Sustainable binders for the creation of a dredge-based tile

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ABSTRACT

This paper focuses on the search for sustainable alternative binders to cement. These binders will be used in a dredge-based tile created by the start-up Waterweg. After a literature review, a ground granulated blast furnace slag binder named Eco$_2$cem, a metakaolin-based binder named Topcrete, a sludge from wastewater treatment named Kaumera and lignosulfonate were selected and tested in the experimental phase. In this phase, samples containing these binders are created through either compression or pouring. These samples are analysed based on their flexural strength. The poured samples are generally stronger than the compressed ones. Lignosulfonate and Kaumera cannot fully replace cement, whereas Topcrete showed to be able to fully replace in compressed samples. Poured samples containing Eco$_2$cem as a sole binder showed great results that are similar to the flexural strength values of the control group that still contained OPC. Efforts should now go into the optimization of Eco$_2$cem- and Topcrete-based samples.

Key words: geopolymer – dredged sediment – flexural strength analysis – compression – pouring

LAY SUMMARY

Climate change is one of the defining crises of our time and the building industry is a large part of this crisis. The main reason why the building industry forms a large part of the climate crisis is the presence of concrete in almost every part of our built environment. Concrete consists of four main ingredients: gravel and sand to provide strength, cement as a binder to glue everything together and water to activate the binding of cement.

Cement is the main reason why the building industry is so polluting, as the chemical reaction that produces cement releases a lot of carbon dioxide. Additionally, sand is becoming increasingly scarce in the world due to the increasing need for building materials, which includes concrete. It is high time to find more sustainable binders than cement and a replacement for sand to ensure that building materials remain available in the future.

Waterweg is a start-up in Rotterdam that focuses on the utilization of dredged sediment rather than sand in water-permeable street tiles. Dredged sediment is a waste stream that currently has low value in our society; it is often stored at farms or even illegally dumped in nature reserves. Waterweg aims to provide an alternative for sand in building materials and valorises dredged sediment, putting a stop to these practices. At the moment, Waterweg still uses cement for the binding of their tiles, but the aim is to switch to a more sustainable alternative.

Our research investigates these alternatives to cement. We make a selection of alternative binders based on binding capabilities, the sustainable character, the availability and the state of the research field. We moved the following four binders to the experimental phase of our research:

- Eco$_2$cem, a waste stream formed during the production of iron
- Topcrete, a waste stream that is created by the burning of paper pulp
- Lignosulfonate, a chemical that is retrieved from plants and forms a waste stream of the paper industry
- Kaumera, a by-product that comes from the wastewater treatment

With these four binders, small tiles were created by either compressing a mixture or by pouring a mixture in a mould.

In general, samples that were created by pouring were stronger than samples that were created by compressing. We noticed that lignosulfonate and Kaumera did not suffice as replacements of cement, but they might be useful to Waterweg later as an additive for certain mixtures. Eco$_2$cem worked well as a binder in samples that were poured. These samples were as strong as the tiles that were compressed and still used cement as a binder. Topcrete worked fairly well as a binder in samples that were compressed, but it did not work well as a binder in poured samples yet.

Theoretical research was also carried out to analyse a possible third method of production next to compressing and pouring, namely 3D printing. This method has the potential to increase the sustainability of the final product of Waterweg and opens an avenue to the creation of different products made out of dredged sediment.

My advice is to continue the investigations into Topcrete and Eco$_2$cem, so that one these two binders can eventually fully replace cement and create a more sustainable product for Waterweg.

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ABBREVIATION LIST

DS = Dredged Sediment

EC = Eco$_2$cem

FTIR = Fourier-Transform Infrared Spectrometry

K = Kaumera

MKPC = Magnesium Potassium Phosphate Cement

OPC = Ordinary Portland Cement

RBW = Relative Breaking Weight

SEM = Scanning Electron Microscopy

SS = Siliceous Sand

TC = Topcrete

UCS = Unconfined Compression Strength

XRD = X-Ray Diffraction

XRF = X-Ray Fluorescence

Wt% = Weight Percentage

INTRODUCTION

When you look around you at the spot you are currently in, there is a large chance you will see a construction that contains concrete. It could be that you are seeing a building, a pavement or even a flowerpot. Based on this quick look at our current environment, it is not surprising that every person on this planet used about 1 tonne of concrete per year in 2007, making it the most utilized building material on Earth (Flower and Sanjayan). Concrete is favoured so heavily thanks to its excellent properties for the construction industry. It is chemically inert when hardened, it shows fairly good strength which can be improved through reinforcement and it lasts for a long time.

Concrete consists of aggregates, like sand and rocks, and a mixture that glues these aggregates together, the binder. In the case of concrete, this binder is cement. Unfortunately, the production of cement is polluting and energy-intensive. The cement industry emits 5 – 7 % of all CO_2 emitted by humanity (Meyer, 2009; van den Heede & de Belie, 2012; Worrell et al., 2001). Additionally, the production of cement consumes a large amount of energy and water (Meyer, 2009).

The main materials used to create cement are limestone, chalk and clay. These materials are pulverized and mixed to create a blend that is ready for the next step: pyro-processing to achieve calcination of the blend. Calcination is the heating of materials without airflow or oxygen to remove impurities. In the case of cement production, the impurity is carbon. During the process of calcination, the calcium carbonate in the blend undergoes the following reaction to produce calcium oxide and carbon dioxide (Meyer, 2009):

$$\begin{aligned} &\mathrm{CaCO_3\ (s) \rightarrow CaO\ (s) + \ CO_2\ (g)} \\ &\mathrm{1\ kg \qquad\qquad 0.56\ kg + \ 0.44\ kg} \end{aligned}$$

As can be seen from the reaction formula above, around 44 mass percent of calcium carbonate forms carbon dioxide. This is one of the main factors why the cement industry emits such large amounts of CO_2. The other important factor is the combustion of fossil fuels to apply heat for the calcination process to occur (Meyer, 2009). The main cement type produced is ordinary Portland cement (OPC).

Because of the large environmental impact of the concrete industry, it is high time to look into sustainable alternatives for the construction industry. The main goal of Waterweg is to create a water-passing tile made from dredged sediment to fit a climate adaptive city. The idea for these tiles originated with Wies van Lieshout and Eva Aarts. With the development of this idea Wies and Eva won the Blue City Circular Challenge in 2018. This challenge is a design competition organized by Blue City, a hub in Rotterdam that helps start-ups in the field of circular economy. Through winning this competition, Wies and Eva were able to turn their idea into the start-up Waterweg.

With the creation of water-passing tiles from dredged sediment, an opportunity arises to solve multiple problems at once. The first problem that can be solved is the excess of dredged sediment in the Netherlands. In the region between Rotterdam, Zoetermeer, Gouda and Schoonhoven alone, more than 150,000 m^3 of sediment is dredged every year (Hoogheemraadschap Schieland en de Krimpenerwaard [HHSK], 2020). In the period between 2002 and 2011, around 400 million m^3 of sediment was estimated to be dredged in the Netherlands (Advies en Kenniscentrum Waterbodems [AKWA], 2001). This dredged sediment is officially treated in the following four ways (Mul et al., 2020):

- Spread out on an empty field close to the dredging site
- Stored at farms, who are obliged to accept this storage by Dutch law
- Transported to a waste treatment facility
- Used in a project close to the dredging site, for example to heighten the ground below highways

Unfortunately, polluted dredged sediment is also dumped in nature reserves through illegal activities (Bosma, 2019). The main reasons for this are the large excess of dredged sediments and the lack of a specific use for this waste stream. All treatment options mentioned in the list above do not utilize the dredged sediment to create a high-grade product, meaning this waste stream is available for valorisation. The utilization of dredged sediment in a construction material like a water passing tile aids in the diminishing of the excess. This decrease of the excess hopefully eliminates the illegal activities that are taking place right now.

The creation of water-passing tiles from dredge sediment might also help reduce the worldwide sand shortage. Due to unawareness and a lack of supervision, the demand for sand has soared and will continue to rise drastically in the coming years if no action is taken (Bendixen et al., 2019). It is estimated that between 32 and 50 billion tonnes of sand and gravel are mined on a yearly basis (Koehnken & Rintoul, 2018), mostly for the concrete industry, alongside the glass industry and electronics (Bendixen et al., 2019). As mentioned before, sand and gravel are used in concrete as aggregates that are glued together by cement. The goal of Waterweg is to replace sand with dredge as an aggregate. If this goal becomes reality, the demand for sand from the construction industry can reduce over time.

The third problem that the tiles created by Waterweg can hopefully tackle is the lack of water-passing tiles in cities, for example Rotterdam. With heavy rainfall, cities often struggle to get rid of the water, as most pavements and roads are covered with non-permeable asphalt and concrete. The creation of a water-passing tile with a fitting foundation under the tiles can help alleviate the problems of heavy rainfall in urban areas.

This research will be focused on the search for a sustainable alternative to cement as a binder of the dredge-based tiles. The goal is to provide Waterweg with an overview of the most suitable binders. Suitability will be based on the properties of the binder, its environmental impact and the mechanical properties of the final product. These mechanical properties will be analysed in the experimental section of our research.

The following literature review provides an overview of the sustainable alternatives to cement as a binder for dredge sediment. First, a quick overview will be given of the types of tests that are needed for the characterization of our dredged sediment. This characterization is needed to optimize the mixture of dredge material and binder.

After this first section, an analysis of the possible alternatives of cement is carried out. Properties of the different alternatives will be given in this section. These properties will not be compared yet, as they are often dependent on the mixture they are in. We will compare these properties within our controlled mixture in the testing phase. Additionally, found information on the environmental impact of all found alternatives will be given. Finally, the availability of these binders will also be taken into account.

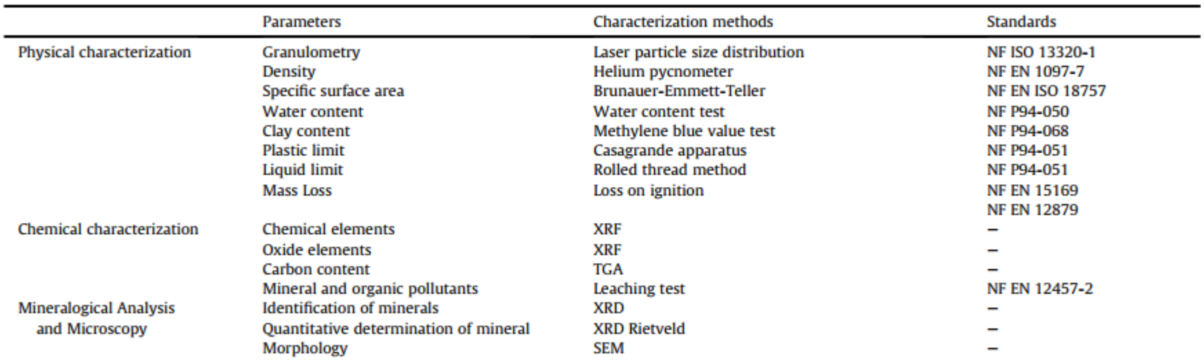
Based on the results found throughout this literature review, we will select the most suitable binders which will be tested in the experimental phase of our research.

LITERATURE REVIEW

Dredge sediment

In order to investigate the functionality of sustainable binders in combination with our dredged sediment, we need to have extensive knowledge on the physical and chemical character of our dredged sediment.

Amar et al. (2021) provide a clear overview of all the parameters that are potentially important to the characterization of dredged sediment. These parameters are summarized in Table 1.



	Parameters	Characterization methods	Standards
Physical characterization	Granulometry	Laser particle size distribution	NF ISO 13320-1
	Density	Helium pycnometer	NF EN 1097-7
	Specific surface area	Brunauer-Emmett-Teller	NF EN ISO 18757
	Water content	Water content test	NF P94-050
	Clay content	Methylene blue value test	NF P94-068
	Plastic limit	Casagrande apparatus	NF P94-051
	Liquid limit	Rolled thread method	NF P94-051
	Mass Loss	Loss on ignition	NF EN 15169 NF EN 12879
Chemical characterization	Chemical elements	XRF	–
	Oxide elements	XRF	–
	Carbon content	TGA	–
	Mineral and organic pollutants	Leaching test	NF EN 12457-2
Mineralogical Analysis and Microscopy	Identification of minerals	XRD	–
	Quantitative determination of mineral	XRD Rietveld	–
	Morphology	SEM	–

Table 1: Overview of parameters important for the characterization of dredged sediment. Retrieved from Amar et al. (2021).

Some of these parameters are already known for our dredged sediment, like the granulometry and the potentially toxic elements. The most important parameters that still need to be tested are the chemical elements and oxide elements through x-ray fluorescence (XRF), the qualitative and possibly quantitative identification of minerals through x-ray powder diffraction (XRD) and eventually the morphology through scanning electron microscopy (SEM). The XRF of chemical and oxide elements is of importance because a clear overview of these elements helps us in predicting possible reactions between these elements and elements in the binder. The XRD will give us insight in the mineral composition of the dredge, which aids us in the determination of the homogeneity of the dredged sediment. It might also give us insight in the reactivity and binding capabilities of the dredged sediment itself. XRD analyses of certain binders could also prove useful if the mineral composition is not yet clear upon arrival. Finally, the SEM provides a comparison tool on the morphology of different mixtures of dredged soil and binder. SEM could also be used to compare the morphology of dredged soil before and after mixing with a binder.

Binders

In this section, we introduce a variety of binders that could prove to be a suitable alternative to cement for binding dredged sediment. The field of sustainable binders is still in its infancy, hence the limited amount of literature on certain binders. This will be noted per binder, as it is a factor that influences our decision whether to test this binder in our lab. Additionally, just like dredged sediment, most binders are variable in composition due to their source; a waste stream. This will also be considered when deciding on the different binders.

Finally, if a study on a certain binder uses dredged sediment as an aggregate, the nature of this dredged sediment will be mentioned in this literature review.

Fly ash geopolymers

Fly ash is a residue retrieved from the flue gases that leave the boiler in coal-fired powerplants (Zhuang et al., 2016). The content and concentration of these fly ash residues is largely dependent on the type of coal that has been processed. For this reason, fly ash is a variable waste stream. The main components of fly ash are the following oxides:

- SiO_2
- Al_2O_3
- CaO
- Fe_2O_3

Fly ash can be classified as class C and class F based on the calcium oxide (CaO) content. Class C fly ash contains 20 wt.% (weight percentage) of CaO, whereas class F contains less than 10 wt.% CaO (Zhuang et al., 2016). If fly ash shows to be a suitable binder for our purpose, the difference of these classes on the mechanical properties of our tiles should be investigated.

Geopolymerization

The aluminium and silicium-containing components of fly ash can be used to form a geopolymer. A geopolymer is an aluminosilicate network that can express binding characteristics similar to cementitious materials (Duxson et al. 2007). Figure 1 shows a simplified overview of the process pathway for geopolymerization as it is now understood. There are still doubts about details of this pathway, as it is a new and developing field. It must also be noted that the steps shown in Figure 1 are often not distinctly separated and can occur simultaneously.

First, a solid aluminosilicate source is dissolved with the help of an alkaline activator (M^+ and OH^-). During this process the aluminate and silicate split up from the aluminosilicate source and this reaction consumes water. The dissolution of aluminosilicate goes very fast when in a solution of high pH. Next, a mixture of aluminate, silicate and aluminosilicate species is formed that gels at high concentrations of aforementioned species (Duxson et al., 2007). The duration of this gelation period varies based on the content of the solution, the type of aluminosilicate source and the working conditions. After this gelation period, the network continues to reorganize, which is specified in the second gel step. Eventually, the network is fully polymerized and the gel subsequently hardens. During each step after dissolution, the consumed water is expelled as a result of condensation in these steps.

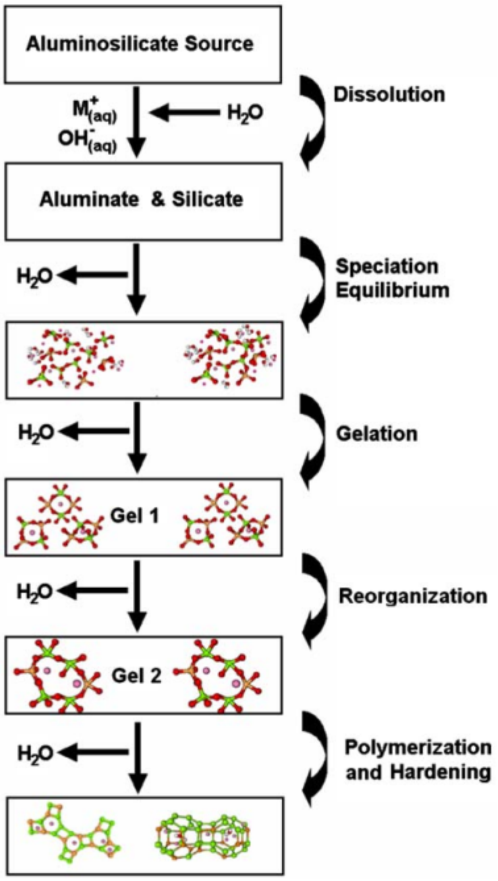


Figure 1: Process of geopolymerisation. Image retrieved from Duxson et al. (2007)

State of the research field

As opposed to other binders discussed below, a fair amount of research has gone into the creation of building materials from dredged sediments and fly ash. Lirer et al. (2017) produced cylindrical structures from a combination of fly ash with either siliceous sand (SS) or dredged sediment (DS). The fly ash used is classified as class F, meaning it is fly ash with a low calcium oxide content. The dredged sediment was retrieved from the harbour of Naples, Italy. The mechanical properties did not differ significantly between the DS constructs and the SS constructs. Also, scanning electron microscopy (SEM) analysis revealed a more compact surface structure for the DS constructs compared to the SS constructs, which might result in better mechanical properties of these DS constructs.

The results in this study are very promising for our study. Additionally, the experimental section for this paper is highly detailed and could definitely be of use to us in our testing phase. In this research Fourier-transform infrared spectrometry (FTIR) is used to define the degree of geopolymerization within the final product. This technique might be valuable to us later if geopolymers turn out to be the preferred binders for our dredged material.

Additionally, leaching tests showed that the produced specimens did not cross any boundaries set by Italian laws for non-hazardous waste. Italian laws take into account directives on this topic set by the European Commission.

More recently, it has been discovered that a combination of binders might result in the best chemical and mechanical properties of the product. Furlan et al. (2021) investigated different mixtures of fly ash with cement and/or lime, a mineral with a high calcium concentration. The calcium is present in oxide or hydroxide form. These mixtures were added to a sediment dredged from a bay close to Nantes, France.

The three test groups respectively contained 2% lime and 9% fly ash, 7% cement and 9% fly ash and finally 2% lime, 7% cement and 9% fly ash. These percentages are weight percentages of the dry mass of the dredged sediment. With these mixtures, cubic samples of 10 mm^3 were created through compression. Again, the methodology of this paper is highly detailed which could be very useful for our testing phase.

Unconfined compression strength results (UCS) and a drop in moisture content show that the addition of fly ash has a positive effect on the mechanical properties of the specimens.

A very interesting idea by Wang et al. in 2020 is the addition of fibres to a mixture of cement, fly ash and dredged sludge. In order to improve the poor mechanical properties of dredged sludge, chemical stabilization or physical reinforcement are needed (Wang et al., 2020). Currently, chemical stabilization is preferred and often lime and/or cement are used for this. However, due to the high emission profile of lime and cement, this solution is not ideal. Additionally, the alkalinity of these binders often inhibits growth of flora (Wang et al., 2020).

Physical reinforcement of dredged sediment through fibres is an alternative that might alleviate some of the problems that come with the exclusive use of chemical binders such as lime and cement. Wang et al. tested a variety of different fly ash, cement, dredged sludge concentrations and polypropylene fibre contents. This study used dredged sludge rather than dredged sediment because of the higher water content. This made the mixing of fibres through the sludge easier. The sludge was retrieved from a river in Nanjing, China. The final product was a cylinder.

During this project it was noticed that inclusion of fibres did not have a significant effect on the water content of the final product. The reinforcement with fibres did however significantly increase the UCS of the cylinders. The fibre content that reached the highest UCS was 0.1%. Additionally, the fibres can help reduce stiffness and brittleness that often comes with the chemical stabilization through cement and/or fly ash. The 'bridging effect' of these fibres (see Figure 2) plays a role in this.

The fibres used by Wang et al. (2020) are made of polypropylene, which is a plastic that is recyclable but not biodegradable. As the study by Wang et al. (2020) is very recent, we believe that there are still a lot of improvements possible on the environmental aspect of this approach through the utilization of more sustainable fibres.

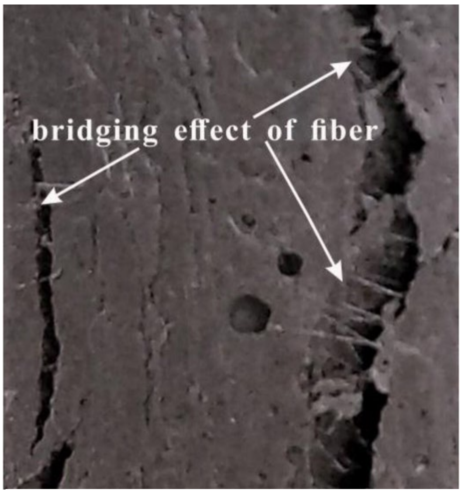


Figure 2: The 'bridging effect' of polypropylene fibres within a stabilized dredged sludge construct. Retrieved from Wang et al. (2020)

Environmental aspect and verdict

Fly ash can provide a 80% reduction of carbon dioxide emissions compared to OPC (Davidovits, 1993). This is a great improvement but even though fly ash is a waste stream, it is still retrieved from a very polluting process, namely the incineration of coal. Due to the high emissions related to coal-fired power plants, the government in the Netherlands and other countries are shutting down these plants in favour of cleaner energy sources (Purtill, 2020; van Santen, 2020). Fortunately, fly ash is not the only source of geopolymers so we first believed this binder should still be moved to our testing phase to familiarize ourselves with geopolymers. This decision was based on the relatively large amount of external research that has gone into this binder and the promising properties of the binder itself.

We did receive fly ash from a waste treatment facility in Duiven, the Netherlands, but unfortunately it was not possible to meet the safety requirements needed to work with this fly ash in our lab. This fly ash was too polluted with lead to be handled safely.

Fortunately, geopolymer-based binders can also be created from slags, volcanic rocks and even biomass fly ash (Saeli et al., 2019). Hence, we moved onto slag-based geopolymers.

Slag-based geopolymers

Slag is a waste stream created by the metal industry during the processing of metal ores and cokes. The slag produced during the processing of iron is especially useful as a binder. The composition of these slags varies largely based on iron ore and coke sources, but the slag consists mainly of lime, silica, alumina and magnesia (Ramezanianpour, 2014). The silica and alumina sources make geopolymerization possible. The main minerals present in these slags are melilite, as well as merwinite, diopside, lime, wustite and ferrite (Ramezanianpour, 2014). Slag is often granulated after the production of iron and subsequently ground. This process also determines the properties of the slag as a binder. The material that is produced is called ground granulated blast furnace slag (GGBS).

Slag shows very little capabilities as a binder on its own. However, when it is introduced to a highly basic environment, slag can be activated to show cementitious properties. Examples of compounds that can create this basic environment for slag are Ordinary Portland Cement, sodium hydroxide and sodium carbonate (Ramezanianpour, 2014; Slama et al., 2021). Unfortunately, the exact mechanism behind the activation of slag is still unclear, but it seems to follow the geopolymerization reaction pathway.

Ground granulated blast furnace slag has recently been combined with dredged sediment from a harbour in Tunisia to create road layers (Slama et al., 2021). This research is still ongoing, but the initial tests are positive. Even though road layers are different from the tiles that Waterweg aims to create, it is encouraging to see that dredged sediment can be combined with GGBS to create a load-bearing structure.

In 1999, 33.8 million tonnes of granulated blast furnace slag was produced in Europe alone (Ramezanianpour, 2014). This number probably changed over the years but it is clear that this is a large waste stream that could be put to good use as an alternative binder to cement.

We were able to retrieve a sample of ground granulated blast furnace slag (GGBS) from Ecocem Benelux. This GGBS is named *Eco₂Cem* and is already used commercially in the foundation of windmills and bridges (Ecocem, 2021). It was also used in a different study on the activation of slag by Yuan et al. (2017).

Based on the promising binding capabilities of GGBS, the availability of a sample and the fact that it is already being used as a binder, we made the decision to move Eco₂cem to our testing phase.

Metakaolin based geopolymers

Metakaolin is usually not a by-product, as it is created by incinerating kaolin, or *China Clay*, an abundant clay mineral (Ramezanianpour, 2014). During this heating, also called calcination, the following reaction occurs where the chemical structure of kaolin changes to release water:

$$\underset{kaolin}{Al_2Si_2O_5(OH_4)} \underset{800\ ^oC}{\rightarrow} \underset{metakaolin}{Al_2O_3.2SiO_2} + 2H_2O$$

To create metakaolin, the temperature that kaolin is exposed to should not exceed certain limits, as kaolin will then be burned and become unreactive.

Metakaolin usually has a particle size of less than 2 mm and mostly consists of aluminium oxide and silicium dioxide. Metakaolin does not require a basic environment to activate its binding properties, like GGBS. It is also mentioned that metakaolin increases the longevity of concrete constructs (Ramezanianpour, 2014). However, there seems to be no sign of studies that successfully replaced all cement by metakaolin.

We found a product named Topcrete that contains metakaolin. Topcrete is a product created from paper at a waste treatment facility in Duiven, the Netherlands. After around 7 times, paper fibres cannot be recycled anymore. The paper fibres that cannot be recycled again are heated in an installation and processed to form Topcrete, a material with a high calcium carbonate and metakaolin content. Additionally, the heating of paper pulp also supplies heat and electricity for households in the Netherlands (Afval Verwerking Rijnmond [AVR], 2021). Topcrete is currently used by some construction companies to replace cement. Unfortunately, we were not able to get in contact with these construction companies.

Topcrete seems to have a fairly sustainable character and is freely available, so we decided to move Topcrete to our testing phase.

Kaumera

Kaumera is made from a waste stream coming from the wastewater treatment. Kaumera consists of a variety of organic materials that are extracted from sludge granules which appear during water purification in the Nereda® process (van der Knaap et al., 2019). Kaumera consists of ~ 50% proteins, ~ 25% polysaccharides and the last ~ 25% are different organic materials like humic acids. Additionally, Kaumera contains 2-3% phosphate (P) and 6-9% nitrogen (N) as a percentage of the dry matter (van der Knaap et al., 2019).

Kaumera has an affinity for multivalent cations. When Kaumera is combined with a calcium source, it reacts to form small blobs that float on the surface. This affinity of Kaumera will not play a role with the type of dredged sediment we currently use, as the multivalent cations present in our dredged sediment have very low concentrations. This affinity does need to be taken into account if polluted dredged soil is used for future projects. Additionally, Kaumera could be combined with a secondary binder containing multivalent cations to possibly enhance the binding capabilities of both binders.

Unfortunately, when Kaumera is stored, it has a tendency to start moulding (van der Knaap et al., 2019). By changing the storage conditions, this can be avoided. However, if Kaumera is used within our tiles, these storage conditions cannot always be altered, as these tiles will move outside after the hardening process. Additionally, we noticed the moulding of our tiles after storage for around half a year in our lab. The possibility of mould formation needs to be taken into consideration. A possible way to avoid moulding is to increase the pH of the Kaumera and dredge mixture, as an increased pH generally reduces pathogenic activity (van der Knaap et al., 2019). Calcium carbonate or sodium hydroxide are possible additives to increase the pH. The use of calcium carbonate is a bit tricky due to the affinity of Kaumera with multivalent cations, including calcium. However, this research will not focus on eradicating mould activity. Future studies can help identify the gravity of this problem.

State of the research field

Kaumera is not known for its binding capacities. It has been discovered that Kaumera works well as a bio stimulant or as a coating on multiple materials. It also works as a curing compound in concrete to decelerate drying in climates where the temperature is too high or where the humidity is too low. A design studio in Arnhem named "Omlab" has been able to utilize Kaumera as a binder in a 3D printing project, but the properties of their resulting product do not suffice for dredge-based tiles.

Kaumera is a product that might be commercialized in the future. For this reason, literature is only available that is written by the creators of Kaumera. This situation makes it tougher to make an objective judgement of this possible binder. However, thanks to the connection with the creators of Kaumera, we are able to discuss our progress with them.

Environmental impact and verdict

An LCA for Kaumera has only been conducted with the aim of using it as a bio stimulant, making it difficult to critically assess the sustainable character of Kaumera with the aim of using it as a binder. The extraction of Kaumera from wastewater sludge does help decrease the amount of sludge that needs to be removed by 20 – 35 % (Kaumera, 2021). Between 33 and 47 MJ of energy is needed to produce a kg of Kaumera. The main chemicals used in the extraction process are hydrochloric acid and a form of hydroxide. Additionally, wastewater treatment is a process that will be continued for a long time, which secures the availability of Kaumera. Judging from this small data sample, it seems like Kaumera is a sustainable alternative to cement. However, more research needs to be conducted to quantify the sustainability of Kaumera.

All in all, Kaumera might be a suitable alternative to cement as a binder. It has a fairly sustainable character and will most likely be available for a long period of time. The main concern is the binding ability of Kaumera. This can only be analysed in practice so for this reason we want to test Kaumera in our lab.

Lignin

Lignin is an important molecule in plant-based lifeforms, where it provides strength and protection. It is a complex polymer that forms the second most abundant source of aromatic bindings behind petroleum (van Dam et al., 2016). Lignin is a polymer that has no regularly repeating set of monomers, making it a heterogeneous and highly variable material. Due to this variability, lignin does not have a crystalline structure. The large amount of aromatic structures within lignin give it a brown colour. For this reason, lignin is removed from paper pulp during the production of white paper. As paper production is a large industry, lignin has often been viewed as a by-product and not as a valuable resource in itself (Ekielski & Mishra, 2021).

The removal of lignin is a difficult process, as lignin has a high tendency to interact with other components within the plant. Multiple processes have been developed to isolate lignin (Ekielski & Mishra, 2021):

- Kraft process
- Organosolv process
- Sulfite process
- Soda process
- Hydrolysis process

The properties of the isolated lignin, also named technical lignin, are highly dependent on the isolation process and the type of biomass that the lignin is isolated from.

Environmental impact

Lignin is in theory an inexhaustible resource, as it is a large waste stream in the paper industry. Additionally, plant-based lifeforms can be grown for the purpose of lignin extraction. Additionally, there are waste streams that contain lignin, like grass cuttings. Grass contains a smaller concentration of lignin than hardwood, so whether lignin extraction from grass is worthwhile still needs to be determined. Grass cuttings are freely available for Waterweg through water authorities around Rotterdam, which would help closing the cycle of this waste stream and making it circular. Finally, there is a risk that certain lignin-containing material streams are mistakenly seen as a waste stream, like crop residues for example. Crop residues are vital to the soil of agricultural lands and crop residue removal can only be done in a harmless manner in specific cases (Blanco-Canqui & Lal, 2009).

State of the research field

The field of lignin binders is still in its infancy, especially when it comes to the use in construction materials. There are projects where lignin was used as a binder in consumer products, like the research by Jiang et al. in 2019. In their research, Jiang et al. reinforced a cellulose paper material with a lignin binder. This new combination of cellulose and lignin showed to have a much higher isotropic tensile strength and Young's modulus than plastics or combinations of plastics with either lignin or cellulose. A tensile strength test determines the load that a material can bear when stretched and a higher Young's modulus results in a more rigid material. It is also mentioned that lignin has a low material price, thanks to its abundance in plant-based lifeforms. It is proposed that this newly created material is used in medicinal packaging for example.

In 2017, Xie et al. mentioned the use of lignin as a binder modifier for road asphalts. In this study, no binding capabilities of lignin were mentioned. In the same year, Liu et al. created a hard material from a cement-lignin binder and marine dredge from Shanghai. Unfortunately, the exact composition of the binder was not mentioned. In 2021, Liu et al. used a lignosulfonate binder to granulate a mixture of tea waste and limestone. This lignosulfonate binder was supplied by Borregaard, a company that provides lignin for a variety of purposes.

Through the paper by Liu et al. (2021), we learned about Borregaard and decided to contact them. Borregaard is a company that originated in Norway and has expanded with locations over the whole world. The source of lignin used for their products are mainly Norwegian spruces. Next to lignin, cellulose, vanillin and bioethanol are also extracted from these spruces (Borregaard, 2020). They have clear goals when it comes to sustainability and all the wood they use within their biorefineries is certified.

As their binders were freely available for testing and their sustainable character seems decent, we decided to also test lignin binders within our lab. We received two binders from Borregaard, which are lignosulfonate and ammonium lignosulfonate. These products are retrieved from the sulphite process mentioned above. The sulfonate groups make lignosulfonate water soluble, which is a property that many lignin products do not possess (van Dam et al., 2016). Lignosulfonates are mostly implemented as plasticizers for the creation of concrete (Gargulak, Lebo & McNally, 2015).

Magnesium potassium phosphate cement

Another promising option as an alternative for OPC is magnesium potassium phosphate cement (MKPC). Phosphate binding was first used in dental surgeries where quick hardening was required (Chau, Qiao & Li, 2011). This quick hardening is one of the strong characteristics of MKPC, which is currently researched on its applicability in construction materials and radioactive waste immobilization (Pyo, Um & Heo, 2021; Gardner et al, 2021).

The binding reaction with MKPC comes from an acid-base reaction between dead-burnt magnesium oxide (MgO), monopotassium phosphate (KH_2PO_4) and water. The formed product is called struvite-K ($MgKPO_4$) (Chimphango et al., 2021; Gardner et al., 2021).

$$\mathrm{MgO\ (s) + KH_2PO_4\ (s) + 5\ H_2O\ (l) \rightarrow\ MgKPO_4 \cdot 6\ H_2O\ (s)}$$

Advantages that MKPC has over OPC is an almost neutral pH, a small amount of water needed and the quick hardening mentioned earlier (Gardner et al., 2021). A neutral pH provides a more friendly environment for the growth of flora.

As it is difficult to retrieve samples for this binder and the sustainability of this product is unclear, we decided not to move this binder to our testing phase.

Dredged sediment

Ferone et al. (2013) investigated the possibility of using dredged reservoir sediments to form geopolymer binders. These sediments were retrieved from various lakes in the south of Italy. Sediments were dried in an oven at 105 °C. The sediments contained an average of 49.4 wt.% silicium dioxide and 16.0 wt.% aluminium oxide and these were the most prevalent compounds within the sediments. Two test groups of sediment were thermally activated at respectively 650 °C and 750 °C. This thermal activation step showed to enhance polycondensation of the sample. Enhanced polycondensation results in a higher mechanical strength.

Additionally, leaching tests were conducted to classify the sediments. The results concluded that these sediments can be classified as non-hazardous waste following the European Waste Catalogue (Ferone et al., 2013).

The research into dredged sediments as a resource for geopolymeric binders is still very much in its infancy. Additionally, there are no dredge-based binders readily available as of now. For these reasons we have decided to omit this type of binder for now. It does remain a promising option, especially if other binders become scarce and a new source of geopolymers is needed.

Conclusion

Based on the amount of research found, the availability of the binders and the environmental impact, we have picked the following binders to move to our testing phase:

- Eco_2cem
- Topcrete
- Kaumera
- Lignin

For most binders, depending on the literature and advice from the company that produces the binder, we will start with mixing it in with cement and then gradually increasing the concentration of sustainable binder while decreasing the cement concentration.

3D printing

Next to the choice for a binder of dredged sediment, there is also a variety of production mechanisms with different influences on the carbon footprint of the production process overall. Within the experimental phase of our research, mixtures will either be compressed or poured in a mould. These mechanisms are two of the most utilized ones in the building industry at the moment of writing and for this reason have the most literature dedicated to them. Utilizing these two mechanisms allows for comparisons between our product and products that are currently available and created using the same methods.

An alternative method that could be used, is additive manufacturing (AM), which is the umbrella term for a variety of techniques that create three dimensional objects by adding layers on top of each other. An example of additive manufacturing is 3D printing. Additive manufacturing provides numerous advantages over pouring or pressing of cementitious mixtures. First, most mixtures are prepared within the additive manufacturing apparatus, removing the health risks that are associated with the handling of cement (Bos et al, 2016). An advantage of 3D printing is that less material can be used to achieve similar results to poured or pressed constructions and no material is needed for a mould, which leaves the pouring method at a disadvantage. Also, different materials could be printed simultaneously to create complex structures which cannot be produced through pouring concrete (Bos et al., 2016).

Finally, less physical work will be needed with AM, especially when an apparatus can produce the desired construction on-site. The removal of physical work allows for a larger age group that is suited for this industry, which might provide new job opportunities (Bos et al., 2016).

At the moment, three types of 3D printing have been applied in the concrete industry (Wu, Wang & Wang, 2016; Gosselin et al., 2016)):

- Concrete printing
- Contour crafting
- D-shape

The mechanism behind concrete printing - which is a very confusing name as all three techniques involve the printing of concrete - and contour crafting is based on the extrusion of a concrete mixture layer by layer in open air. The concrete mixture is extruded from a computer controlled nozzle and the extruded material is then pushed in the right direction with a trowel accompanying the nozzle (see Figure 3). The main difference between concrete printing and contour crafting is the higher resolution of printing for concrete printing.

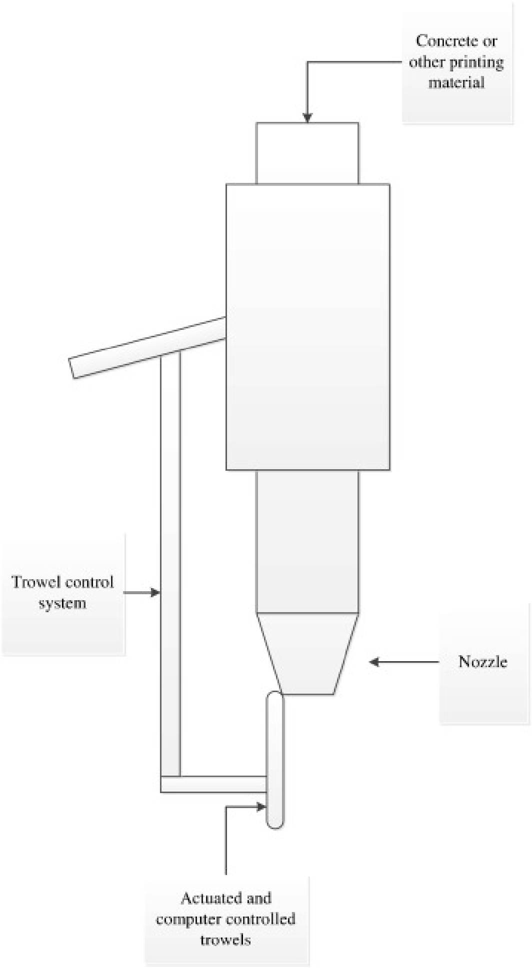


Figure 3: an overview of the process of contour crafting and concrete printing. Figure taken from Wu, Wang & Wang (2016).

Finally, D-shape printing is similar to an inkjet printer. A nozzle moves through a powder bed while extruding a mixture that glues the powder together layer by layer. In the case of D-shape printing of concrete, the glue is a mixture of cement, while the powder is sand (Lowke et al., 2018).

Although the field of concrete 3D printing is very promising and developing rapidly, there are still various hurdles to overcome before this technique can be applied widely. Firstly, the body of knowledge on 3D printing of materials for the building industry is still small (Bos et al., 2016; Wu, Wang & Wang, 2016). This is due to the novelty of this field in general and the secrecy of companies researching topics in this field.

Additionally the varying consistency of the extruded filament and the different levels of hardness per layer due to the curing of the concrete make it difficult to create a stable end product. Innovative materials, for example containing fibres, could help tackling some of these problems (Korniejenko & Łach, 2020)

Only one source could be found on multiple literature research engines (WebofKnowledge & Google Scholar) regarding the additive manufacturing of a mixture containing dredged sediment. In 2019, Liu and Liu explained at a conference that they were able to print a vault with the use of dredged sediment retrieved from lake Erie in Ohio in the United States. Before printing, the dredged sediment was successively dried, pulverized and sieved with a mesh size of 150 µm. The mesh size that Liu and Liu have chosen filters out a large amount of organic matter, gravel and sand. The main components that go through this sieve are silt and clay. This is in line with the choice they made for a 3D printer that is well-suited to print clayey materials. A water content of 60 wt% was reported to show the best results. With this water content, a filament of 4500 mm in length could be printed in up to 7 layers of 4 mm thickness (Liu & Liu, 2019). The layers at the bottom of the construct would not change under the pressure of the upper layers. The optimal shear strength range of dredged sediment for their 3D printing setup was 0.05 – 0.15 kPa. After printing, the constructs were left to dry in air and subsequently heated in an oven to increase their strength, just like other clayey constructs would. Finally, an analysis was carried out of the construct design to analyse the points in the design that would undergo the most stress due to the particular build. The design was then modified to spread these forces more evenly over the construct.

We also visited the design studio Omlab in Arnhem to gather more knowledge on the additive manufacturing of new materials. The people at Omlab created structures with a mixture of cellulose, Kaumera, calcite and water. They had also previously used river clay to create 3D printed constructs. One tip of theirs was to assess whether a mixture is right for 3D printing by running it through a regular plastic syringe (Looze, 2021). If the extruded filament is uniform and it does not take a large amount of pressure to push the material through the nozzle, the mixture is probably usable for 3D printing.

Additionally, we found promising studies on the replacement of cement with geopolymers like fly ash in the mixture used for additive manufacturing (Panda et al., 2017; Korniejenko & Łach, 2020). This is of great importance, because these alternatives are desperately needed to tackle the carbon footprint of the construction industry.

The future of the additive manufacturing of concrete is looking bright as the technology has a lot of potential and an increasing amount of research is published in this field. A realistic possibility is that additive manufacturing finds a niche next to conventional methods of production like compressing and pouring (Panda et al., 2017).

For Waterweg, additive manufacturing provides an additional pathway to reduce the carbon footprint of a dredge-based product, while offering new possibilities in designing this product. The research by Liu and Liu (2019) provides an accessible methodology to get started with the additive manufacturing of silt and clay present in dredged sediment. Bos et al. (2016) noted that their research group created a printing apparatus at the Technical University of Eindhoven. My advice would be to get in contact with this research group to gain more knowledge and to discover possibilities of printing dredged sediment with a sustainable binder.

MATERIALS AND METHODS

1. Materials

1.1 Material preparation

Dredged sediment (DS)

All dredged sediment (DS) used in this study comes from the same batch that was retrieved from the municipality of Rotterdam.

Upon retrieval, the DS was dried in an oven at 105 ^{o}C for 60 min to remove all water without changing the chemical composition of the DS drastically. Next, the DS was sieved with a mesh size of 3 mm. This process removed large pollutions like twigs and pieces of plastic. The DS is now ready for sample creation.

Lignosulfonate

Two different types of lignosulfonate have been tested, namely ammonium lignosulfonate and lignosulfonate. Both were retrieved in powder form.

Ammonium lignosulfonate was retrieved from Lignotech Florida in powder form. Total solid content is 98% and the pH is 4.3 (Lignotech Florida, 2018).

Lignosulfonate was retrieved from Borregaard in Norway. The pH range of this binder is 3.5 – 5.1. The exact chemical composition of this binder was not noted (Borregaard AS, 2021).

Kaumera (K)

A Kaumera solution was retrieved from the Kaumera installation in Zutphen, the Netherlands. The concentration of Kaumera is 7.45% in water.

Topcrete (TC)

Topcrete was retrieved in powder form from the waste treatment facility in Duiven, the Netherlands. The chemical composition of Topcrete is the following (AVR Afvalverwerking B.V., 2014):

Chemical name	**Weight %**
Calcium Carbonate	40 – 50
Meta-kaolin	25 – 30
Calcium oxide	20 – 25
Manganese oxide	< 5
Titanium dioxide	< 2
Di-iron trioxide	< 1

Table 1: Chemical composition of Topcrete

Eco2Cem (EC)

Eco$_2$cem was retrieved in powder form from Moerdijk, the Netherlands. The exact chemical composition of this binder was not noted. As mentioned before, Eco$_2$cem needs an activator in the form of OPC or an alkaline solution to express binding properties (Ramezanianpour, 2014). Additionally, Eco$_2$cem should not be combined with an ammonium-containing source, as ammonia gas will then be produced (Ecocem Benelux, 2018).

Cement (C)

Cement type CEM I 52.5 R was used for the creation of control group samples. The Roman number *I* stands for the type of cement, which is Ordinary Portland Cement (OPC) in our case. *52.5* stands for the compressive strength that this cement reaches in a concrete mixture after 28 days of curing. The letter *R* stands for rapid, meaning this type of cement reaches a decent compressive strength value quickly, in approximately 2 days.

1.2 Creation of compressed samples

During this process, the Airbench was running the whole time and a FFP3 mask together with safety glasses and gloves were worn. First the DS and binder were mixed for approximately 3 min with a regular tablespoon. Then, water was added while mixing with a tablespoon. After all the water was added, an immersion blender was used to mix the blend thoroughly. This was done for approximately 3 min. 90 grams of mixture was weighed off and loaded into a mould. A workshop hydraulic press was used to compress the mix in the mould. A force of 1.2 metric tonnes was applied. The sample was retrieved from the mould and left to cure on netting to ensure that all sides of the sample were able to breathe.


Figure 4: The hydraulic press that was used for the production of compressed samples

1.3 Creation of poured samples

Before poured samples were created, moulds were made. These moulds were made with two component silicon rubber PS 8520 by Poly Service. Same amounts of the two components were poured into a mould of cardboard. After hardening for two hours, the cardboard mould could be removed and the moulds were ready for use.

Poured samples with water

The mixtures for poured samples with water were created using the same method as the mixtures needed for the compressed samples. Gravel was added to most of the poured samples, as this is a common ingredient that strengthens the samples created with the pouring process. The 3:2:1 ratio was used for every mixture unless noted differently. The 3:2:1 ratio is the ratio between gravel, sand (or DS in our case) and binder respectively. This ratio is used frequently for the creation of poured concrete.

After the mixture was ready, it was added to the moulds. A tablespoon was used to ensure the filling of the corners of the mould. After the moulds were filled, they were shaken and vibrated gently to remove air bubbles in the samples. The samples were then left to cure in the moulds until they were broken.

Poured samples with NaOH

Eco$_2$cem was mixed with 30 wt% sodium hydroxide (NaOH) and 37 wt% sodium silicate to investigate the activation potential of these alkaline solutions.

Figure 5: Samples created with a mixture of DS, gravel, NaOH, sodium silicate and Eco$_2$cem.

First, the dried dredge sediment and binder were dry mixed for approximately 3 minutes. Depending on the concentration needed, both sodium hydroxide and sodium silicate were diluted. It should be noted that when sodium hydroxide was diluted, the NaOH was added to water. This should not be done the other way around, as the reaction between H_2O and NaOH is highly exothermic and this could create a dangerous situation. Next, the sodium hydroxide was added to the sodium silicate and mixed for approximately 1 minute. This addition was done slowly to avoid extensive heating of the mixture due to the exothermic nature of the reaction. This alkaline solution was then added to the dry mixture and is mixed for approximately 5 minutes. The mixture was then added to a mould. The mould was vibrated gently for 10 seconds to remove entrapped air bubbles. The samples were left to rest for 28 days in a fumehood before testing their strength. This methodology for the creation of samples with the aid of NaOH is based on the work by Lang, Chen & Chen (2021) and Luhar & Luhar (2020).

1.4 Strength test of samples

Before testing the strength of the samples, their dimensions (length, height and depth) and weight were measured. Next, a construction to test the flexural strength was installed upon a scale. The sample was placed on the construction and pressure was applied on the middle of the sample with a metal bar. The scale showed the highest weight the samples was able to bear without breaking. This value was noted.

The value noted was divided by the height or the weight of the tile respectively. This was done because the height and weight of the tiles varied significantly between different test groups. The values retrieved after dividing by either height or weight are called the relative breaking weight (RBW). In text, these values might also be referred to as flexural strength values.

It must be noted that this flexural strength analysis is sufficient for this introductory research. These values can be compared relatively to each other, but should not be compared to values from professional flexural strength test machines. When a strong mixture is created, this should be scaled up to create a large tile that can be tested with a proper apparatus.

2. Analyses

2.1 X-Ray Diffraction (XRD)

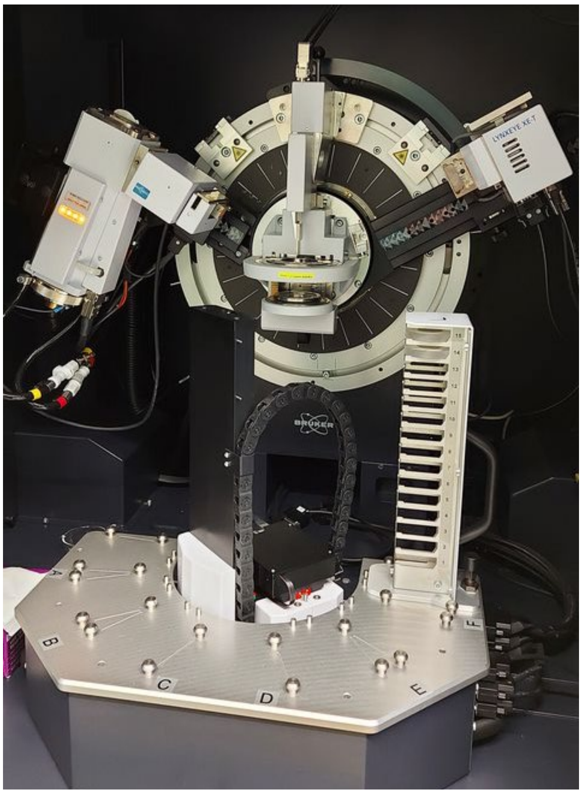


Figure 6: The Bruker D8 Advance Diffractometer that was used for our experiments.

Preparation

An X-ray diffraction (XRD) was conducted to analyse the dredged sediment from this study on its mineral content. This XRD analysis took place at the University of Utrecht at the department of Earth Sciences.

First, three samples of dredged sediment from the same batch were taken apart. These samples were filtered by sieve with a mesh size of 3 mm. After filtering, 2-propanol (isopropanol) was added to 2 grams of every sample to prevent heating because of friction during milling. Subsequently, the samples were ground in a McCrone mill by Retsch at 1500 rpm for 5 minutes. Between every milling session, the milling chamber should be cleaned with a little bit of the sample itself to avoid background measurements. After milling the sample should normally be left to air-dry for 24h. As time did not allow for this step, our samples were put in the oven for an hour at 60 °C to accelerate the drying process. As our dredged sediment was already dried at 105 °C before, this drying process at 60 °C should not influence the mineral structure significantly. About one gram is then added to the sample holder. The sample should not exceed the top of the sample holder as this would influence the measurements in a negative manner.

XRD

For the XRD analysis a Bruker D8 Advance diffractometer was used (see Figure 6). Samples were measured while applying the following settings; a 2θ range between 3 and 80° with steps of 0.02 2θ with 0.85 sec per step, 40 kV, 20mA, 17 mm sample measurement, 15 rpm rotation of sample. The X-ray source is Cu-k alpha with a wavelength of 1.54 nm. This analysis took around 55 min per sample.

The results retrieved with the diffractometer were analysed with Diffrac. Eva.

2.2 Scanning Electron Microscopy (SEM)

Preparation

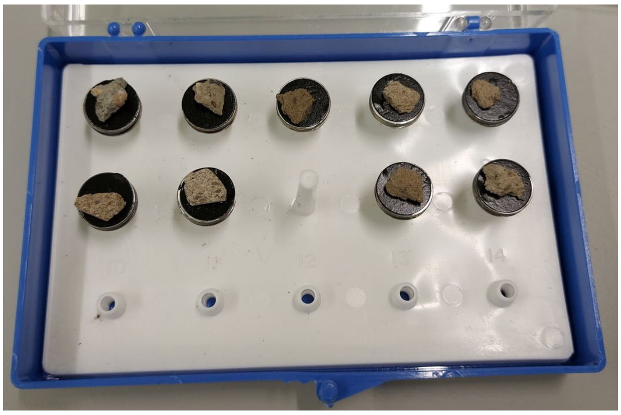


Figure 7: The uncoated samples stuck onto the stubs

First, samples that already underwent the breaking process were broken into smaller pieces. This was done by putting them in a plastic bag and subsequently hitting this bag with a rubber hammer two or three times. Next, the pieces that were smaller than 9 mm by 9 mm and as flat as possible were selected. Next, a round conductive sticker was stuck onto the necessary stubs and the samples were then stuck onto the stickers (See Figure 7). The samples were placed into a JEOL JFC2300HR sputter coater where they were coated with a conductive platinum – palladium coating of 8 nm. As our samples were fairly high in the z direction, it was important to let the samples rotate diagonally during the coating process. This guaranteed a homogeneous conductive coating in contact with the sticker, so no static electrical charging would occur during imaging of the samples by the electron beam.

SEM

For the scanning electronmicroscopy (SEM) analysis a JEOL JCM-6000 tabletop scanning electron microscope was used. The sample never exceeded the height of the sample holder, as this would negatively affect the image. The following settings were used; high-vacuum, SEI, 15 kV, standard filament current, std-probe current. The software used to operate the SEM and analyse the images is JCM-6000. It should be noted that the maximum zoom level was 2 µm. This zoom level should not be used for a long period, as it might damage the coating on the sample.

RESULTS & DISCUSSION

I have opted to add the results and discussion section together, as the large amount of results would have caused the discussion section to become very unorganized.

Sieve analysis

A sieve analysis was carried out already for the dredged sediment that would be used in all experiments in this research. This revealed that 79.4% of the dredged sediment consisted of dry material. 3.4% of this dry material consisted of organic material.

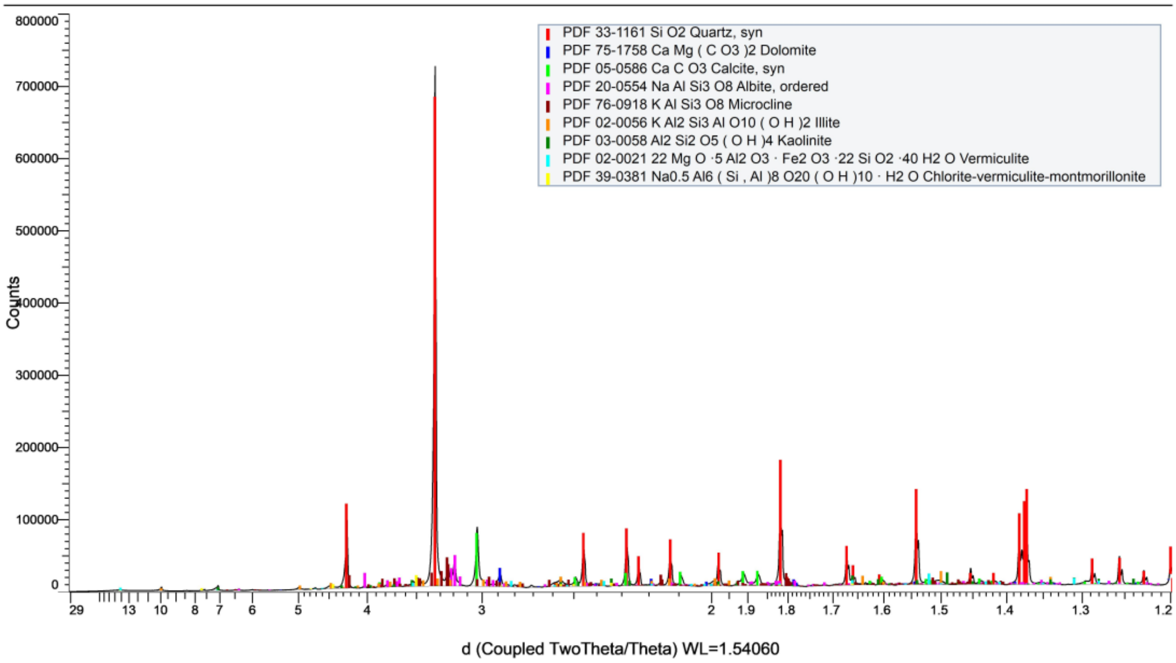
XRF of dangerous metals

An X-Ray Fluorescence (XRF) was conducted by the municipality of Rotterdam to analyse the levels of heavy metals and other possibly toxic elements within the dredged sediment. All the materials analysed did not reach levels where they could be toxic, resulting in the labelling of this dredged sediment as non-hazardous.

An XRF analysis of the oxide elements in the dredged sediment was also planned for our research; however, due to unforeseen circumstances this analysis could not be conducted and should be done in future research for Waterweg.

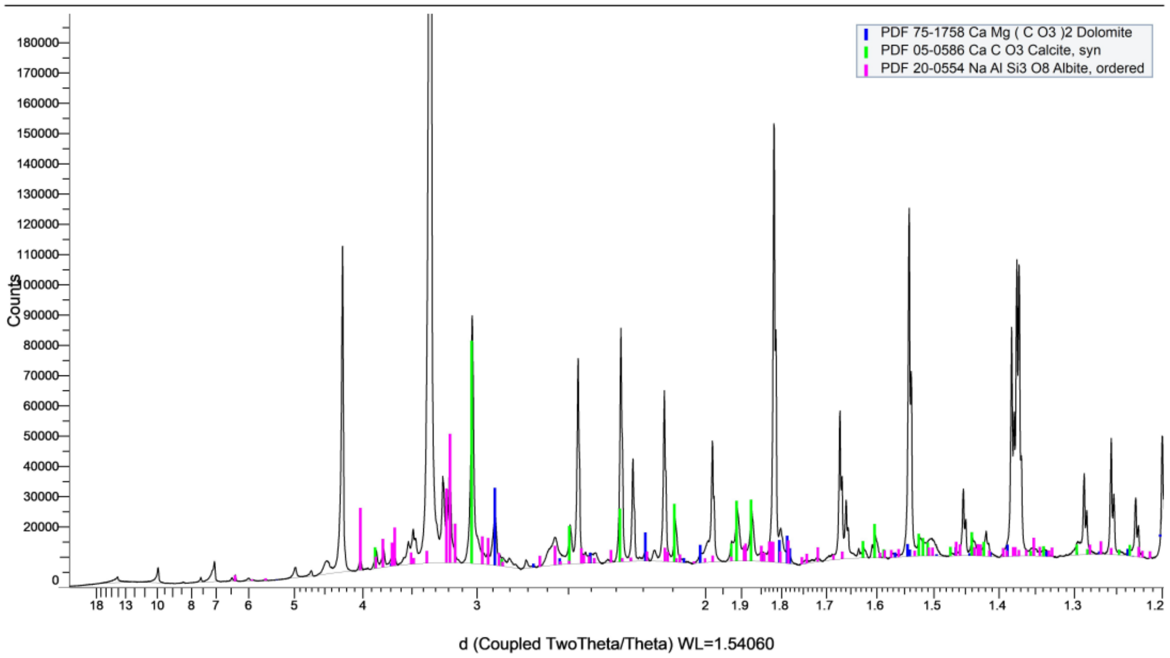
XRD

An X-Ray Diffraction (XRD) analysis was conducted to analyse the mineral composition of the dredged sediment, as well as the variance within the dredged sediment on a mineral level.



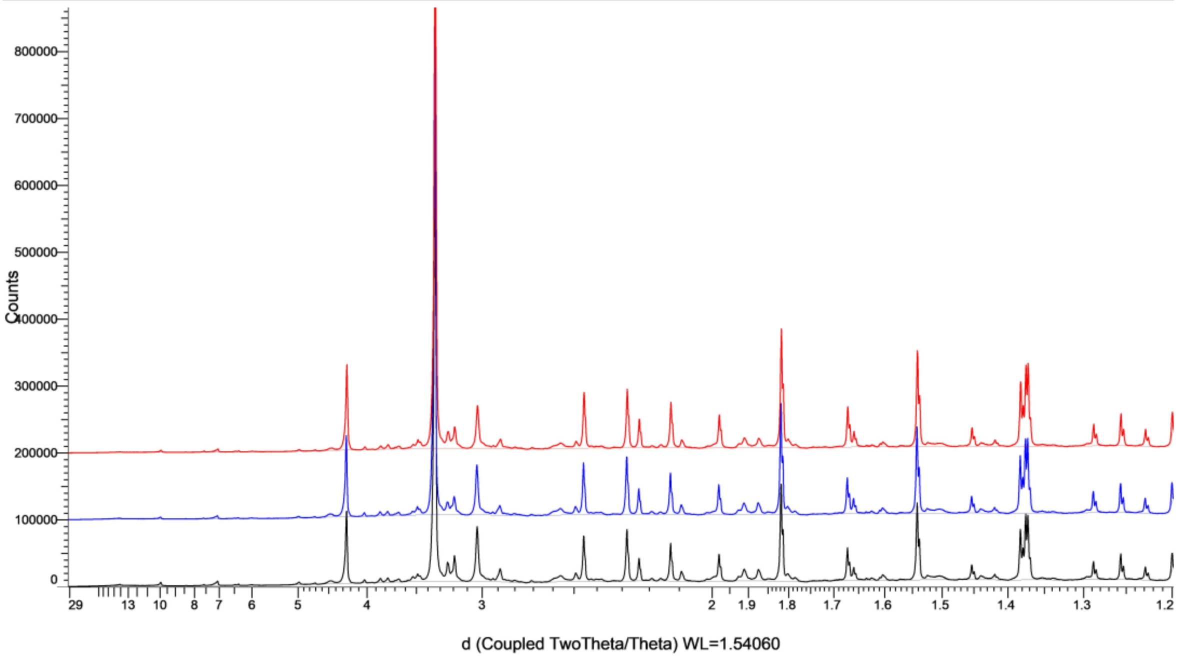
Graph 1: XRD analysis of the dredged sediment with all plausible minerals indicated

Graph 1 shows the full XRD profile of the dredged sediment used. The quartz peaks align properly with the quartz lines from the library, as well as the calcite peaks to a lesser extent. As can be seen in Graph 2, the other peaks hint at the presence of dolomite and albite.



Graph 2: XRD analysis of the dredged sediment with dolomite, calcite and albite indicated

Between 18 and ~ 3.4 d clay minerals are observed within the XRD graph. Unfortunately, this part of the graphs was difficult to analyse as the peaks seemed to have shifted relative to the values in the XRD database. It could be that the clay minerals in the DS have different chemical compositions, with more iron or aluminium for example, than the standards in the XRD library. The peak shifts could also be due to the drying of dredged sediment at 105 ^{o}C as this process may alter the minerals within the DS because of the removal of water from their structure (as reviewed by Ramezanianpour, 2014). If the DS was not completely dried out, the water that stayed within the clay minerals can also cause a shift or elongation of the peaks. The one conclusion that can be drawn from Graph 2 is that a mixture of clay minerals is present. Additional research should be conducted to analyse the exact clay composition.



Graph 3: XRD analysis of three samples of the same dredged sediment without any minerals indicated

Graph 3 shows the XRD profile of three different samples from the same batch of dredged sediment. The three profiles show similar peaks at similar d-values. This is an indication that there is not a large variance in mineral composition throughout this batch of dredged sediment. However, there are some differences between the profiles in terms of peak intensity, for example around d = 2.9 nm.

Compressed samples

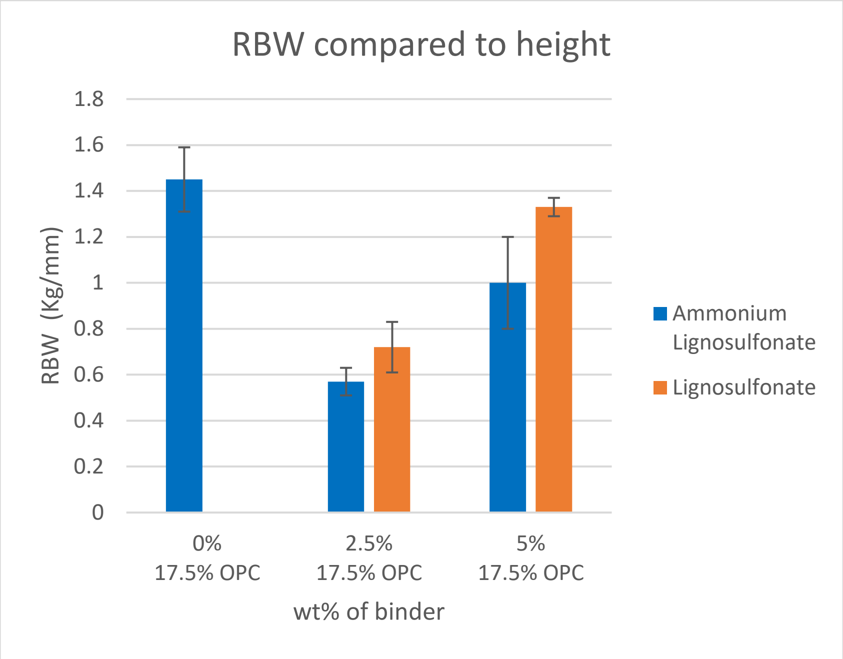
The compression production method was used first for the creation of samples.

All binders in this research underwent the same procedure. First, pressed samples were made in which OPC was gradually replaced by the alternative binder. This experiment showed to which extent the new binder had the capability of functioning as an independent binder. After this research, samples were created with different ratios of water to binder to investigate the ideal recipe for these mixtures. Finally, with the optimized mixture from the previous experiment, samples were created and stored for different durations to investigate the curing time of this binder. The results are ordered per binder below.

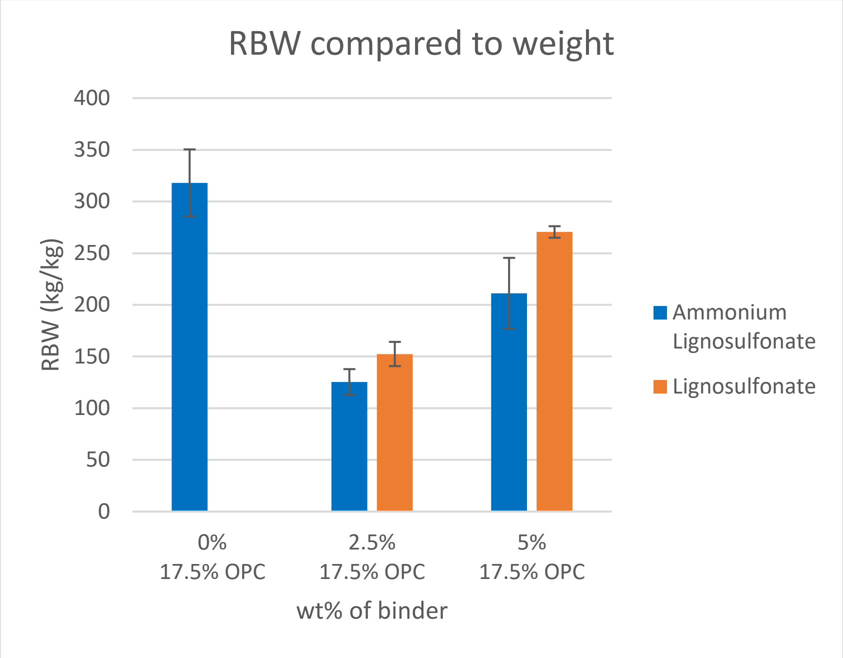
Samples with OPC are used as a control group as well as a benchmark for the new binders.

Lignosulfonate

The first binders that were tested were the two different types of lignosulfonate. The flexural strength results of these binders are presented in Graph 4 and 5 with RBW standing for relative breaking weight. This breaking weight is either relative to the height or the weight of the tile.



Graph 4: Relative breaking weight compared to height of cement tiles with the addition of either ammonium lignosulfonate or lignosulfonate. The 0% group is the control group. The black bars represent the standard deviation.



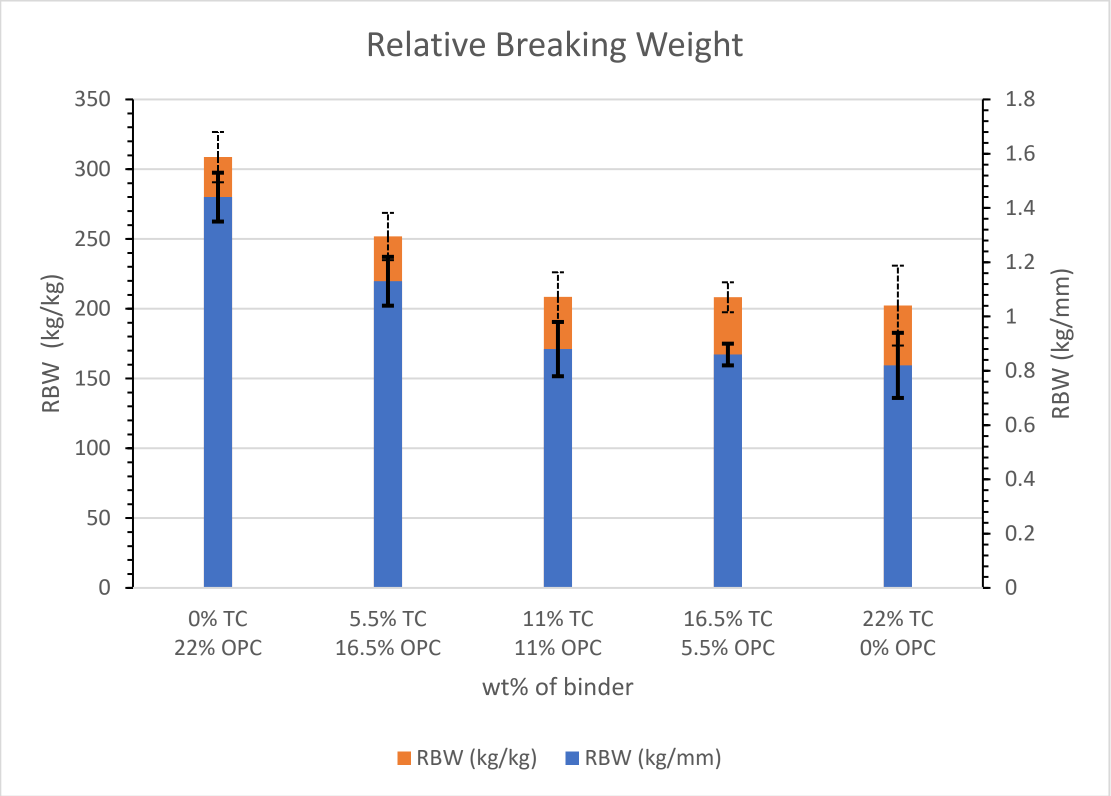
Graph 5: Relative breaking weight of cement tiles with the addition of either ammonium lignosulfonate or lignosulfonate

These were the first experiments conducted and it was decided that no more tests would be started with lignosulfonate after this. The results in Graphs 4 and 5 seem promising but it should be noted that these samples still contained 17.5 wt% of OPC. Secondly, only three samples were created per test group with the idea that these experiments had a very introductory nature. This makes the possible variance within the test groups larger, especially since some samples did not survive the compression process, resulting in an even smaller test group. Five samples were created for any subsequent test group after these lignosulfonate experiments. One thing that was noted after the experiment is that a mix of lignosulfonate, OPC, DS and a high percentage of water had hardened in a cup to form a very strong cylinder. For this reason, tests were carried out to investigate the utilization of lignosulfonate in poured samples. This research will be addressed below.

Topcrete

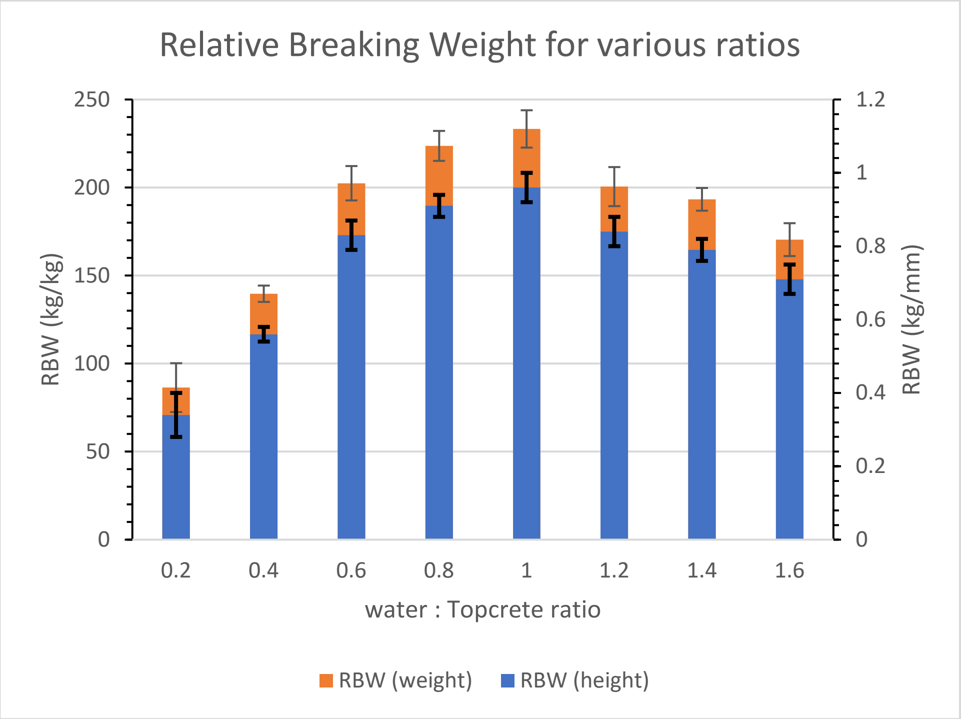
Next, Topcrete was tested as an alternative binder. First, samples were created with an increasing wt% of Topcrete and a decreasing wt% of OPC (see Graph 6). The OPC wt% decreased by the same amount as the amount by which the TC wt% increased.

The flexural strength of the samples decreases with an increasing concentration of TC and a decreasing concentration of OPC. However, the flexural strength seems to reach a plateau at 11 wt% TC (ratio TC : OPC is then 1 : 1). Based on these results, it was decided that the next experiments would be carried out with a full replacement of OPC by TC (22 wt% TC).



Graph 6: Relative breaking weight of samples created with different weight percentages of Topcrete

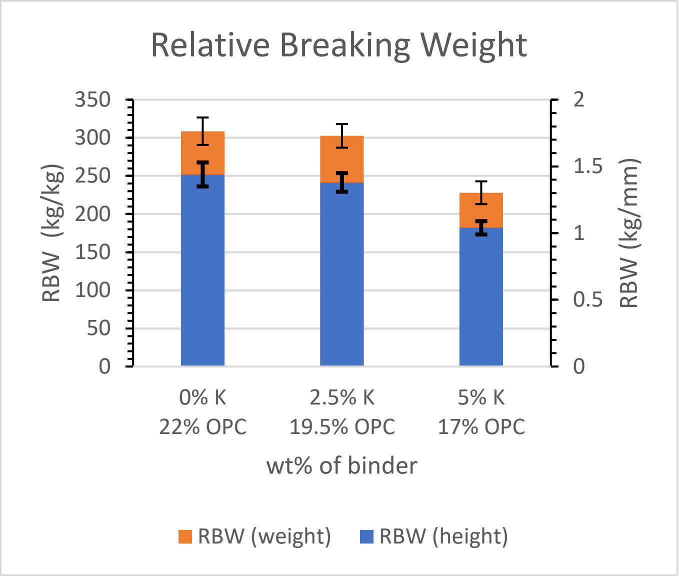
Next, the ideal ratio between TC and water was investigated (see Graph 7). The limits of this graph (ratio of 0.2 and 1.6) were chosen because outside of these limits no samples could be created either because of a lack of water below a ratio of 0.2 or an excess of water above a ratio of 1.6. These results clearly show that a ratio of TC to water of 1 : 1 gives the highest flexural strength for the production of samples through compression. This ratio was then used to create samples that were left to cure for respectively 1, 2, 3 and 4 weeks before breaking them.



Graph 7: Relative breaking weight of samples created with different water : Topcrete ratios.

Kaumera

Kaumera was also tested as a possible replacement of cement for dredge-based tiles. First, cement was gradually replaced by Kaumera. The results can be seen in Graph 9.



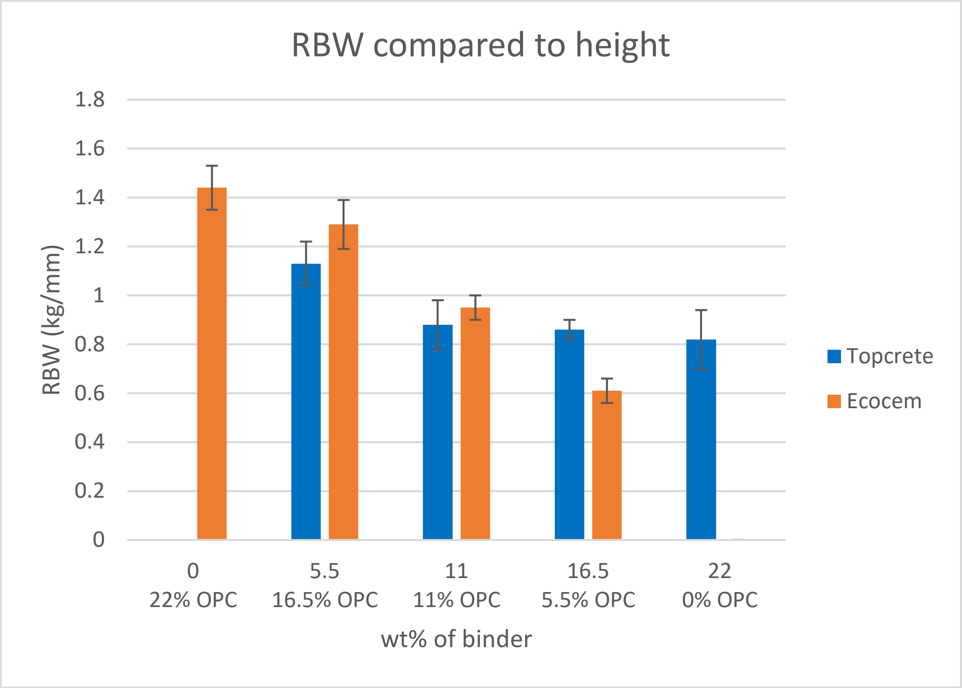
Graph 9: Relative breaking weight of samples created with different weight percentages of Kaumera (K) and OPC

These results are not very promising unfortunately. A maximum of 5 wt% of Kaumera could be used to replace OPC, which was still present for 17 wt%. A higher replacement of OPC by Kaumera was not possible, as the mixture could not be compressed. The RBW values reached for 5 wt% of Kaumera are very comparable with the samples of TC, where cement was fully replaced.

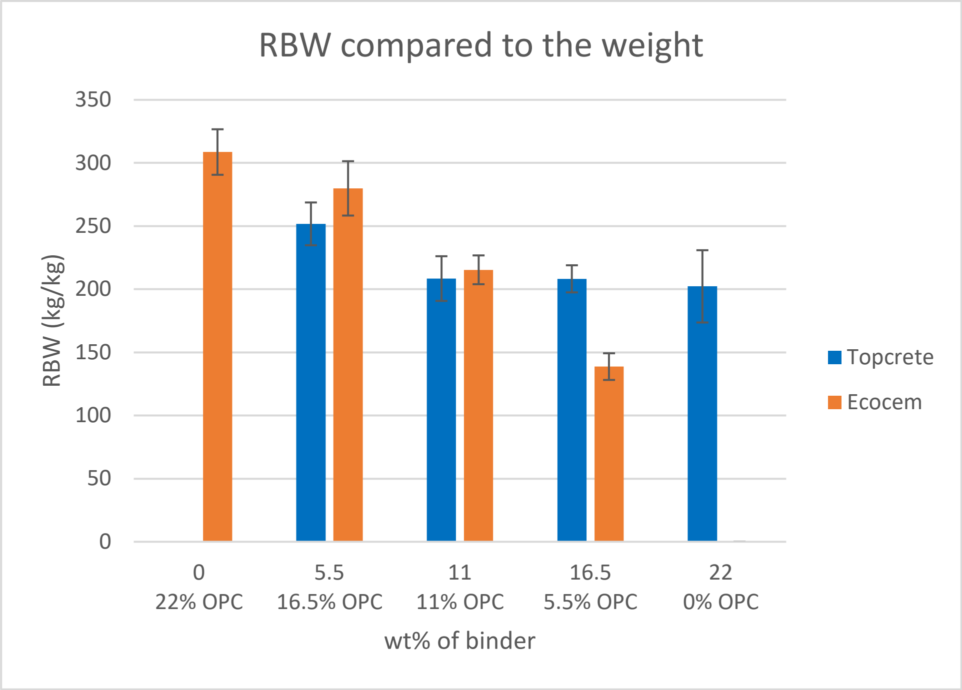
Based on these unpromising results, the decision was made to terminate the experiments with Kaumera for the duration of this research.

Eco$_2$cem

Graphs 10 and 11 show the results of the experiment where OPC was gradually replaced by EC, alongside the previously discussed results of TC. As mentioned before, Eco$_2$cem needs an activator to work as a binder. OPC or an alkaline solution are two possible activators. For this reason, a full replacement of OPC by EC is not possible, which can be seen in Graphs 10 and 11 at the 22 wt% of binder. The highest possible replacement of OPC by EC was chosen, because this would achieve the best results from a sustainability standpoint. The concentrations of EC and OPC respectively in this mix are 5.5 wt% and 16.5 wt%. With this mixture, the ideal ratio between water and EC together with OPC was analysed.

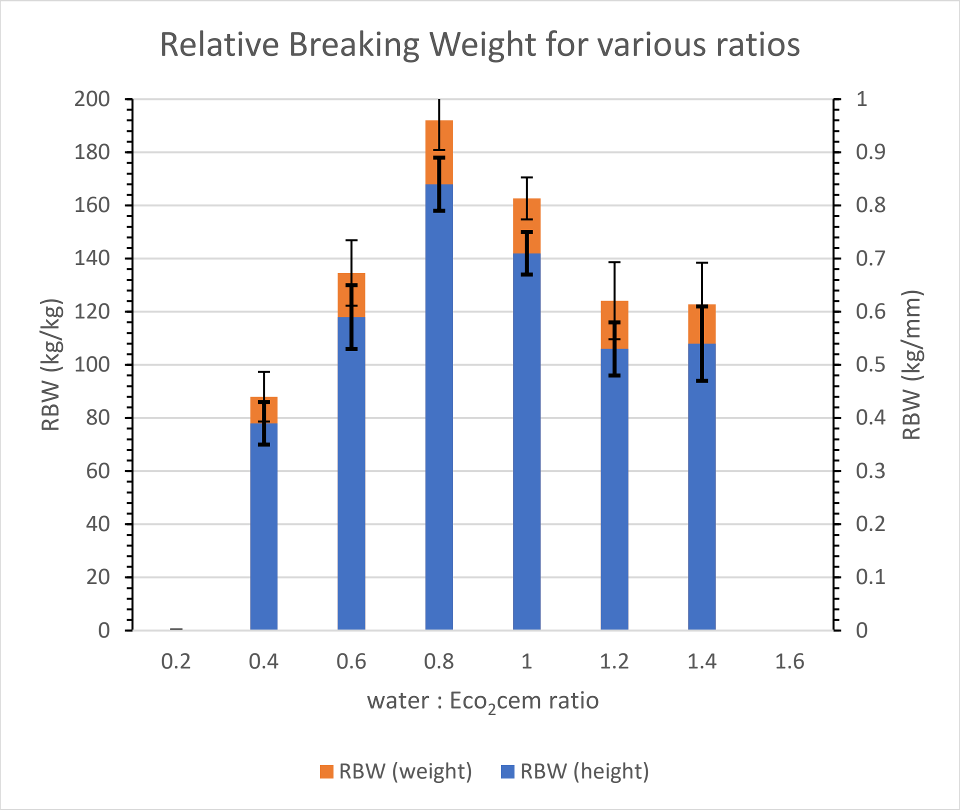


Graph 10: Relative breaking weight compared to the height of samples created with different weight percentages of Topcrete or Eco$_2$cem. The 0 wt% group represents the control group.



Graph 11: Relative breaking weight compared to the weight of samples created with different weight percentages of Topcrete or Eco$_2$cem. The 0 wt% group represents the control group.

As can be seen in Graph 12, the ratio of water to EC of 0.8 shows a significantly higher flexural strength. This ratio, alongside the weight percentages used for EC and OPC form the optimized mixture for this binder in a compression process. For Eco$_2$cem, no samples were stored for longer than one week, as the results for similar experiments with OPC and TC showed that either our storage conditions or the production process do not allow for the creation of useful results. Eco$_2$cem was then tested in a pouring process.



Graph 12: Relative breaking weight of samples created with different water : Eco$_2$cem ratios. The OPC wt% in these samples is set at 5.5.

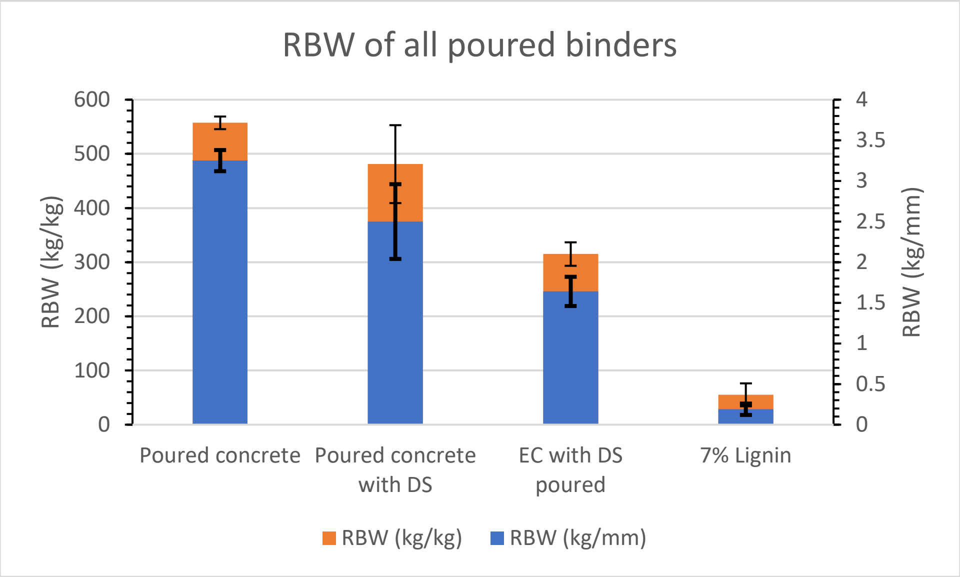
Poured samples

The idea of pouring samples was brought up after it was noticed that the compression process did not deliver the wanted results with the new binders.

Ordinary Portland Cement

Poured concrete samples were created following the 3:2:1 ratio with gravel, ordinary sand and OPC, after which the samples were left to rest for 7 days. The flexural strength results are shown in Graph 13 alongside the results of the other poured samples. The values that are achieved by poured concrete are the highest of any of the samples, either poured or compressed. This was to be expected, as the flexural strength of poured concrete is the benchmark for any materials that are to be used for street tiles in the Netherlands. These retrieved values for the poured concrete samples will also serve as a benchmark within our studies.

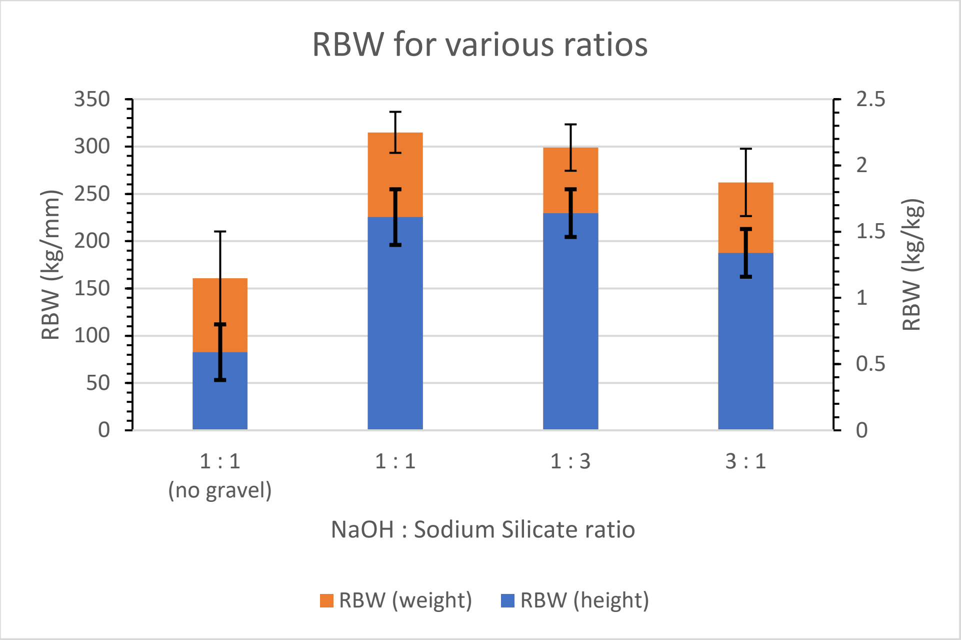
Next, samples were created with the same ratio as poured concrete, but with DS replacing ordinary sand. As can be seen in Graph 13, these samples showed good flexural strength, with a rather large standard deviation. This is due to the fact that there were samples that reached flexural strength values similar to the values of poured concrete, but there were also samples that reached lower values.



Graph 13: Relative breaking weight of all poured binders

Eco$_2$cem

The last samples shown in Graph 13 are poured samples containing Eco$_2$cem and a mixture of sodium hydroxide and sodium silicate. These samples were first created without gravel. it was noticed during production of these samples that the consistency of their mixtures was very different from the previously produced mixtures with gravel. For this reason, gravel was added with later test groups and it was noticed that less solution was needed to make the mixture ready for pouring. The flexural strength analysis revealed that the samples containing gravel were more than twice as strong as the samples without gravel (See Graph 15). For the three test groups containing gravel, the ratio of sodium hydroxide to sodium silicate was analysed. There was no difference between the 1:1 and 1:3 NaOH to sodium silicate ratios, but the 3:1 ratio was slightly weaker, although not significantly. The 1:3 ratio is displayed in Graph 13.



Graph 15: Relative breaking weight for various ratios of NaOH to sodium silicate.

Lignosulfonate

A brief test was carried out with the utilization of lignosulfonate as a binder in poured samples, but this was no success. The RBW compared to height was 0.19 ± 0.07 and the RBW compared to weight was 55.09 ± 21.19 (See Graph 13). These values were not promising enough to warrant a continuation with this binder.

Topcrete

Poured samples were also created with Topcrete using the 3:2:1 ratio and water as an activator. These samples turned out to be very weak, as they broke before the application of any pressure.

SEM

Scanning Electron Microscopy was used to analyse the produced samples based on the character of their surface. This research was conducted halfway through the production process, so not all samples discussed above could be analysed by SEM. Figures 8 A to E show the presence of needle-like mineral structures within the poured concrete and to a lesser extent also in the poured concrete with DS. These needles are not observed on the surface of compressed samples (See Figures 8 C to E) and might explain the higher flexural strength of poured samples. The compression process might inhibit the formation of these needles. Also, the inclusion of DS instead of ordinary sand seems to influence the needle formation. Crystal growth can be influenced readily by different additions so DS might play an important role in this. The presence of water could also play a role in crystal formation. In compressed samples, less water is present, which might influence the ability to form certain crystals. The lack of water was also noticed during the experiments where samples were left to cure for longer periods. These samples seemed to dry out and subsequently showed lower flexural strength compared to samples that were left to cure for shorter periods.

Additionally, it seems like the poured concrete has a larger particle size distribution compared to the poured concrete with DS. A sieve analysis could clarify this suspicion.

Next, Topcrete samples seem to consist of loose sheets of crystals, whereas this phenomenon is less obvious for the Eco$_2$cem samples.

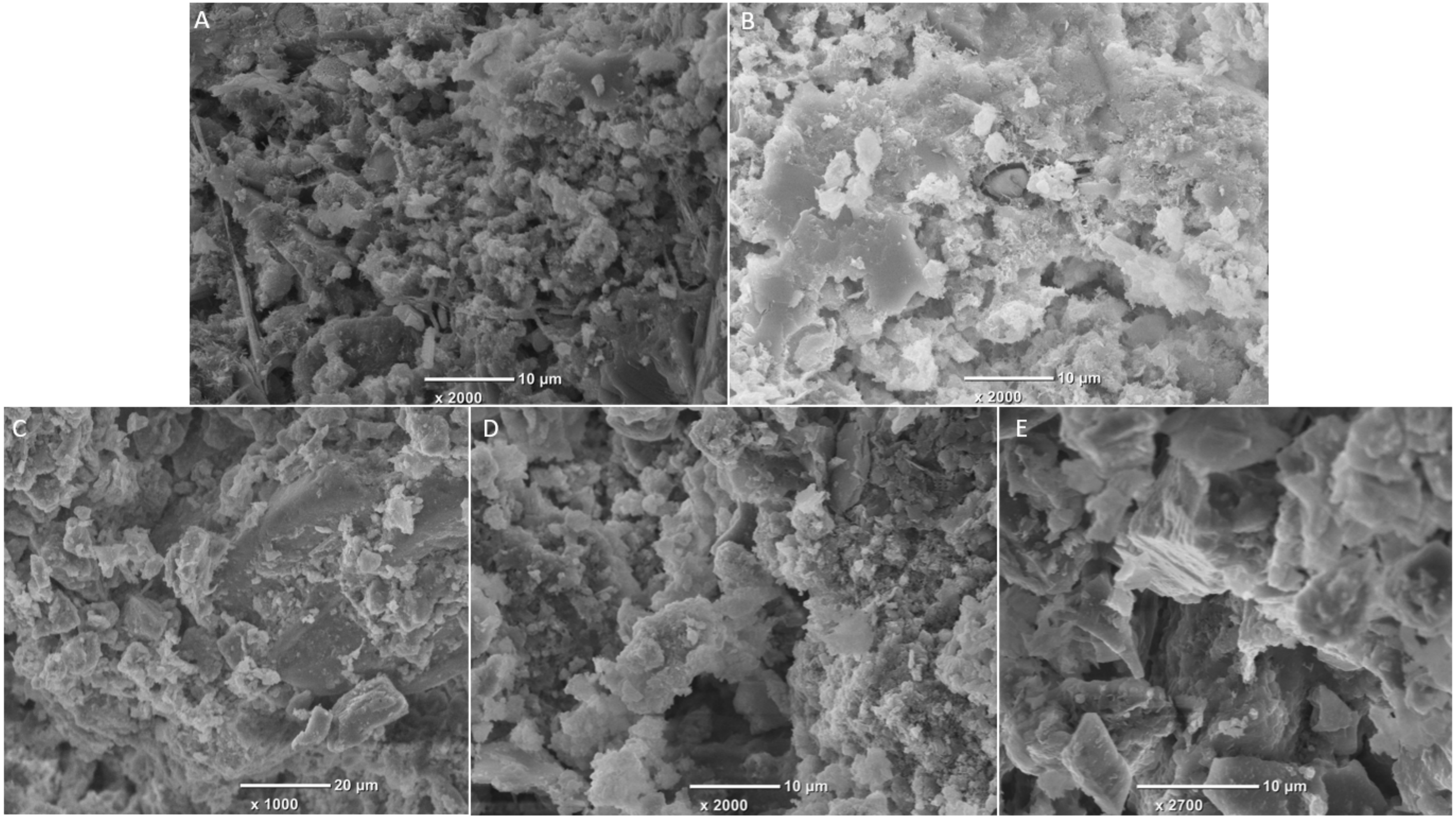
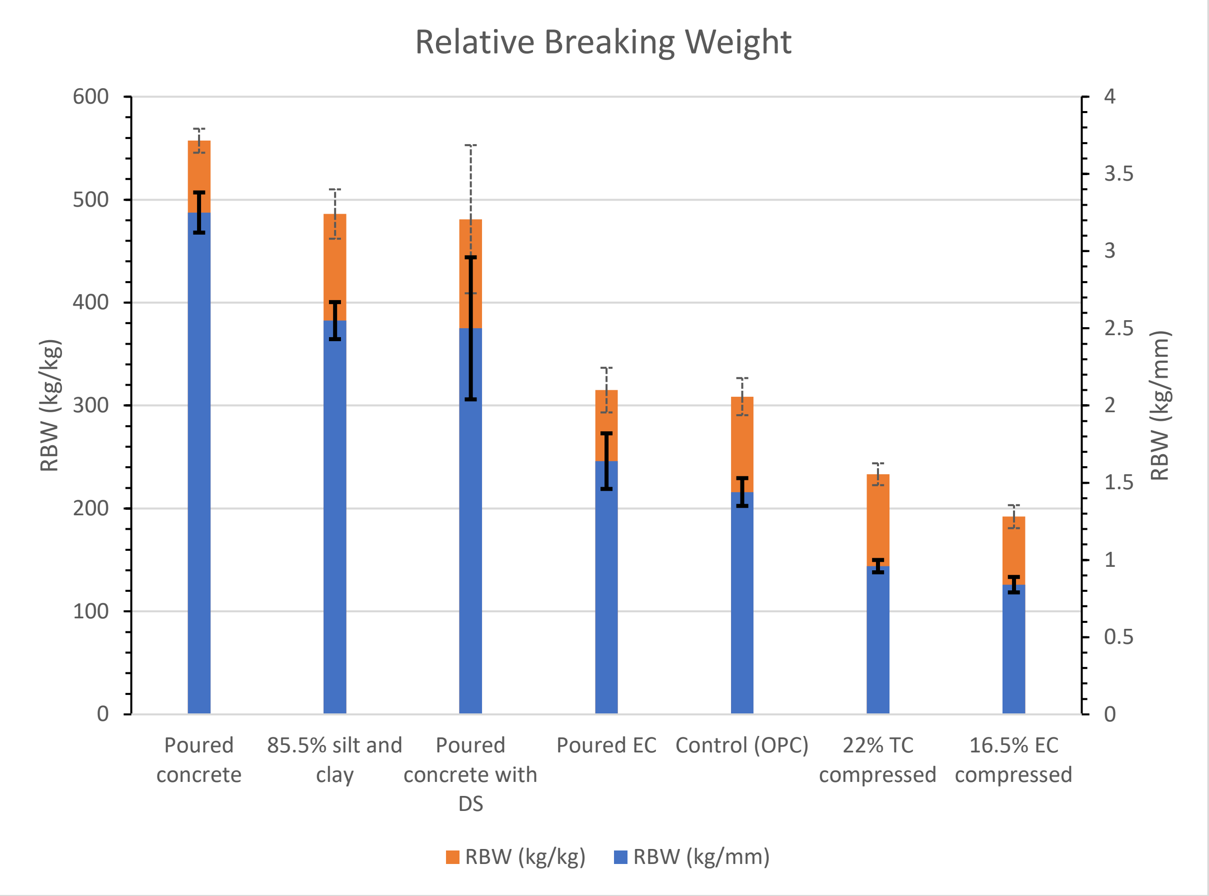


Figure 8: A) SEM image of a poured concrete sample, at 2000x magnification. B) SEM image of a sample made of poured concrete with dredged sediment instead of sand, at 2000x magnification. C) SEM image of a compressed sample created with OPC, at 1000x magnification. D) SEM image of a compressed sample created with Eco$_2$cem, at 2000x magnification. E) SEM image of a compressed sample created with Topcrete, at 2700x magnification.

Comparison of promising samples

Graph 16 shows the results from every promising binder, either poured or compressed. The test group with 85.5% silt and clay are compressed samples created by Bas van Middelkoop, a different intern at Waterweg (van Middelkoop, 2021). The values of this test group are very impressive, but it has proven difficult to find a significant amount of dredged sediment with this concentration of silt and clay. Additionally, the poured Eco$_2$cem is just as strong as this control group. This is good news, as this means that the same flexural strength can be achieved for this sustainable binder as for the control group which still contains OPC. Additionally, the pouring process is not optimized yet. Optimalization will most certainly increase the flexural strength of these Eco$_2$cem-based samples.

Additionally, the poured concrete with DS shows flexural strength values almost similar to poured concrete, which should warrant more investigation into the pouring method of production. The results in Graph 16 also suggest that Topcrete and Eco$_2$cem might not suffice as binders for compressed samples.



Graph 16: Relative breaking weight for every promising binder.

CONCLUSIONS

In this research, a large amount of knowledge has been gathered on dredged sediment and a selected group of binders:

- An **XRD analysis** revealed the main minerals present in the dredged sediment that was used, were quartz, calcite and probably dolomite and albite. It was impossible to distinguish the clay minerals present as the dredged sediment had been heated at 105 °C to remove water.
- Mixtures of **dredged sediment**, OPC, gravel and water were poured and the resulting samples showed relative breaking weight values that are close to the values of poured concrete, which is the benchmark for all the samples created in our research.
- **Lignosulfonate** did not suffice as a sole replacement for Ordinary Portland Cement in both the compressing method and the pouring method. A maximum of 5 wt% of lignosulfonate could be added to the samples in the compressing method, while still adding 17.5 wt% of OPC. However, lignosulfonate can play a role as an additive as it reportedly improves certain properties of the end product made with cement (Arel, 2017). This was not analysed in our research.
- **Kaumera** did not suffice as a sole replacement for OPC in either production method. It could be used as an additive, but it is less promising than lignosulfonate. It has only been proven to have a slowing effect on the drying of cementitious mixtures, which could be useful in warm climates. Unfortunately, Kaumera might increase the microbial activity within the created tiles, which is not wanted due to the organic matter that is already present within dredged sediment.
- **Topcrete** is the only binder that could fully replace OPC in the compressing method of production. The poured samples containing Topcrete and water turned out to be very weak. Topcrete seems like a sustainable binder for the medium to long term, as it is a waste stream that is created at the end of the paper recycling industry.
- Around 75 wt% of OPC can be replaced by **Eco$_2$cem** in the compression method of production. These samples with 75 wt% of Eco$_2$cem and 25 wt% of OPC have a flexural strength that is similar to samples created with Topcrete as a sole replacement. Mixtures of Eco$_2$cem, sodium hydroxide, sodium silicate, gravel and dredged sediment showed flexural strength values similar to values of the control group.
- **Fly ash** could not be tested as a replacement of cement as it could not be handled safely in the lab of Waterweg due to lead pollution in the fly ash that was delivered. Fly ash is a promising alternative to OPC in the short to medium term, because of the relatively large amount of research that has gone into this binder compared to other alternatives for OPC. The sustainability of fly ash as a binder is hard to gauge due to the process in which fly ash is produced as a waste stream; coal incineration. As countries are scaling down coal incineration for energy production, fly ash might also become more scarce in the future.

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