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Life-cycle analysis of geopolymers

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Abstract: Geopolymers as alternative binder systems are attracting increasing interest in research and development. They can display outstanding technical properties, such as high strength, high acid resistance, and/or high temperature resistance.

It has been documented by several investigations that good performance of geopolymers can also be obtained by the utilisation of secondary raw materials (industrial wastes like fly ash or slag). This explains the strong interest in this technology from countries with growing industrialisation. These countries accumulate large quantities of industrial wastes and do not have a developed recycling pathway. The use of waste for geopolymer production could not only solve a waste problem, but also reduce the consumption of primary raw materials.

Nevertheless, only limited knowledge exists about the ecological implications of geopolymer production. In this chapter, the effects of geopolymer production will be investigated from an environmental point of view. Furthermore, the predominant drivers of ecological impacts of geopolymers will be discussed to provide guidance for the development of geopolymer compositions for different applications.

Key words: life cycle assessment, geopolymer, concrete, cement, waste.

10.1 Introduction

According to the publications of Glukhovsky, geopolymers or inorganic polymer binder systems had already emerged in the late 1960s (Glukhovsky, 1959). In Russia and the Eastern part of Europe, some industrial applications of geopolymers (e.g., railway sleepers) were reported (Petrova *et al.*, 2005), but it seems that geopolymers were never in a state of mass production. Until today, only niche applications (e.g., fire-resistant glue, exhaust fume pipes, heat/fire barriers) have been established in Western Europe.

Owing to increased environmental concerns as well as increased costs of raw materials, however, research and development of geopolymers is currently experiencing a renaissance. Upcoming industrial nations, in particular China and India, are searching for a recycling option for increasing amounts of industrial wastes to prevent large-scale dumping as well as to help resolve cement shortages.

Geopolymer systems are suitable, in principle, for the utilisation of industrial wastes (e.g., hard/soft coal fly ash, blast furnace slag, red mud) as a secondary resource. Hence, upcoming industrial nations are keenly interested in the development of geopolymers for mass applications.

Although production of applicable geopolymers on the basis of secondary resources has been demonstrated on laboratory and semi-industrial scales, very little knowledge exists about the ecological impact of geopolymer production, especially in comparison to competitive systems.

On a quantitative level, some estimations or rough calculations of single environmental aspects (without clear system boundaries) can be found in literature. Most of them refer to CO_2 emissions only and compare geopolymer (concrete) production with that of Portland cement (concrete). Estimated potential reductions of CO_2 emissions by geopolymer systems vary from a moderate 20% up to an ambitious 80% (Duxson *et al.*, 2007c; Komnitsas and Zaharaki, 2007; Nowak, 2008). But the assessment of new materials on the basis of CO_2 emissions alone is not sufficient. Other emissions and the consumption of resources should be taken into account to gain a more detailed impression.

The most common approach to assessing potential environmental impacts is the method of life cycle assessment (LCA). LCA is a cross-media approach and allows for a more holistic investigation of products and services. The goal of this study is to provide guidance to geopolymer designers for the development of ecologically more advantageous geopolymers.

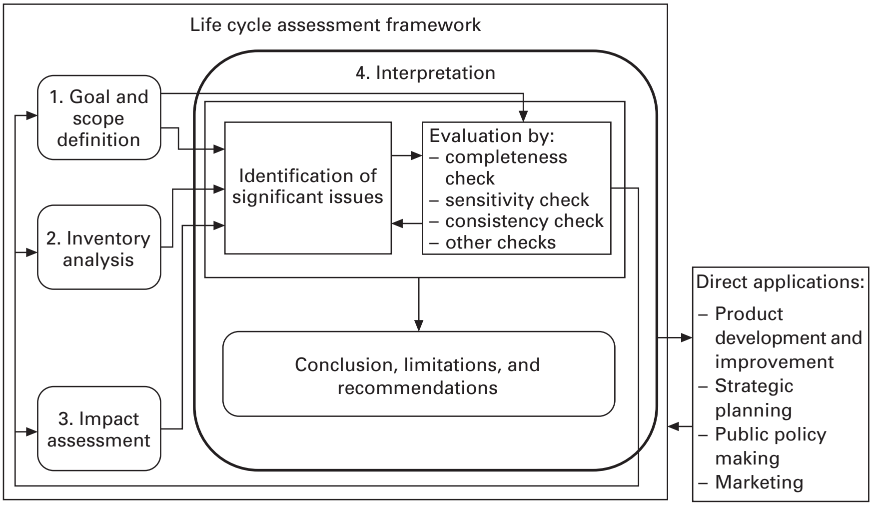
10.2 Life cycle assessment

10.2.1 Methodology of life cycle assessment

The production phase, the utilisation phase as well as the after-use phase (recycling, disposal) of every product is associated with the consumption of resources and emissions into air, water, and earth. To understand the complexity of the product, life cycle analysis methods as well as a quality-assured database are necessary.

Life cycle assessment (LCA) is an established method to evaluate the potential ecological impact of materials, products, or technologies. In LCA, an inventory of all material and energy flows of all processes relevant to an item is taken over the entire life cycle ('cradle to grave') to assess its environmental effects. The LCA methodology is described in the international ISO standards 14040 ff (ISO 14040, 2006). These standards represent a valid framework and permit consistent approaches.

A complete LCA generally comprises four major steps (Fig. 10.1):



10.1 LCA framework according to ISO Standard 14040 series (ISO 14040, 2006).

Goal and scope

The ‘goal and scope’ step defines the objective and intended application of the investigation. Life cycle assessment of a product will never be exhaustive. As a consequence, the ‘goal and scope’ determine the necessary degree of precision and correctness of a study and which part of the life cycle is modelled (e.g., cradle to gate, cradle to grave, or gate to gate). Closely related to the goal of the study is the determination of the functional unit which explicitly defines the functionality of a considered system. This is an important step, because careful definition of a functional unit prevents apples from being compared with oranges. The LCA results always refer to the functional unit.

Inventory analysis (LCI)

Inventory analysis (LCI) indicates the relevant inputs and outputs of a product or service system. On the input side, all energy and raw material consumption are considered, for instance, whereas products and emissions into air, water, and land are counted on the output side. LCI results represent an important information source for the LCA interpretation step.

Impact assessment (LCIA)

Impact assessment evaluates potential environmental impacts associated with the environmental inputs and outputs identified by the LCI.

By applying different models of environmental mechanisms (e.g., global warming due to the emission of greenhouse gases), the inventory (LCI) is translated into potential environmental impacts. For the 'translation', different methods are available with various strengths and shortcomings, (Dreyer *et al.*, 2003). A selection of often used impact categories (and indicators) is presented below:

- Depletion of resources (abiotic depletion potential – ADP)
- Climate change (global warming potential – GWP)
- Acidification (acidification potential – AP)
- Eutrophication (eutrophication potential – EP)
- Human toxicity (human toxicity potential – HTP)
- Ozone depletion (ozone depletion potential – ODP).

Interpretation

The final interpretation step identifies significant environmental impacts (e.g., energy use, greenhouse gases) as well as significant unit processes in the system. Both LCI and LCIA results are considered to identify the environmental drivers. In addition, this final step includes a consistency check, a sensitivity analysis, and a completeness check. Conclusions are drawn and recommendations are made, taking into account the assumptions, limits, and original goal and scope of the study.

10.2.2 Database

In the present study, LCIs were accomplished based on data from different sources. The most frequently used basic data for raw materials, auxiliary materials, and semi-finished products were taken from the Ecoinvent database (Ecoinvent, 2006). Ecoinvent provides scientifically sound, transparent, and quality-assured data sets. Nevertheless, suitability of a specific inventory of the database for a planned investigation remains to be proven in each case. For some inventories of specific raw materials (e.g., metakaolin), new and product-specific data sets had to be provided. In case of metakaolin, basic data (e.g., energy consumption) were obtained from a metakaolin producer/seller in Europe[1]. In Table 10.1, all materials and processes utilised are displayed together with the data source and data quality. The estimated quality of the applied data ranks from very good to adequate. Adequate data quality means that there are some restrictions (e.g., in the form of data gaps, or different regional contexts), but the data are still acceptable for the investigation carried out.

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[1] BASF (formerly Engelhard) declare quite low CO_2 emissions for metakaolin production, cited in Duxson *et al.* (2007c), which cannot be confirmed by our data.

Table 10.1 Utilised inventories with data source and estimated data quality

Process	Data source	Data quality
silicate solution	ecoinvent, literature	very good
NaOH solution	ecoinvent	good
blast furnace slag	industry, literature	adequate (particle emissions into air from grinding are not included)
metakaolin	industry	good (particle emissions into air are not included)
hard/soft coal fly ash	estimation, literature	adequate
Portland cement	ecoinvent	good
gravel/sand	ecoinvent	good
water	ecoinvent	good
HDPE	ecoinvent	good (human and ecotoxicity assessment is severely limited)
auxiliary materials	ecoinvent	good
mixing	industry	adequate (particle emissions into air are not included)
plastic forming	ecoinvent	good
transportation lorry/ship	ecoinvent	very good

10.2.3 Impact assessment method

For this study, the CML method is applied (Guinée *et al.*, 2001) and the cumulative energy demand (CED, [MJ]) is also considered. The CML method comprises several environmental sectors with the related indicators. Two important environmental indicators are analysed:

- Global warming potential (GWP, [kg CO_2 equivalent])
- Abiotic resource depletion potential (ADP, [kg antimony equivalent]).

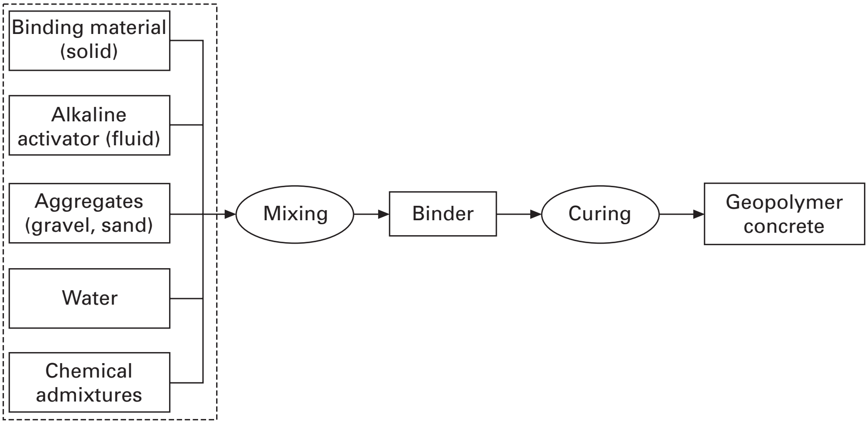
It would also be interesting to analyse the indicator of human toxicity, but unfortunately the database does not allow for the calculation of potential impacts. Basic data for particle emissions (dust), which have a great impact on human health, are lacking in particular for the mineral processing processes (e.g., grinding of slag).

10.3 Influence of the geopolymer composition on environmental impacts

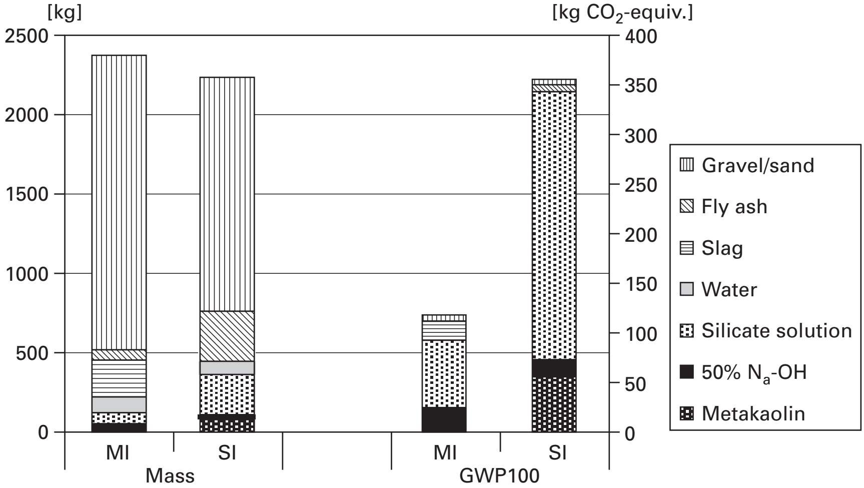
Geopolymers can be produced from a broad range of solid and fluid raw materials of significantly different qualities. Apart from the processing conditions, the raw materials selected are the important parameters, which determine the setting behaviour, the workability, and the chemical as well as the physical properties of geopolymeric products (Duxson *et al.*, 2007a). Furthermore, the environmental profiles depend largely on the raw materials used. There are significant differences between resource-intensive primary solid raw materials (e.g., metakaolin) and less resource-intensive secondary solid raw materials (e.g., fly ash) and between resource-intensive primary fluid raw materials (e.g., NaOH solution, silicate solution) and less resource-intensive primary fluid raw materials (e.g., water). The system boundaries for a comparison of different geopolymer (raw material) compositions are demonstrated in Fig. 10.2, whereby transportation processes are not being included. A comparison of the ratio of raw material mass (Fig. 10.3, left) to the share of environmental impacts represented by the indicator GWP 100 (Fig. 10.3, right) of two different geopolymer compositions reveals the following important aspects:

- Despite their high mass proportion, sand and gravel contribute only a little to the GWP.
- Fly ash (hard coal fly ash or soft coal fly ash) does not significantly contribute to the GWP.
- Slag (only a composite in mixture MI, Fig. 10.3) contributes noticeably to the GWP.
- The provision of water does not noticeably contribute to the GWP.
- Silicate solution contributes significantly to the GWP and dominates the environmental profile of both mixtures.
- Moderate use of NaOH solution (50%) in both mixtures causes a significant contribution to the GWP.
- Moderate usage of metakaolin (only in mixtures SI) contributes significantly to the GWP.

Some general conclusions can be drawn for geopolymer mixtures, which do not consider the influence of composition changes on the technical performance of the final geopolymers. As far as possible, the application



10.2 System boundaries for the comparison of different geopolymer compositions.



10.3 Comparison of mass balances and GWP results of two different geopolymer compositions (MI, SI).

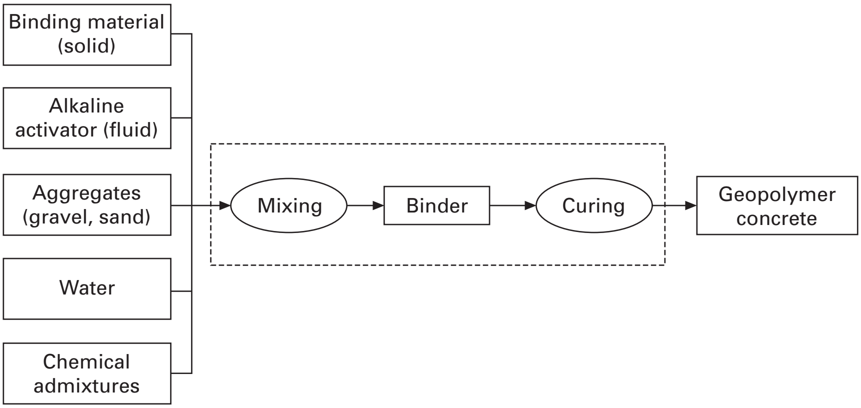
of silicate solution and NaOH solution should be reduced to a minimum, or these materials should be replaced by a more environmentally friendly activator. This is also true for metakaolin which should be replaced by alternatives in order to reduce the environmental load. It has to be stated that the applied (commonly available) quality of metakaolin is very high (in terms of purity). It should therefore be checked whether a lower quality would be more adequate for a broad application in geopolymer systems. Utilisation of secondary raw materials, such as fly ashes (no grinding and no thermal activation necessary) or blast furnace slag from iron production (only grinding necessary), is more favourable, provided that these secondary resources (wastes) are available and environmental limit values are not exceeded.

10.4 Influence of the geopolymer production process on environmental impacts

The geopolymer production process (Fig. 10.4) can be divided into the following major production steps:

- mixing of components
- heat curing.

An additional compaction process (by using a vibration table) during casting is not considered, but its contribution to the environmental impacts is known to be insignificant in other cases (common use). This is also true



10.4 System boundary for the identification of relevant production processes.

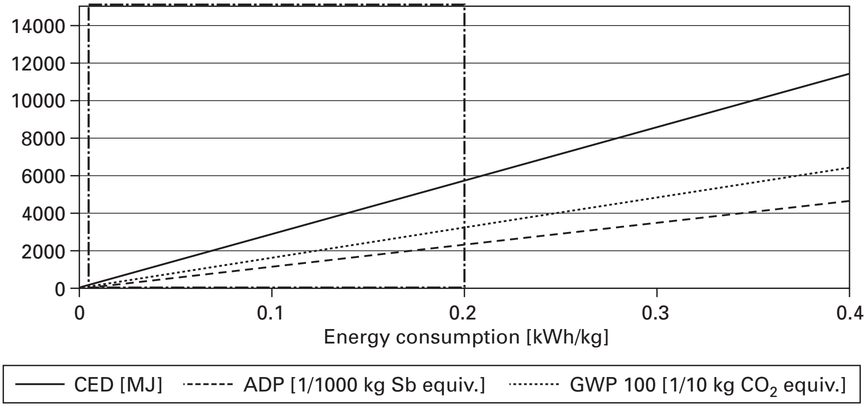
for the mixing process, which contributes less than 1% to the environmental effects (geopolymer production).

In contrast to this, the heat curing process may change the environmental profile of geopolymers significantly. It must be noted that not every geopolymer composition needs the heat curing step. Especially slag-rich geopolymer compositions achieve the desired technical properties within a few hours or days at room temperature without any heat curing (Duxson *et al.*, 2007b; Bakharev *et al.*, 1999).

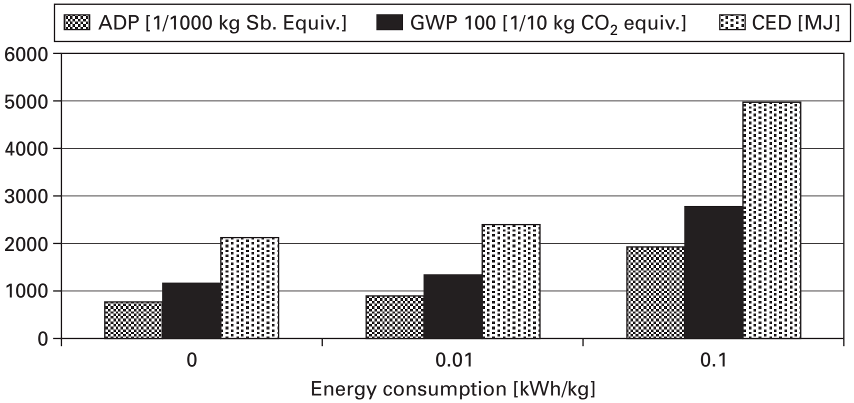
Mixtures containing predominantly fly ash (or other slowly reacting raw materials) require a heat treatment to enhance the maturity of the geopolymers. The moderate temperature ranges usually between 20°C and 80°C. Heat curing within a similar temperature range is also quite common in the pre-cast concrete industry, where the strength development of the concrete elements is accelerated.

The energy consumption for concrete production of common pre-cast companies ranges between 20 and 500 kWh/m^3 (Menzel, 1991), or approximately 0.01–0.2 kWh/kg. For the assumption that an electric conditioning cabinet (<100 kW) is used, the effects of energy consumption on the environmental indicators (CED, GWP, ADP) are highlighted in Fig. 10.5. A simple linear relationship between the energy consumption and the environmental indicators becomes evident. In addition, the normal range of energy consumption for heat curing is indicated (dashed line, Fig. 10.5).

The effect of heat curing on the environmental impacts of geopolymer production is demonstrated in Fig. 10.6. In this specific case, all indicator values are more than doubled, if heat curing consumes a moderate amount of energy (0.1 kWh/kg). If a gas fan burner is used instead of an electric conditioning cabinet, the increase of the environmental indicator values



10.5 Effects of heat curing (consumption of electric energy) on the environmental impact indicators.



10.6 Effect of heat curing on the environmental impacts of geopolymer production.

would be less dramatic, but still significant. From a process perspective, the application of heat curing should therefore be reduced to a minimum. Another option to reduce the environmental impact may exist in the use of waste heat from other processes.

10.5 LCA comparison of geopolymers to other product systems

The understanding of the interdependence of composition, processing, technical performance and ecological/economic effects is essential to develop competitive geopolymers for different application fields (Weil *et al.*, 2006; Dombrowski *et al.*, 2008). Research tends to focus on the improvement of one or two properties to produce so-called high-performance materials. In fact, the market needs a product of suitable technical performance (broad requirement profile, minimum properties are specified in guidelines and/or standards) at moderate costs (competitive to existing technologies) that preferably have low environmental impacts. A comparative LCA, i.e. a comparison with existing traditional systems, already during the development phase of geopolymers is very helpful to identify optimisation potentials or issue warnings from an environmental perspective. As geopolymers have not yet reached product character in many application fields, the comparison with traditional systems is subject to some restrictions. Whereas long-term experience (e.g., durability) exists for traditional systems, information on geopolymers are often limited (Gruskovnjak *et al.*, 2006).

Nevertheless, the utilisation period of materials (including maintenance) has a significant influence on the total environmental performance. To deal with this problem, comprehensible assumptions have to be applied or the results obtained have to be discussed in the light of the known restrictions.

Focusing on two quite different applications, an approach towards an adequate comparison will be presented.

10.5.1 Geopolymer versus cement concrete

In industrialised countries, cement may be considered a cheap product suitable for concrete mass production. To compete with such an optimised system, cheap raw materials, such as blast furnace slag and coal fly ash, may be used for geopolymer production.

The use of secondary raw materials also reduces the environmental impact of the geopolymer system. Nevertheless, the question arises of whether the geopolymer system is competitive to a cement-based system from an environmental point of view. For comparison, a concrete complying with increased requirements is considered (functional unit): freeze-thaw-resistant concrete of exposition class XF2 and XF4 according to DIN EN 206-1/DIN 1045-2. The system boundaries are shown in Fig. 10.7.

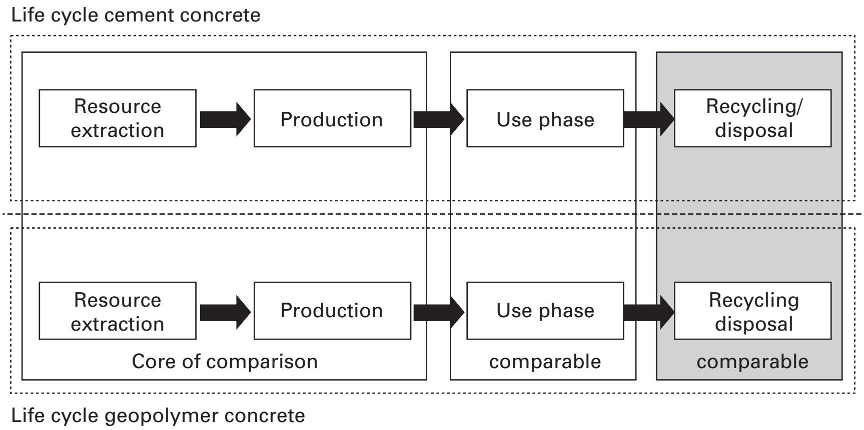
After use, geopolymer concrete and cement concrete might be crushed and used for different applications in civil engineering. Thus there is no evidence of geopolymer concrete having major ecological advantages or drawbacks in comparison to cement concrete as far as the ‘recycling/disposal’ phase is concerned. Hence, both after-use phases may be considered comparable. This conclusion may only be drawn, however, if both the raw materials and the product meet the environmental threshold values existing (e.g., for heavy metals).

Comparability of the utilisation phase of both materials is ensured by fulfilling minimum requirements of the functional unit (freeze-thaw-resistant concrete of exposition class XF2 and XF4). The necessary investigation basically covers strength development and freeze-thaw tests -CDF- (Dombrowski *et al.*, 2008), according to which geopolymer concrete is comparable to cement concrete or even better (as far as major technical properties are concerned).

The life cycle phase that is comparable for both materials may be excluded in a comparative LCA. The investigation will therefore focus on the production phase and the related pre-chains of resource extraction (Fig. 10.7). Although the transport of raw materials contributes at least noticeably to the environmental profile, it is not included in the system boundaries, because the efforts needed to transport the raw materials needed are quite different worldwide. The impact of transportation processes should only be assessed in a specific regional context.

The compositions of the compared systems are shown in Table 10.2. All mixtures have an identical content of gravel/sand and the concretes are produced without any chemical admixtures (e.g., superplasticisers). The Portland cement concrete mixture contains 340 kg cement/m^3, which is 20 kg above the minimum cement content for freeze-thaw-resistant concrete (XF2-XF4) according to DIN EN 206-1/DIN 1045-2. The already optimised geopolymer concrete (Dombrowski *et al.*, 2008) with a slag/fly ash ratio of approximately 80:20 does not need any heat curing and hardens at room temperature (20°C).

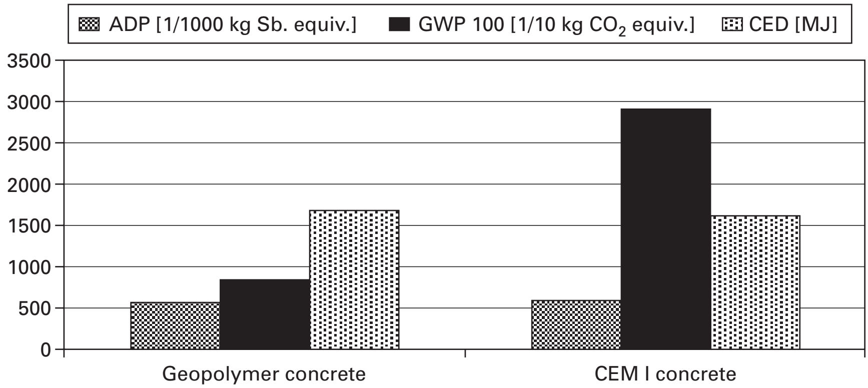
In the case where the utilisation phase and the recycling/disposal phase are considered to be equal, the results in Fig. 10.8 exhibit comparable environmental impacts in terms of ADP and CED of the geopolymer system



10.7 System boundaries for the comparison of geopolymer concrete and cement concrete production.

Table 10.2 Composition of geopolymer and cement concrete [kg/m^3]

Material	Cement concrete	Geopolymer concrete
cement CEM I 32.5R	340	
slag		230
fly ash		57
reactive waste		83
silicate solution/Na silicate (37%)		33
NaOH (50%)		24
de-ionised water	170	99
gravel/sand (0-16mm)	1878	1878
kg/m^3	2388	2404



10.8 LCA results of geopolymers and cement concrete.

for this specific application (freeze-thaw-resistant concrete). In contrast to this, the geopolymer concrete contributes much less to the global warming potential (GWP). In comparison to the Portland cement concrete, the GWP of geopolymer concrete is approximately 70% lower.

Compared to cement concrete, two aspects have to be considered critical:

- the cement content used exceeded the prescriptive minimum cement content (DIN EN 206-1/DIN 1045-2) by 20 kg per m^3
- Portland cement (with the highest energy and resource requirements of all known cement types) is used instead of blended cements, such as CEM II or even CEM III, which may be also be applied for class XF2-XF4 according to DIN EN 206-1/DIN 1045-2.

In fact, both mentioned aspects would improve the environmental profile of cement concrete. The reduction of the cement content to the required minimum (320 kg) will reduce the indicator values by 3–5%, use of a slag-rich CEM II cement will reduce the indicator values by 11–16%. In such a best case assumption for cement concrete and provided that such mixtures have the technical performance required, the cement concrete mixture will have noticeable advantages in terms of ADP and CED. But still, the geopolymer would have a considerably lower impact on the global warming potential (GWP indicator). To facilitate decision, the regional transport efforts have to be taken into account.

The utilisation phases of both systems are considered to be comparable, as the requirements according to DIN EN 206-1/DIN 1045-2 are fulfilled. Owing to its different carbonation performance, varying durability of reinforced cement and geopolymer concrete should be investigated from a technical point of view. By means of a durability model considering complex environmental conditions, the lifetime of each system could be assessed more detailed. Such lifetime predictions would be an important input for a more holistic LCA comparison of geopolymers and cement concrete.

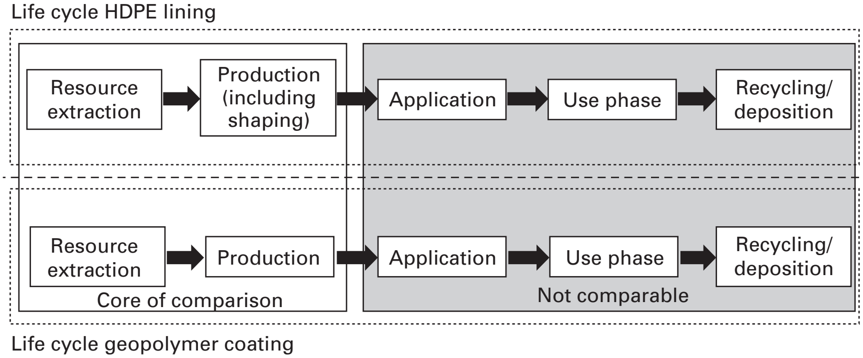
10.5.2 Comparison of geopolymer coating with HDPE liner systems in sewage sludge pipes

Geopolymers may exhibit remarkable technical performance, such as a high strength, high heat resistance, and high acid resistance. The latter property is used to design an acid-resistant coating for sewage sludge pipes. Unprotected concrete pipes are usually used for domestic wastewater transport. If increased resistance against acid is requested and/or a longer life cycle is desired, concrete pipes are often lined with high-density polyethylene (HDPE). Sewage sludge pipes with an HDPE lining are well protected against chemical and mechanical corrosion (Guermazi *et al.*, 2008).

In principle, geopolymer coatings are also qualified to protect a concrete surface against chemical attack (Bakharev, 2005; Bakharev *et al.*, 2003) and mechanical abrasion.

The system boundaries for a comparative investigation of both protection systems are shown in Fig. 10.9. The concrete pipe (the protected item) may be assumed to be identical and will not be considered within the system boundaries. The shaped HDPE ‘pipe’ is applied to the inner part of the tube directly after stripping the moulds of the concrete pipe without using additional adhesive components. In contrast to the HDPE lining, the geopolymer coating must be applied by a spray technique. There is no evidence of either application technique causing a considerable environmental impact.

Owing to the use of different materials in both systems (Fig. 10.9), the utilisation phase and the recycling/disposal phase might be different as well. In this case, the utilisation phase (including maintenance) probably contributes decisively to the environmental profile. The recycling/disposal phase is estimated to be of small importance. Since data on the application, use, and recycling/disposal phases are lacking, in a simplification the investigation



10.9 System boundaries for the comparison of geopolymers coating and HDPE lining.

focuses solely on the production phase. But it must be kept in mind that the significance of the results gained is limited, especially due to the exclusion of the utilisation phase.

The functional unit of the comparative assessment is a 100 m^2 protective layer:

- HDPE lining of 5 mm thickness (density 0.97 kg/dm^3)
- geopolymer coating of 10 mm thickness (density 2.23 kg/dm^3).

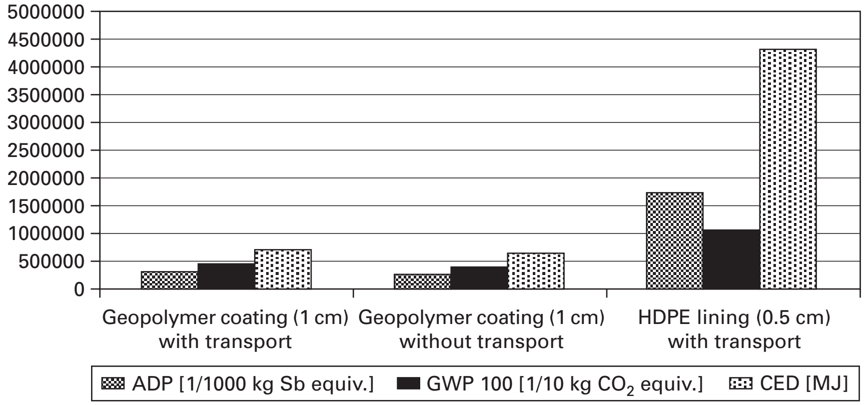
A suitable geopolymer mixture with very promising technical properties for this application has been designed by Anja Buchwald. The mixture contains:

14.3% soft coal fly ash (lorry transport distance 300 km)
6.1% metakaolin (lorry transport distance 300 km, ship transport distance 6000 km)
11.8% silicate solution (lorry transport distance 300 km)
2.2% NaOH solution (lorry transport distance 300 km)
3.9% water (no transport)
61.7% sand/fine sand (lorry transport distance 10 km).

To compare the HDPE lining (which includes already the transport of the raw materials) and the geopolymer coating system in a consistent way, a transportation scenario for the geopolymer-based system (focused on central Europe) is considered in addition (Fig. 10.10). The assumed transport distances for the raw materials are given above and are rather high (worst case assumption).

The geopolymer coating reaches much lower values for all environmental impact indicators considered. This is true for the geopolymer coating with and without consideration of raw material transportation.

The GWP of the HDPE system is higher by a factor of 2.3, the CED by



10.10 LCA results for geopolymers coating (with and without transport) and HDPE lining.

a factor of 5.9. As mentioned above, the use phase presumably contributes decisively to the environmental profile. Hence, the results presented do not favour one option, unless comparable information exists with respect to the durability of both systems.

However, the discussion may also be reversed. Although the geopolymer coating reaches only half of the lifetime of the HDPE lining and hence needs in-situ maintenance (recoating with geopolymer), it might have environmental advantages. The geopolymer coating might also be an attractive compromise between an uncoated concrete pipe and an expensive HDPE-protected concrete pipe for a moderately aggressive environment.

10.6 Geopolymers and the utilisation of secondary resources

A geopolymer composition can be optimised in terms of environmental impacts (and costs) by replacing primary raw materials with secondary resources. This kind of optimisation is recommended by the LCA results, but in practice is associated with several problems when good technical performance is desired. Secondary raw materials tend to have a greater variety of chemical compositions compared to primary raw materials. Consequently, technical performance of geopolymers also varies considerably, which is quite problematic for an industrial application. Some secondary raw materials also contain heavy metals, hence the use of such materials for industrial production might be restricted by environmental threshold values.

Another problem is the availability of secondary resources, especially these with a desirable low variation in chemical composition and a low content of heavy metals or impurities. Even if the availability varies a lot worldwide, the problem remains the same, as other traditional or new systems compete for these secondary resources. For instance, blast furnace slag is already used in traditional cement-based systems in Germany. Competition with a traditional utilisation of the same secondary resource does not facilitate market introduction of a new technology. Thus, the development of geopolymer mixtures for different applications should preferentially favour secondary resources which are not already used as raw materials to a large extent in other industrial sectors.

10.7 Conclusions

The environmental impact of products should be reduced to a minimum. To develop more environmentally friendly materials, the material designer needs some knowledge about the environmental drivers of new material systems, but also knowledge of the environmental impacts of competitive traditional material systems. Geopolymers can be produced from different raw materials at variable process conditions to achieve different properties which make them suitable for a variety of applications. Hence, the issue of environmental implications of geopolymers is a rather complex one.

This investigation reveals that a careful raw material selection should focus on both the solid and fluid components. To improve the environmental profile of geopolymers, the replacement of silicate solution and N_a-OH solution shows great potential. Furthermore, the application of heat curing for geopolymer production should be reduced to a minimum level, or waste heat from other processes should be utilised in order to reduce the accompanied environmental burdens.

A comparative assessment shows clearly the competitiveness of geopolymer systems with cement based systems from an environmental point of view. It will depend on the exact composition (in both systems), for a specific application field, to favour one option (assumed that the technical performance is comparable). The comparison of geopolymer pipe coating with an HDPE lining demonstrates a quite different picture. In this specific case the geopolymer system exhibits considerable environmental advantages as far as the production phase is considered. In general there is a great need to understand the durability and lifetime performance of geopolymer systems in comparison with traditional systems. Only with this important information can a final conclusion on the environmental impacts of geopolymer products be drawn.

10.8 Acknowledgement

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