37]. Despite this, Si-rich materials like volcanic ash, and Si-rich fly ash are still suitable for geopolymerization due to their overall efficiency and reduced chemical dependency.

Summarizing the normalized results of the endpoint damage categories, the geopolymerization process shows improved results in the overall score (25 mPt avoided per ton of waste glass) compared to conventional glass recycling (4.7 mPt avoided per ton of waste glass). In addition, geopolymer materials can immobilize heavy metals. The safe consolidation of all these materials in a geopolymer matrix is far more preferable to landfilling, as the degradation of landfill cells can pose risks to human health, the surrounding soil, air quality, groundwater sources, and surface water [38].

The economic potential has been reported in the literature for the geopolymerization process for different materials. An increase in financial gain of 5% has been exhibited for the geopolymerization bricks compared to conventional fire bricks for a case study in the region of France [12]. In the same study, the lower production cost was estimated at EUR 114 t^{-1}. A major contribution in the total product cost can be attributed to the high market price of NaOH (EUR 490 t^{-1}); thus, the use of different alkaline precursors may lead to further cost reduction, enhancing the process's sustainability [39].

The proposed process presents a comprehensive strategy for waste valorization for the production of recyclable materials in the context of the circular economy and zero-waste politics. Thus, its implementation aligns with and contributes to the relevant SDGs of the 2030 Agenda [40]. By converting this waste into construction material, the process implementation can reduce the waste amount that ends up in landfills, which is aligned with the SDG 11, 'Sustainable Cities and Communities'. Geopolymerization also supports the recycling and reuse of materials, lowering the carbon footprint associated with conventional cement tile production, and contributing to SDG 12, 'Responsible Consumption and Production' and SDG 13, 'Climate Action'. Finally, an indirect connection can be identified with SDG 9, 'Industry, Innovation, and Infrastructure', since this process can foster the development of resilient infrastructure and alternatives in industrial practices.

5. Conclusions

The environmental impact of the waste glass geopolymerization process was evaluated through the application of the life cycle analysis methodology, which revealed a net positive impact. The primary factor contributing to this observation was the displacement of other materials with high environmental costs by the product of the process. The deployment of the geopolymerization process was validated as a supplementary process for seasonal fluctuations in the quantity of glass collected by a municipality. This method of waste treatment has the potential to further reduce the overall impact of waste remediation in comparison to conventional recycling. It is important to note that the geopolymerization process is not intended to compete with recycling. Rather, it is designed to address the accumulation of materials in sorting centers or end up in landfills. The proposed process offers a valuable alternative for reducing landfill use and advancing the sustainability goals set forth by the European Union for the transition to a sustainable and "green" future.

Author Contributions: Conceptualization, M.K., D.Z., and C.G.; methodology, G.M. and D.Z.; software, D.Z.; validation, G.M., D.Z., and C.G.; formal analysis, G.M. and D.Z.; investigation, G.M. and D.Z.; resources, M.K. and C.Z.; data curation, G.M. and D.Z.; writing—original draft preparation, G.M. and D.Z.; writing—review and editing, C.Z., C.G., and M.K.; visualization, G.M. and D.Z.; supervision, C.Z. and M.K.; project administration, C.G. and M.K.; funding acquisition, C.Z. and M.K. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: Data can be provided on request.

Conflicts of Interest: Author Constantina Zafiri was employed by the company Green Technologies Ltd. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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