CAN GEOPOLYMER CONCRETE PAVEMENTS CONTRIBUTE TO A SUSTAINABLE AND DURABLE FUTURE?

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ABSTRACT
This paper will investigate to what extent geopolymer concrete can contribute to the further sustainability of our infrastructure. The impact is examined in 4 areas: availability of raw materials, characteristics, circularity and experiences when applied in (road) infrastructure. A brief introduction to geopolymer concrete will also be given……

KEY WORDS
GEOPOLYMER / CO2-REDUCTION / ALTERNATIVE BINDER

1. INTRODUCTION

In the Netherlands, as in the rest of the world, people are constantly looking for ways to reduce the CO_2 footprint in general. Within the (road) construction process, possibilities to even further reduce the CO_2 emissions of concrete, and specifically cement, are then quickly looked at. '*To even further reduce*' because the Netherlands has been a global leader in the use of low-CO_2 cement for decades (figure 1).

The current CO_2 savings in cements used are mainly achieved by:

- High share of blast furnace slag cements
- Use of fly ash and alternative fuels/raw materials
- Transportation over water

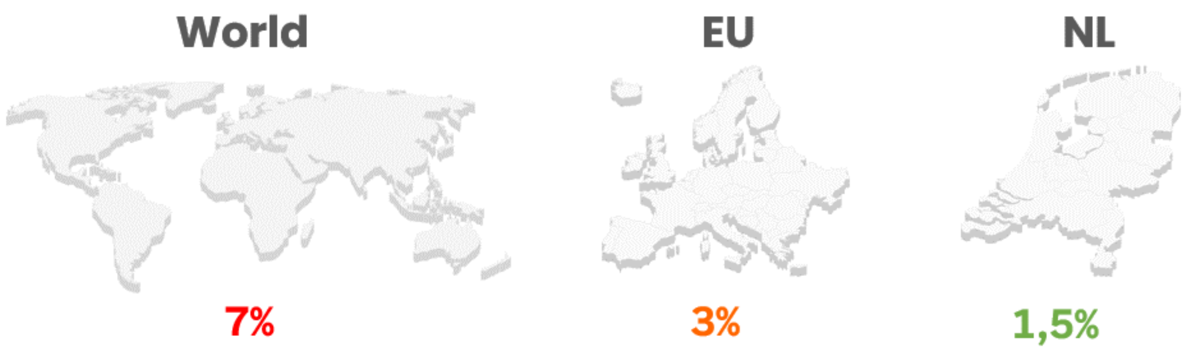


Figure 1 Comparison share of cement in total CO2 emissions

Within the search for a reduction of the CO_2 profile of concrete, the focus is mainly on binders with a low CO_2 profile. Geopolymer concrete is, in the Netherlands, increasingly seen as 'the solution' to significantly reduce the CO_2 profile of concrete. (Since the share of materials, and cement in particular, is relative large in the sustainability impact of concrete, a reduction in the share of these raw materials will mean a reduction of the impact (figure 4))

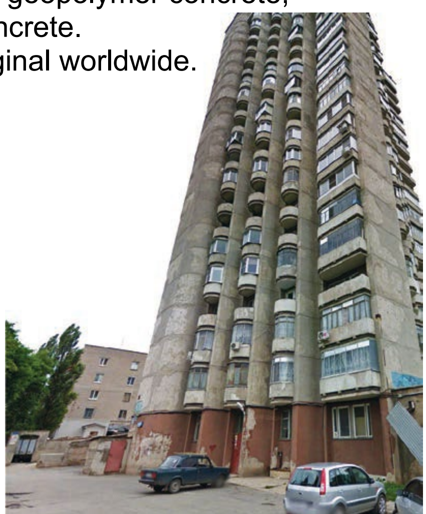
In addition to a much more favorable environmental profile, it would technically perform the same or better than regular concrete. (a claim that will be assessed in this paper based on practical experiences)
The vast majority of articles on geopolymer concrete, mostly from universities and research institutes, are positive about this material. However, the application is still very limited which is mainly due to the lack of regulations. However, there are more reasons why geopolymer concrete is not (yet) used on a large scale. In this paper, the environmental aspects and properties of geopolymer concrete are considered, we look at the obstacles to scaling up and we report on the extensive experience gained with geopolymer concrete within the ‘in-situ cast’ (road) infrastructure.

2. GEOPOLYMER CONCRETE

The term 'geopolymers' is a common designation for alkaline activated binders. Materials such as pulverized coal fly ash and blast furnace slag are used in this process not. Not as usual, activated with Portland cement clinker but with a strong base.
With raw materials that mainly contain silicon, such as pulverized coal fly ash, a more or less polymeric structure can be formed. With materials that also contain a lot of calcium, such as blast furnace slag, a CSH gel mainly forms, just like with regular cement. The calcium/silicon ratio in the CSH gel of 'geopolymer' concrete differs strongly from that in regular concrete. In alkaline activated slag, the ratio of calcium/silicon is approximately 1, while the ratio with regular cement stone is approximately 1.5 to 1.8.

The binder of geopolymer concrete usually consists of – as for regular cement suitable – raw materials such as blast furnace slag and pulverized coal fly ash and an alkaline activator. The activator ensures a high pH, so that the slag and fly ash dissolve and can react. The most common activators are sodium hydroxide (NaOH, referred to as caustic soda when an aqueous solution) and sodium silicate (Na_2SiO_3, water glass). Sodium carbonate (Na_2CO_3, soda) is also used as an activator. Compared to water glass and sodium hydroxide, the weak base soda is much less aggressive (lower pH) and more environmentally friendly, but also much less effective. However, in combination with heating, blast furnace slag can be effectively activated with it. An even weaker activator than sodium carbonate is sodium sulfate. However, in combination with blast furnace slag and a small proportion of Portland cement (5%), the strength development is sufficient, although it is not a fully alkaline activated binder. In this combination, sodium hydroxide is formed by the formation of calcium hydroxide during the hydration of the Portland cement clinker: $Na_2SO_4 + Ca(OH)_2 \rightarrow 2NaOH + CaSO_4$. The sulfate also aids in strength development through the formation of ettringite. This type of binder was marketed in Belgium in the 1950’s as Purdocement and was used to a limited extent. Examination of some structures that have been recovered, incidentally, show considerable carbonation at an age of approximately 55 years and, as a result, corrosion (Vanooteghem, 2011).

The first patent for a geopolymer binder (alkaline activated slag) was filed in 1908 issued in the US (US PATENT,1908). However, large-scale application accelerated in the sixties in Russia and Ukraine in particular, driven by a shortage of Portland cement in the former Soviet Union (figure 2).
In recent decades there has been a great deal worldwide research into geopolymer concrete, because of the potential to decrease significantly the CO2 profile of concrete.
Till now the application of geopolymer concrete remains relatively marginal worldwide.



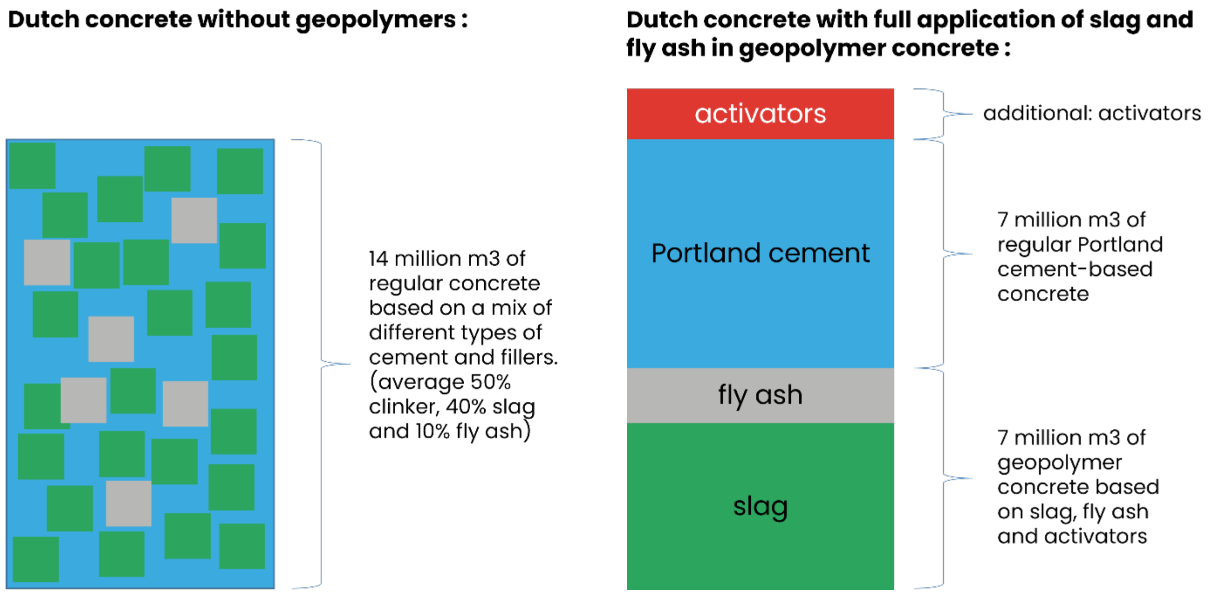


Figure 3 Comparison use of raw materials used in the Dutch cement and concrete industry

Geopolymers can contribute to a CO_2 reduction on the condition that no use is made of raw materials that are already fully used in the cement and concrete industry, such as blast furnace slag and pulverized coal fly ash. Blast-furnace slag and pulverized coal fly ash are scarce raw materials that are already used almost entirely in Europe (and blast-furnace slag worldwide) in cement and concrete, and Portland cement clinker has been partially replaced for many decades. By applying high contents of slag or fly ash in geopolymer concrete, but also, for example, in the form of blast furnace cement CEM III/C, therefore does not yield any environmental benefits. This only leads to a reallocation of raw materials (fig.3).

In The Netherlands, circularity and green public procurement are becoming increasingly important. Results from LCAs are used increasingly in public procurement tenders, especially in the Dutch construction sector. In these tenders, the Environmental Cost Indicator (ECI) is used as an important criterion to determine the winning bid. The Environmental Cost Indicator (ECI) unites relevant environmental impacts into a single score of environmental costs. In the Netherlands the ECI is known as MKI (Milieu Kosten Indicator).

At a local (project) level, geopolymer concrete can yield a low ECI, but on average the MKI on a national scale will not decrease and therefore neither will CO_2 emissions. Of course, due to increasing demand, we can import more slag and fly ash from neighboring countries than is already the case. In the Netherlands, the average MKI may fall, but of course the average MKI of concrete in the surrounding countries will then rise, because slag and fly ash are already almost completely used in concrete there too. Due to increased transport, total CO_2 emissions will even increase. The activators also result in extra CO_2 emissions. In addition, Portland cement clinker is a more effective activator than strong bases: to achieve the same strength, generally more slag is needed in geopolymer concrete than blast furnace cement in regular concrete.

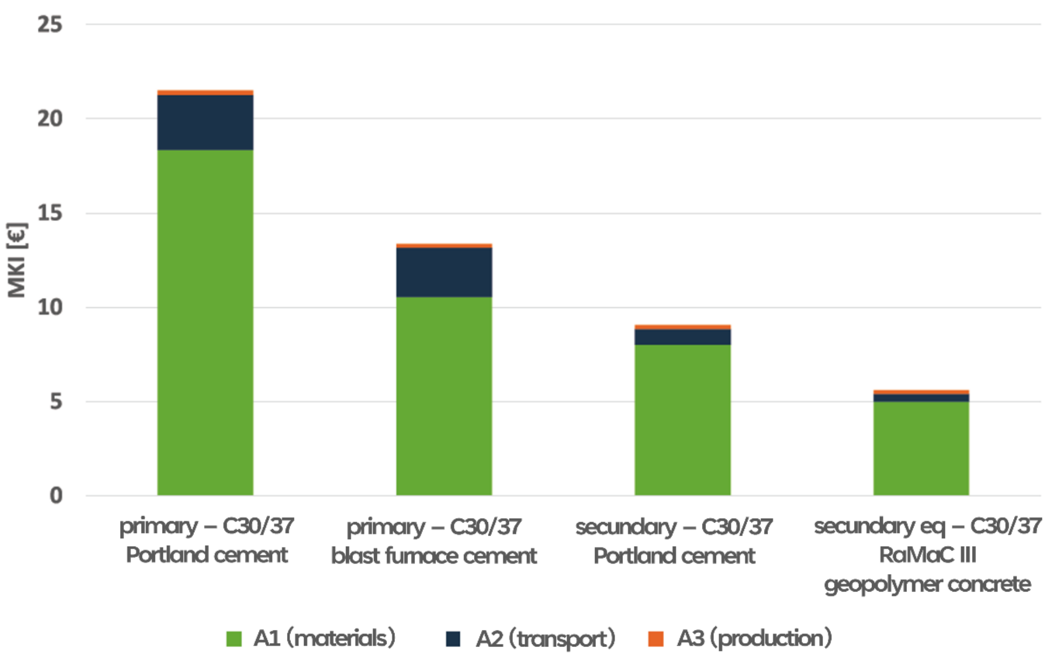


Figure 4 ECI (MKI in axes) scores per life phase A1 – A3 of different concrete mixtures.
(RaMaC = alkali activated Ready Mix Concrete with Sqape binder)

Geopolymers can therefore only contribute to limiting CO_2 emissions if materials are used that we do not yet use in concrete. This can be an artificially produced slag, but also secondary material flows other than pulverized coal fly ash and blast furnace slag. In the case of alternative secondary material streams, the available volumes are unfortunately usually limited. Calcined clay can be produced indefinitely and used as a raw material for geopolymer concrete. It is highly questionable whether there will be any environmental benefits compared to a regular cement with a low CO_2 profile.

3. AVAILABILITY OF RAW MATERIALS

It can be roughly stated that the effectiveness of a raw material in geopolymer concrete is comparable to its effectiveness in combination with Portland cement clinker. In cement, granulated blast furnace slag can replace the greatest share of Portland cement clinker and blast furnace slag is also most suitable for geopolymer concrete. When dosed with a high proportion of blast furnace slag, geopolymer concrete can be made that also develops good strength without heating. With less reactive materials, such as pulverized coal fly ash, ground brick, trass or calcined clay, very high doses strong activator and/or heating are necessary. As indicated earlier, the use of granulated blast furnace slag and pulverized coal fly ash in geopolymer concrete results in an environmental loss (instead of the environmental benefit that was looked for) but the volume of these by-products is also relatively limited. Together they make up about 13% of the global cement production (SGS INTRON, 2021).

There are other residual and waste streams that are suitable for making geopolymer concrete that are not yet used in cement and concrete, but the volumes of these streams are limited and considerably less than the amount of blast furnace slag and fly ash. For example, a relatively large amount of copper slag is released in Belgium which seems suitable for the production of geopolymer concrete. However, this is less than 200,000 tons per year, while approximately 6.5 million tons of cement are used in Belgium each year.

Clay is widely available and can suitable for use in both cement and geopolymer concrete after being calcined (heating between approximately 700 and 800 °C). It mainly concerns the type of clay kaolin (Chinese clay) which is rich in the mineral kaolinite ($Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$). By heating, the aluminum silicate is converted into metakaolin: crystal water is evaporated

and a complex more or less amorphous structure is created, giving it pozzolanic properties. Mainly because of the amount of energy required for heating, calcined clay has, compared to blast furnace slag and pulverized coal fly ash, a higher CO_2 profile (270 - 423 kg CO_2 per ton) (RILEM TC 282-CCL, 2022)(SGS INTRON, 2021)). This results in little or no environmental benefit in combination with activators when compared to regular cement-based concrete with a low CO_2 profile. Trass is also widely available and does not have to be heated but, as with calcined clay a very high dose of strong activator is required to achieve an acceptable strength development.
Presumably even more problematic for scaling up the production of geopolymer concrete is the limited availability of activators. The production of NaOH (caustic soda), the most important and most effective activator, amounts to approximately 60 million tons per year via electrolysis of a sodium chloride solution, which releases chlorine gas. The market for chlorine (Cl_2) is limited, which means that scaling up the production of NaOH is not easy (Luukkonena et al., 2018). Moreover, the demand for NaOH from other applications at this time is already greater than the actual production. NaOH is used, among other things, for the production of paper and for numerous chemicals processes. The limited availability therefore hinders the upscaling of geopolymer concrete (SGS INTRON, 2021).
The worldwide production of sodium silicate (water glass), an activator often used in combination with NaOH, is less than 10 million tons per year, which can be used to make a maximum of 50 million tons of binder (Scrivener et al., 2016). That roughly corresponds to 1% of the total global cement volume. But water glass also has many other applications, which means that current production can only be used for geopolymer concrete to a limited extent. The production of the weak base sodium carbonate is approximately 50 million tons, through extraction and mainly through a chemical reaction between limestone and atrium chloride (The Essential Chemical Industry). Roughly half of this is used for the production of glass. The production of sodium carbonate could be increased (and there are large stocks in the US), but this activator is not very suitable for the activation of widely available raw materials such as (calcined) clay and trass.

4. CHARACTERISTICS

The properties of geopolymer concrete strongly depend on the chosen combination of filler(s) with binder function (powders) and activator(s), while also minor variations in the composition of the powder(s) have more influence on the properties than when used in regular cement and concrete. However, there are some general statements to be made that apply to most variants of geopolymer concrete.

4.1. Constructive characteristics

Geopolymer concrete has, compared to regular concrete (at least, as long as it is not carbonated), a higher tensile strength at the same compressive strength. This is favorable for unreinforced concrete. In reinforced concrete, the higher tensile strength is generally unfavorable: to keep the crack width limited and avoiding the risk of brittle fracture often more reinforcement is necessary than with regular concrete. Geopolymer concrete usually shows much more shrinkage than regular concrete. Twice as much shrinkage is common, although there are also studies and geopolymer variants showing little difference. Geopolymer concrete also exhibits more creep than regular concrete (Shi et al., 2016). The fact that geopolymer concrete generally shrinks and creeps to a bigger extend compared to regular concrete is usually unfavorable from a structural point of view.

4.2. Deterioration mechanisms
With regard to various deterioration mechanisms, such as freeze-thaw cycles and penetration of chlorides, geopolymer concrete appears to perform well. However, the studies have almost without exception been carried out on young geopolymer concrete. The resistance to these degradation mechanisms is strongly reduced by carbonation, while the resistance to carbonation in particular is not a strong aspect of geopolymer concrete. During carbonation of geopolymer concrete, at least with alkaline activated slag, the CSH gel is directly affected (Puertas et al., 2005).
In addition to calcium carbonate, this creates a type of silica gel that is more porous and weaker than the original CSH gel. As a result, with geopolymer concrete, the cement stone does not become harder and denser due to carbonation like with Portland cement, but softer and more porous. This has a negative effect on the density and thus the resistance to, among other things, the penetration of chlorides, and it also strongly reduces the resistance to frost-thaw cycles.
In geopolymer concrete based on fly ash (and with it presumably also other pozzolanas, such as calcined clay and trass because of the low content of CaO), carbonation also increases the porosity and reduces the resistance to various deterioration mechanisms (10).

As a result, geopolymer concrete performs not as good in the real world than may be expected based on laboratory research on young (non-carbonated) concrete. By the way, laboratory research on carbonated concrete in the laboratory also appears not to be representative for practice, because the real world conditions (variably wet/dry) cannot be simulated in the laboratory (Pasupathy, 2018).

There is no doubt that geopolymer concrete can be used to build. But the common statement that the properties of geopolymer concrete are generally equal or better than that of regular concrete is, based on the above, incorrect.

5. CIRCULARITY
In the recent CROW-CUR guideline 2 ‘*Assessment system raw materials for suitability for circular concrete*’ (CROW-CUR, 2021), a criterion for the alkali content of concrete granulate has been included of up to 0.4% m/m. This criterion has been included to prevent ASR from occurring in new concrete with recycled concrete aggregate (RCA). As a rule, geopolymer concrete will not meet this criterion to fulfil. Geopolymer concrete must therefore be reused in geopolymer concrete when recycled or it must be demonstrated that there is no increased risk of ASR when used in regular concrete. However, that is not easy. The content of alkalis in geopolymer concrete is at such a level that the so-called ‘cement section’ of CROW-CUR Recommendation 89(CUR, 2017) cannot be used in case of reusing geopolymer concrete in the same way as traditional recycled concrete aggregate. Because the '*alkali contribution of other components*' already applies when 30% of ‘traditional’recycled concrete aggregate is used. With the reuse of geopolymer concrete this would be considerably higher, even when using blast furnace cement CEM III/B.
Therefore, ASR cannot be ruled out. With smart crushing, the alkalis will mainly end up in the cement stone fraction. The cleverly crushed gravel can be reused, but reuse of the cement stone fraction in regular concrete then becomes problematic.
Recycling geopolymer concrete separately and reusing it in new geopolymer concrete is in the trend practice is not easy to organize and it has not been demonstrated that there is no increased risk on ASR when reused. Geopolymer concrete can therefore not be regarded as circular for the time being considered.

6. EXPERIENCES DUTCH PILOT PROJECTS

Since 2016, about 50.000 m2 of road surface in the Netherlands have been made of geopolymer concrete, most of them (>95%) cycle paths. Cycle paths are seen as an ideal test in practice: Identical method of application as through roads (machine), limited traffic load (limited impact in case of failure) and an almost identical load with regard to frost-thaw load (identical use of de-icing agents).
Since the construction of the first pilot project in 2016, Cement&BetonCentrum (Betonhuis Cement) has been monitoring the state of the pavement. In particular, the road surface, joints and any crack formation are registered.

In this paper three of these geopolymer infrastructure projects are presented until their inspections in 2022:

1. Roundabout near airport Twente
2. Red colored bicycle lane near Almelo
3. Bicycle lanes along both sides of a regional road between Deventer - Holten

These projects will also be inspected in 2023. The latest observations will be incorporated in the presentation during the conference.

Despite the fact that 'only' 3 projects are presented in this paper, the experience with polymer concrete, as described in this paper, is considered across the entire acreage. The projects chosen for further in-depth study in this paper have therefore been deliberately chosen so that they represent a good picture of the total surface area of geopolymer concrete (in concrete roads) in the Netherlands.

The monitoring has been carried out over time by the same (2) persons and the findings have been recorded with high-resolution photographs. This way it we ensured that the assessment of the state of the pavement is as independent as possible.

6.1. Roundabout near airport Twente

Project information:
Location: crossing Vliegveldstraat with Oude Vliegveldweg
Constructed: November 2017

Way of construction: Manual, both main road (between formwork with vibrating beam) and truck apron (with print) and bicycle crossings in median strip.
Construction: Non-reinforced with dowels
Width main road: 5 m[1]
Particularities: sample plate in bypass for monitoring. No roundabout kerb applied; the truck apron is cut diagonally on the side of the carriageway. Main road surface was treated with water glass after pouring and before commissioning.

Figure 5 Aerial view of the location


Figure 6 Sample plate in by-pass

Visual inspections were executed in May 2019, March 2020, February 2021 end July 2022. First inspection after 1,5 years in March 2019 included two winter periods.
There is a difference in surface damage between main road and bicycle crossings and truck apron. The main road shows hardly any damage, but the bicycle crossings already show scaling. The same for the truck apron, where surface particles remain in the ‘joints’ of the printed concrete.


Figure 7 Main road surface in 2019


Figure 8 Main road surface in 2019


Figure 9 Surface of the bicycle crossing in 2019


Figure 10 Surface of the bicycle crossing in 2022

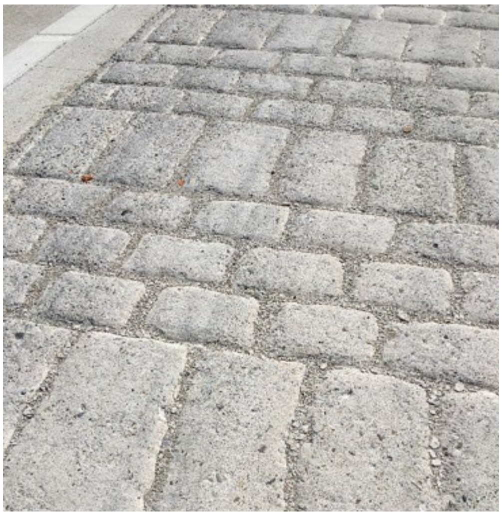


Figure 11 Truck apron in 2019


Figure 12 Truck apron in 2022


Figure 13 Truck Apron in 2019


Figure 14 Truck Apron in 2022

In 2022, after 5 years in use, the surface of the main road is still in good condition. Hardly any scaling. Probably due to the surface treatment with water glass after construction of the pavement as far as our information extends, no air entrainers have been used
The surfaces of the truck apron and the bicycle crossings showing severe damage by scaling. Increasing in time. The white markings deteriorate with the increase of scaling.
The edge of the truck apron looks like exposed aggregate concrete. De-icing salt increases the effect of scaling.

6.2. Red coloured bicycle lane near Almelo

Project information:
Location: Aadorpweg near regional road N36
Constructed: autumn 2019
Length: about 1 km
Way of construction: mainly with slipformpaver. Additional parts near crossings: manual, including curve at the end near a roundabout.
Construction: Non-reinforced concrete, throughout colored in red
Width: 4 m; every 4 m a transfer joint and every 100m an expansion joint filled with cold asphalt.
Particularities: at both sides a machine paved grey concrete kerb was constructed. Transfer slope to one side in the direction of the ditch.

Visual inspections were executed in March 2020, February 2021 and July 2022.

Figure 15 Overview of the bicycle path in March 2020

The first inspection took place in March 2020, six months after construction.
First impression was that the red bicycle path is looking nice. Especially with the grey kerbs on either side. But at a closer look, the detailing and finishing could be better. No wild cracks were observed. Contraction joints look tight.

The most northern part, near the N36, shows some spots with scaling. The longitudinal flatness leaves much to be desired over the entire route. The unevenness in the longitudinal direction is noticeable in the car. There are quite some irregularities in the surface of the cycle path.

The manually constructed parts: connecting bicycle crossings already show a lot of surface damage. The surface of the kerbs: the kerbs on the low side shows more damage (and red areas) than on the other side.

Figure 16 Difference between slipform paved and manual constructed (2020)

Figure 17 Severe and progressive scaling

Figure 18 2022…

Figure 19 Scaling of the kerbs on the lower side in time

Figure 20 Slabs with most severe scaling have been reconstructed

Just after no more than 2 years, almost all slabs are showing severe scaling, which looks progressive. The manually constructed slabs with the most severe scaling have been replaced.

Based on the current condition of the bicycle lane, the expected lifetime will not be reached.

6.3. Bicycle lanes along both sides of a regional road between Deventer - Holten

Project information
Location: N344 Holterweg, near Oude Molen
Constructed: autumn 2019
Length: about 11 km; on both sides of the road about 5,5 km.
Way of construction: slipformpaver.
Construction: Non-reinforced geopolymer concrete (C30/37),
Width: 2,25 m; every 4 m a transverse joint and every 100m an expansion joint filled with cold asphalt.
Thickness: 0,16 m

Visual inspections were carried out in March 2020, February 2021 and July 2022

The visual inspections were concentrated on the east part near Oude Molen in the direction of Holten, for a length of about 1 km. As well for the south as the north part of the bicycle lanes.
During construction of this southern part (about 1 km) wild cracks occurred very quickly. The Cement&BetonCentrum was involved to report the probable cause of this.
During the first inspection the occurred cracks in November were marked with red paint. Also other cracks were detected without marking with paint. At several wild cracks material is crumbling. The longitudinal evenness is OK for the north and the southern path.
The northern path has no wild cracks. But at several slabs scaling already occurred. Surface texture (with broom) has been added as well in transverse as in longitudinal direction.

Figure 21 Wild cracks southside, March 2020

Figure 22 Northside, March 2020

In the years after 2020 no further increase of cracking has been noticed. But a large degree of material loss can be observed over almost the entire length of the cycle paths, this even to the extent that clouds of dust are created with wind.

Figure 23 March 2020: Northern and southern side: scaling already visible

Figure 24 Surface with scaling in 2021

Figure 25 Surface with scaling in 2022

In about less than 3 years after construction the surface structure deteriorated into a structure like exposed aggregates. The comfort of a smooth surface for the cyclists disappeared.
The principal (Province of Overijssel) is considering overlaying these paths of geopolymer concrete with a standard concrete.

Pictures of surface details taken in July 2022 of project bicycle lanes Deventer- Holten :

Figure 28 Surface detail Deventer-Holten

Figure 29 Surface detail Deventer-Holten


Figure 30 Surface texture surface Deventer-Holten


Figure 31 Progressive scaling Deventer-Holten

7. CONCLUSION

- It is clear that, due to the limited availability of raw materials and activators, the production of geopolymer concrete, in its current form, cannot be scaled up in such a way that it can replace an important part of regular concrete based on regular cement.
- For most applications, the properties of geopolymer concrete are also inferior to those of regular concrete.
- Furthermore, geopolymer concrete cannot yet be considered circular.
  It is of course good to use residual flows that are not already used in cement and concrete, such as kop copper slag, to be used for geopolymeer concrete, when this application yields the most environmental benefit. In addition, geopolymer concrete can be used because of specific properties such as acid resistance.

Overall can be concluded that the potential contribution of geopolymers to the further sustainability of concrete is limited, specifically when applying to the in-situ (road) infrastructure.

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