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Review paper

Durability, carbon footprint and contaminant immobilization in self-hardening slurries applied to cut-off walls: a review

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Abstract: Cut-off walls built using self-hardening slurries are an important tool for modern engineering pursuing Sustainable Development Goals. Much like cement concrete, this material is affected by the challenges posed by the increasing human pressure on the environment, although it is used significantly less widely than concrete; for this reason, relatively little comprehensive literature data is available describing the interaction of self-hardening slurries with the environment. This article provides a review that complements the current state of knowledge on self-hardening slurries in this area, with a particular focus on the durability of the material and its pollutant immobilization capabilities. To provide context, the material's operating conditions, properties and components are briefly characterized. The resistance of self-hardening slurries to environmental aggression is described extensively, as it is a key factor in ensuring the durability of the material. A sample analysis of the material's carbon footprint in several representative composition variants is presented. The subject of pollutant immobilization by self-hardening slurries is outlined. Lines of further research are proposed to fill gaps in the available knowledge.

Keywords: carbon footprint, cement-bentonite, durability, immobilization, self-hardening slurry, supplementary cementitious materials

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1. Introduction

The development of civilization and its consequences are generating a significant demand for various types of environmental engineering structures, including cut-off walls. The desire to develop a steadily increasing land area requires ever expanding flood protection, which is further amplified by climate change [1, 2]; the increase in demand for water, energy and the drive to reduce greenhouse gas emissions encourage the construction (and renovation) of reservoirs and dams [3, 4]; finally, the growth of industrial output and the disposal of large amounts of waste create a high risk of pollutant emissions, which needs to be addressed [5]. Cut-off walls are used in each of the above-mentioned areas [6, 7] – they are vertical barriers built in the ground in order to limit the horizontal groundwater filtration flow, including that of contaminated water [8]. At the same time, they do not follow the same structural requirements as e.g. underground parts of buildings. As a result, this field often employs specialized materials, including self-hardening slurries [9], the subject of this article.

Self-hardening slurry is a mixture of water, binder (usually cement) and clay material (usually bentonite) with the optional addition of other components. It is used in engineering, apart from cut-off walls, for the construction of foundations and the filling of cracks or holes in the ground. The fundamental characteristics of this material are its predominant water content, thixotropic properties and hardening ability [10–14]. Hardened slurries exhibit special properties, their microstructure can be described as resembling a fibrous sponge medium capable of enclosing a substantial amount of water and, at the same time, capable of transmitting stresses [15, 16].

Self-hardening slurries, like any building material (especially cement-based), carry an unavoidable carbon footprint. A ‘carbon footprint’ is difficult to define, as it requires a clear statement of underlying assumptions and often, the methodological approach. A widely accepted and concrete definition of a carbon footprint does not exist [17]. Carbon footprint is a subset of all Life-Cycle Assessments (LCA) and is generally based on long-lived greenhouse gases using a 100-year global warming potential (GWP) as specified in the Kyoto Protocol. LCA distinguishes four stages in the analysed system, each of them consisting of several modules, and benefits or loads reaching beyond the system boundary (Fig. 1).

Conceptually, a carbon footprint should consider all emissions of a product both backward in time from the point of use to emission sources and forward in time to include the use and disposal stage of products [19]. The concept of carbon footprint can be perceived at many levels – product, economic sector, corporation, country, etc.; from the point of view of the carbon footprint of the construction industry, it will be interesting to look at the product level – the carbon footprint of a product (CFP).

The definition of the CFP can be found, inter alia, in the standard [20]. The CFP is the sum of greenhouse gas emissions and removals in a product system, expressed as carbon dioxide equivalents (a measure explained e.g. in [21]) and based on a life cycle assessment (compilation and evaluation of the inputs, outputs and the potential environmental impacts of a product system throughout its life cycle) using the single

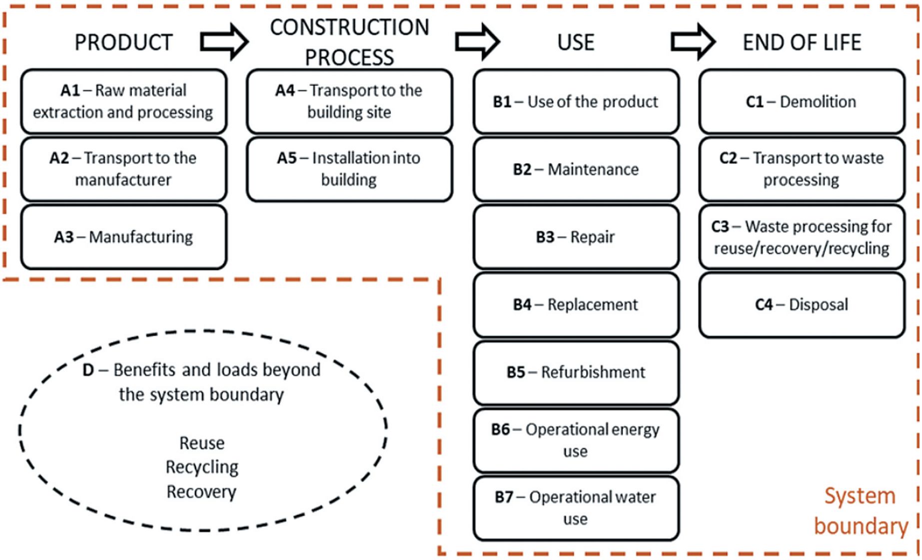

Fig. 1. Four stages and the respective modules (A-D) of LCA in construction works [18]

impact category of climate change. A concept similar to CFP is embodied carbon (EC), the sum of fuel-related and process-related carbon emissions (which can be measured from *cradle-to-gate*). This includes all activities from material extraction (quarrying/mining), manufacturing, transportation and right through to fabrication processes until the product is ready to leave the final factory gate [22] (modules A1–A3).

Emissions from the production and transport of construction materials and the construction process account for 10% of total global emissions [23]. Detailed data on the emissivity of individual construction processes is limited. Still, it is estimated that around 50% of construction-related emissions are attributable to the building material production stage [24] (*cradle-to-gate* according to the Life Cycle Analysis – LCA – nomenclature). Embodied carbon is exceptionally high in cement, which accounts for c.a. 6–8% of annual anthropogenic GHG emissions (the chemical reaction involved in the production of clinker ~4–5% [25, 26], the rest from heating the raw ingredients [27]). Global material use is also expected to double by 2060, and the materials used in the building and construction sector will comprise a third of this rise. At the same time, some analyses show that it is possible to reduce the CF of construction works without increasing their cost [28]. For example, to limit the emissions from the production of cement one can partially replace it with so-called supplementary cementitious materials, mostly industrial by-products [29]. They, however, carry the risk of pollutant emissions to soil and ground water [30, 31], thus impacting the environment in a different way. Another indirect method of reducing emissions is to extend the working life period by ensuring high durability of the structure. This may require greater initial emissions, but will cut their overall amount by limiting necessary repairs, replacements etc. Chemical aggressiveness is a particular threat to the durability of cut-off walls built around landfills [32].

Self-hardening slurries are an essential tool for environmental engineering due to their areas of application (outlined at the beginning) and the importance thereof in achieving the Sustainable Development Goals [33]. The purpose of this article is to complement the current state of knowledge on the use of the material in cut-off walls, relatively to the currently available review articles [8, 9, 34], and to propose lines of further research in response to contemporary challenges. This article supplements the available literature with a review of the durability of slurries under chemical aggressiveness of the water-soil environment, and the pollutant immobilization capacity of slurries. This represents a certain novelty in comparison with the available publications, and at the same time a key element of the Life Cycle Analysis in the post-production, i.e., use and end-of-life phases. Addressing the challenges brought about by climate change and the need to achieve Sustainable Development Goals, a carbon footprint analysis (using the EFFC DFI Carbon Calculator Tool v4.0 [35]) is presented for several representative self-hardening slurry compositions. It highlights the ecological significance of using by-product or waste ingredients in the slurries, combined with the reduction of high-carbon cement (clinker). In the authors' opinion, it is one of the future directions for the development of self-hardening slurry cut-off walls technology, aimed at reducing GHG emissions.

2. Self-hardening slurry

2.1. Working conditions

A comprehensive state of the art of self-hardening slurries is presented, for example, in [8, 34], including the composition of slurries, how the components are selected, and how it affects material properties; this article will not discuss these extensively. Based on their composition several types of slurries are distinguished, most commonly [9, 36]: cement-bentonite (CB), cement-bentonite-slag (SCB) and cement-bentonite-ash (FCB).

The material of a cut-off wall in its working environment is exposed to the following factors, which can adversely affect the durability of the structure [8, 36]):

- Settlement and deformation of embankments and subsoil – under self-weight, the pressure of water (potentially leading to bending, Fig. 2b), the pressure of soil, and friction at the material-soil boundary associated with settlements, (potentially causing hanging of the wall and its stretching) [36];
- Internal erosion – differential pressure acting on the sides of the wall and water filtration can carry away particles and cause hydraulic puncture (piping) or cracking, Fig. 2c [11];
- Changes in ambient humidity – initial moisture content of the hardened slurry reaches up to 100–200%, but water is gradually lost through contact with dry soil [36, 41];
- Freezing and thawing – depending on weather conditions [36, 41];
- Chemical corrosion – particularly affecting the material of the cut-offs which are in contact with contaminated soil and ground water around landfills (Fig. 2a) [42];
- Biological corrosion – in essence, mechanical or chemical interactions, e.g., plant roots growth, animals digging corridors, products of microbial metabolism [36].

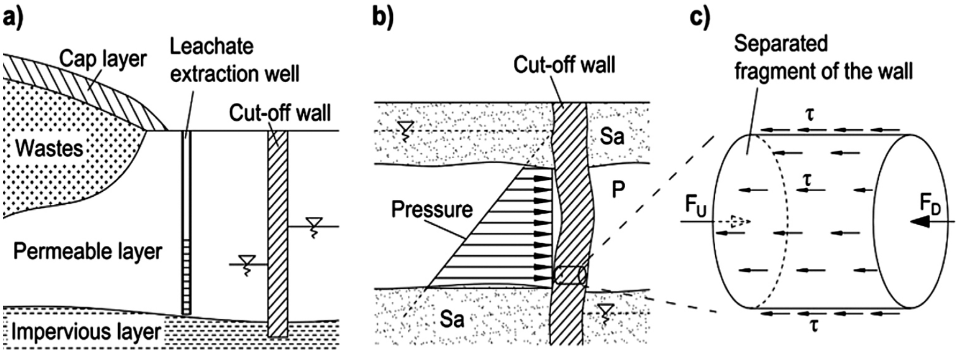

Fig. 2. Working conditions of a cut-off wall: a) around a landfill; b) in soil layers of different compressibility, subjected to bending (hydrostatic pressure in the P layer shown); c) an isolated fragment subjected to uneven pressure (for piping analysis). Sa – low compressibility layer (e.g., sand), P – highly compressible layer (e.g., peat), F_U – upstream hydrostatic pressure, F_D – downstream hydrostatic pressure, τ – tangential stresses on the lateral surface. Based on: [8, 11, 36]

A cut-off wall using the slurry can be made inside an excavated trench, following the single-phase or the two-phase method. The transient working conditions of the material in liquid state should be recognized, distinguishing between the two above-mentioned methods [9, 36].

In the single-phase method, the liquid self-hardening slurry stabilizes the excavated trench and seals its sides (e.g., as a result of soil pore clogging). When the digging is finished, the slurry is left to harden. As a result of unintended continuous mixing of the excavated material with the slurry during excavation, part of the soil is incorporated into the diaphragm, while part of the slurry enters the soil [15]. Moreover, the slurry may already be affected by contaminants present in the subsoil or ground water during the excavation [11].

In the two-phase method, a supporting slurry (usually bentonite-water) is used during the digging, and later replaced with a self-hardening slurry. This ensures greater homogeneity of the wall and reduces the presence of soil particles and contaminants entering the material. In addition, it is also possible to produce a self-hardening slurry in the excavation by adding cement slurry to the supporting slurry.

2.2. Properties

The technological properties of a slurry are tested by methods analogous to those used in drilling [43], while the performance properties are usually determined by methods derived from geotechnical engineering and concrete technology [36]. An extensive description of slurry properties can be found in [8, 9, 34], among others.

Technological properties:

– Bulk density is essential for maintaining the narrow excavation stability and for the slurry's displacement by the target material (two-phase embedment) [36].

- Viscosity is essential for the production and transport of slurry (e.g. pumping), the ease of digging under its cover, and the displacement of slurry [8, 36].
- Water bleed is a measure of slurry sedimentation, homogeneity and stability. Segregation can lead to local differences in composition and properties of the wall material [44].
- Structural (gel) strength of a slurry (shear strength in liquid state) characterises its thixotropic properties [45]. It is responsible for the stability of the excavation sides, prevents the detachment of soil grains and stabilizes the soil-contaminated slurry [36].

Performance properties:

- Shear strength (angle of internal friction and cohesion). Influences the resistance to piping (cf. Fig. 2c)) and suffusion, tightness and mechanical strength [11, 46].
- Compressive strength – it is recommended to design the slurry so that it is mechanically similar to the surrounding soil medium [47]. A detailed review can be found in [9].
- Brittleness index (the ratio of the tensile strength to the compressive strength) – tensile strength helps maintain the continuity of a cut-off wall in the ground [12], e.g. when the lower part of the cut-off wall settles more than the upper part or bending occurs (Fig. 2b) [11, 36].
- Hydraulic conductivity (filtration coefficient k) is a measure of the hardened slurry's filtration conductivity, it depends on the surface area and pore structure of the hardened slurry [12, 37, 48, 49].

2.3. Components of hardening slurries

Basic components of hardening slurries are as follows:

- Water – water deemed useful for concrete mixtures is generally used in slurries [50].
- Bentonite – a strongly swelling sedimentary rock consisting mainly of montmorillonite, a mineral characterised by a high specific surface area and capacity to absorb water and swell [36, 51]. Sodium bentonites exhibit higher water absorption [42]; calcium bentonites are activated by exchanging calcium ions for sodium ions [36, 42].
- Cement – mainly responsible for the mechanical properties of the hardened slurry [36, 53]. Portland cement is most commonly used in slurries (CEM I [54] or Type I [55]), except under conditions of expected significant chemical aggressiveness [36].
- Fly ashes – by-products of combustion, a finely grained dust captured from the flue gases [54]. Their use helps reduce the cost of the slurry and its impact on the environment by substituting cement [39]. Slurries allow for the incorporation of problematic wastes, including ash from thermal treatment of municipal sewage sludge and fluidized bed combustion ashes [38, 39, 53, 56]. Different ashes affect the properties of slurries in various ways, but they generally improve their chemical resistance [9, 13].
- Ground granulated blast furnace slag – a steel industry by-product widely used for its latent hydraulic properties and pozzolanic activity [57]. Like ash, it reduces costs and

negative environmental impacts. The slag hydration requires an activator (alkaline compounds), contained e.g. in lime fly ash or fluidized bed ash [39]. Slurries with the replacement of part (or even all) of the cement with slag achieve higher strengths and lower permeability over an extended time [39, 58, 59].

– Modifiers – added in relatively small amounts, designed to improve selected properties of the liquid slurry (e.g. rheology by the addition of sodium salts [60]) and/or the hardened slurry (e.g. ductility and crack-healing ability with polymers [61, 62]).

3. Durability of self-hardening slurries

In broad terms, corrosion is understood as a gradual change in the technical properties of a material, which leads to the deterioration of its functional characteristics and eventually to its complete destruction; the concept of corrosion aggressiveness refers to the ability of the corrosive environment to act on a given material. Corrosion aggressiveness depends on the type and intensity of corrosive agents that are present in the environment.

3.1. Physical and mechanical factors

Table 1 shows the physical and mechanical factors that can affect the durability of self-hardening slurries and the means of counteracting them.

Self-hardening slurries, due to their hydraulic binder content, once set, are generally resistant to soaking [11]; studies indicate that even bentonite-ash-water slurries (ash acting as a binder) are capable of retaining integrity in water [71]. Self-hardening slurries, on the other hand, are not resistant to drying. The process of water loss is associated with significant deformation and shrinkage cracking [63, 64], highly undesirable considering the purpose of a cut-off wall [11, 65], and a decrease in compressive strength [66]. Cracks formed due to moisture loss are permanent and a recurrent increase in the moisture content of the slurry will not close them [34, 64]. Studies have shown that changes in moisture content and thermal shrinkage of the slurry also cause changes in stress in the structure in the longitudinal direction [63]. Some studies indicate that the durability of the material is maintained under the natural moisture content of the soil in the aeration zone [67], but it also depends on the tensile strength of the hardened slurry. Below the groundwater table, drying of the slurry does not occur [11].

Due to their high moisture content, reaching up to 200%, slurries are not frost resistant [11, 36, 68, 69]. However, due to the operating conditions of the cut-off wall (in soil, mostly below the freezing depth or with a protective layer of non-cohesive soil), this property is not a problem. To counteract the adverse effects of moisture shrinkage and to improve the frost resistance of clay mineral-based slurries, sand can be added [68, 72, 73].

The loads caused by the pressure of groundwater, dammed up on the cut-off wall, and the resulting filtration are resisted by the material's low water permeability, adequate shear strength [11] (ensuring resistance to piping and sufficient bending strength) and proper wall thickness [36]. In the case of suspended type cut-offs (not reaching the impermeable soil

Table 1. Physical and mechanical factors that can affect the durability of self-hardening slurries

Type of impact	Factor	Effects	Means of counteraction
Physical	Change in moisture content [11, 34, 63–67]	Moisture-induced shrinkage or softening, occurrence of longitudinal stress	Addition of cement or sand. Effective protection against drying is provided by covering with protective soil layers of thickness depending on the severity of exposure [36]. Addition of SAP (polymer) [62]
Physical	Change in temperature / ground frost [11, 36, 63, 68, 69]	Decrease in mechanical strength, fracturing, peeling, occurrence of longitudinal stress	Addition of cement, application of a protective layer of non-cohesive soil with a thickness at least equal to the ground freezing depth [36]
Mechanical	Water seepage [11, 36]	Suffusion	Addition of cement, ground granulated blast furnace slag, fly ash
Mechanical	Washout around the bottom end [11, 36]	Erosion	Technical measures, including reinforcement of the wall base and lengthening of the wall
Mechanical	Stress caused by external loads and self-weight [11, 12, 36, 70]	Strain (deformation), cracks, fractures	Suitable selection of components to achieve the required mechanical properties. Addition of SEBS polymer [61]

layers), the critical point is their lower end, where an accumulation of relatively coarse-grained material with less binder (as a result of sedimentation) or native soil particles (in the case of low viscosity slurry) occurs. In addition, this is an area particularly prone to intense washout by the water seepage (density of equipotential lines of the flow net and the occurrence of high pressure gradients) [11], resulting in erosion of the material. A way to counteract this phenomenon is to reinforce the cut-off wall at the base, e.g. by sinking concrete beams into the bottom of the slurry trench, or by creating an excess length of the wall for erosion losses.

3.2. Chemical factors

The problem of chemical corrosion of self-hardening slurries occurs when structures made of them come into contact with a chemically aggressive environment, which is particularly relevant in the case of the cut-off walls used around landfills. The operating conditions of cut-off walls separating contaminated and uncontaminated waters are different from those in walls situated in most embankment structures. Primarily, there is a high level of chemical aggressiveness of the water-soil environment, while, in many cases, the hydraulic gradients acting on the structure are smaller. This is a result of a lack of significant difference in the levels of ground water, as such cut-off walls only serve to separate (and not dam up) the polluted from the unpolluted waters [36].

Cement is the component deemed most susceptible to corrosion in self-hardening slurries [36]. The mechanisms of corrosion of Portland clinker-based binders have been extensively described in the literature [74, 75], hence these issues will be neglected in this paper. A recent review [9] highlighted gaps in knowledge regarding the chemical compatibility/resistance of slurries (a recommendation was stated that cement-based slurries should not be used under conditions of sulfate or acid aggression due to their questionable resistance). The susceptibility of self-hardening slurries to corrosion is an inherently complex issue, due to the variety of material compositions (CB, SCB, FCB) and its potential exposure to a range of impacts, including various aggressive substances. Self-hardening slurries and especially their corrosion resistance have been the subject of numerous studies at the Warsaw University of Technology over the past few decades [6, 7, 11, 12, 36–38, 38, 40]. Not all of their results have reached a wider audience; for example, there are no references to them in [9], which is likely due to language barriers, among other reasons.

The way in which the aggressive environment affects the cut-off wall material depends not only on the chemical composition of the water-soil environment, but also on how the structure was constructed. In the two-phase method, the wall is made without contact with the aggressive environment, it will be exposed to it only after construction. In the case of the single-phase method, it is possible to introduce into the composition of the self-hardening slurries the contaminants present in the subsoil already at the stage of construction, despite the maintenance of the slurry level in the excavation above ground water table. Table 2 shows the effect of selected substances on selected properties of the slurry in liquid and hardened state. After hardening, the slurries were subjected to transfer of contaminants caused by capillary forces and diffusion (non-filtration), which may represent a situation when the value of the so-called initial gradient is not exceeded [76] (the difference in the level of the separated waters is less than the limit, beyond which filtration will occur).

Table 2. Impact of selected aggressive factors on selected properties of self-hardening slurries

Slurry type	Aggressive agent (concentration of aqueous solution)	Effect of the aggressive action	
		Batched water containing the aggressive agent and samples submerged in its solution**	Non-contaminated batched water and samples submerged in the aggressive solution***
CB	CO_2 (aggressive) (40 mg/dm^3) [11]	Marsh viscosity ↑ Bleeding ↓ *Moisture content ↑ *Sample weight ↑ *Compressive strength ↓ *Tensile strength ↓	Compressive strength ↑ Tensile strength ↑ Sample weight (–) Moisture content ↓

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Table 2 – *Continued from previous page*

Slurry type	Aggressive agent (concentration of aqueous solution)	Effect of the aggressive action	
		Batched water containing the aggressive agent and samples submerged in its solution**	Non-contaminated batched water and samples submerged in the aggressive solution***
CB	NH_4NO_3 (1%) [11]	Marsh viscosity ↓ Bleeding ↑ *Moisture content ↓ *Sample weight ↓ *Compressive strength ↓ *Tensile strength (–)	Compressive strength ↑ Tensile strength ↑ Sample weight (–) Moisture content ↓
CB	$Mg(NO_3)_2$ (1%) [11]	Marsh viscosity ↓ Bleeding ↑ *Moisture content ↓ *Sample weight ↓ *Compressive strength ↓ *Tensile strength (–)	Compressive strength ↑ Tensile strength ↑ Sample weight (–) Moisture content ↓
CB	HNO_3 (0.5%) [11]	Marsh viscosity ↓ Bleeding ↑ *Moisture content ↓ *Sample weight ↓ *Compressive strength (–) *Tensile strength (–)	Compressive strength ↑ Tensile strength ↑ Sample weight (–) Moisture content ↓
CB	Na_2SO_4 (1%) [11, 77]	****Marsh viscosity ↓ Bleeding ↓ [11] *Moisture content (–) [11] *Sample weight ↑ [11] *Sample weight (–) [77] *Compressive strength ↑ [11, 77] *Tensile strength ↑ [11, 77]	Sample weight ↑ [11] Cracking [11, 77] Moisture content ↓
FCB	Na_2SO_4 (1%) [11, 77]	–	Compressive strength ↑ [11, 77] Tensile strength ↑ [11, 77]
CB	Distilled water [11]	Marsh viscosity ↑ Bleeding (–) *Moisture content (–) *Sample weight ↑ *Compressive strength ↓ *Tensile strength ↓	Compressive strength ↑ Tensile strength ↑ Sample weight (–) Moisture content ↓
CB (slag cement)	Phenol (up to 35 mg/dm^3) [11, 78, 79]	Viscosity (–) Bleeding (–) Compressive strength (–)	Compressive strength (–)

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Table 2 – *Continued from previous page*

Slurry type	Aggressive agent (concentration of aqueous solution)	Effect of the aggressive action	
		Batched water containing the aggressive agent and samples submerged in its solution**	Non-contaminated batched water and samples submerged in the aggressive solution***
CB (slag cement)	NH_4Cl (1000 mg/l) [79]	–	Compressive strength after 28 days (–) Compressive strength after 42 days ↓
CB (slag cement)	NH_4Cl (10000 and 20000 mg/l) [79]	–	Compressive strength ↓

CB – cement-bentonite slurry, FCB – cement-bentonite-fly ash slurry (ash from the combustion of hard coal
* – based on the comparison of samples made of contaminated batched water and stored in the aggressive solution for approx. 1 year with samples made of potable water and stored in the aggressive solution for approx. 1 year
** – compared to the slurry made using potable water
*** – compared to the slurry stored in potable water
**** – sodium ions usually reduce the viscosity of the slurry; it is difficult to assess the impact of sulfate ions on the reduction of the viscosity
↑ – increase; ↓ – decrease; (–) – no significant change
The impact of simultaneous action of more than one aggressive agent was not tested.

The number of studies of the effect of aggressive agents in the batched water on the properties of fluid self-hardening slurries is scarce, most of them dealing with agents that are generally aggressive to cementitious binder-based materials. A summary of the review of the available literature in this area is presented in Table 2. The results of the cited studies indicate the effects of chemicals dissolved in the batched water on the properties of slurries in the liquid state and on their subsequent performance [11, 77, 78]. Most of the corrosive solutions contained in the batched water resulted in a decrease in the viscosity of the slurry, which translated into an increase in bleeding (except for aggressive CO_2 and distilled water) relative to samples treated with potable water (Table 2), which may affect the durability of the slurry (greater sedimentation, greater contamination with native soil particles).

On the other hand, a comparison of the strength of slurries stored in aggressive solutions indicates that the content of aggressive substances in the batched water (compared to samples made using potable water) has a minor, rather negative effect on the properties tested (Table 2). The exception is the effect of sodium sulfate solution, under the influence of which the samples prepared with potable water cracked, while those mixed with sodium sulfate solution did not, which the authors [11, 77] attribute to a uniform formation of ettringite in the entire mass of the samples (and not only on the surface, which can cause a local increase in stress). The swelling and increase in the weight of sodium sulfate-stored specimens is due to the binding of sulfate ions by the specimen in its structure, a process expected for cement-based materials and described in the literature [74]. A remedy for improving the resistance of CB slurry to the corrosive effects of sulfate is the addition of slag and fly ash. The pozzolanic properties of the industrial by-products and the adsorptive properties of the unburned carbon remaining in the waste weaken the destructive effects of sulfates [11].

A comparison of strength test results for samples made with potable water and stored in various aggressive environments to those stored in potable water suggests that the slurries were essentially unaffected by corrosion (except for those exposed to sodium sulfate), and their average strengths even increased [11].

The cited studies [11] indicate that subjecting samples made with potable water or aggressive solutions to the influence of an aggressive environment slightly affects the moisture content of the samples. In both cases a steady increase in the water content of the samples can be observed, which indicates the chemical transformations taking place in the material.

Changes in the weight of samples subjected to various aggressive environments are usually a good measure of corrosion progress. In the case of self-hardening slurries, other phenomena, such as the hydration of the cementitious binder and the saturation of the samples also affect the mass changes [11], so it is difficult to determine the influence of corrosive factors unambiguously. Particularly intense aggression is found in environments containing nitric acid [11].

The effect of aggressive substances on the permeability of self-hardening slurries has received far more attention in the literature (Table 3). It is widely believed that the low filtration coefficient of self-hardening slurries is a prerequisite for their considerable durability in contact with aggressive media [11], while under the influence of concentrated solutions of acids or alkalis, dissolution of alumina can occur, resulting in an increase in the hydraulic permeability of hydrated bentonite barriers [80].

For most of the aggressive agents (Table 3), there was no corrosion effect manifested as an increase in the filtration coefficient, regardless of the composition of the slurry. In the case of ammonium corrosion, the resistance of CB slurries depended on the concentration of the aggressive solution [11, 78], while slurries with the addition of fly ash showed a lack of resistance to this type of aggressiveness [12, 37].

Exposure of slurries to the filtration of chemically aggressive solutions, potable water and distilled water leads to dissolution and leaching of $Ca(OH)_2$, followed by decomposition of other hydrated phases. The filtration interaction of potable water and distilled water are different; the purer the water, the higher the corrosion rate [84], which is confirmed by the cited results of distilled water filtration of fluidized bed fly ash slurries [12, 37].

The effect of $Mg(NO_3)_2$ solution on self-hardening slurries with the addition of fluidized bed combustion fly ash depends on the type of ash. The addition of ash from hard coal combustion increased the hydraulic permeability of slurries, while the addition of ash from lignite combustion increased the tightness. The authors of the study [12, 37] explain the positive effect of the latter ash by its reactivity (hydraulic and pozzolanic), due to which new phases are formed, sealing the microstructure, thus restricting the access of substances corrosive to calcium hydroxide.

No corrosion was observed as a result of subjecting slurries with ash from fluidized bed combustion of hard coal and lignite to filtration with sodium sulfate and nitric acid

Table 3. Effect of aggressive solutions on the filtration coefficient of self-hardening slurries

Slurry type	Aggressive agent, concentration of aqueous solution	Impact on the filtration coefficient* (final coefficient observed after 100–200 days of test)
CB	K_2SO_4 5 g/dm^3, 27.5 g/dm^3, 50 g/dm^3, 95 g/dm^3	↑ [81]
CB (slag cement)	Test liquid (Sulphate 14 650 mg/dm^3+ Chloride 100000 mg/dm^3+ Ammonium 12000 mg/dm^3+ Phenoles 350 mg/dm^3+ Methylene Chloride 50 mg/dm^3+ Toluole 10 mg/dm^3)	**↑ [79]
FCB(H)***	Na_2SO_4 (1%)	↓ [12, 37, 49]
FCB(H)***	Distilled water	↑ [12, 37, 49]
FCB(H)***	HNO_3 (0.5%)	↓ [12, 37, 49]
	HNO_3 (0.5 M)	↓ [82]
FCB(H)***	NH_4NO_3 (1%)	↑ [12, 37]
FCB(H)***	$Mg(NO_3)_2$ (1%)	↑ [12, 37]
FCB(L)***	Na_2SO_4 (1%)	↓ [12, 37, 49]
FCB(L)***	Distilled water	(–) [49] ↑ [12, 37]
FCB(L)***	HNO_3 (0.5%)	(–) [49] ↓ [12, 37]
	HNO_3 (0.5 M)	↓ [82]
FCB(L)***	NH_4NO_3 (1%)	↑ [12, 37]
FCB(L)***	$Mg(NO_3)_2$ (1%)	↓ [12, 37]
SCB	Aniline (0.5%)	(–) [83]
SCB	Phenol (0.5%)	(–) [83]
CB (slag cement)	Phenol (350 mg/dm^3)	(–)** [79]
CB (slag cement)	NH_4Cl (20 000 mg/dm^3)	↑** [79]
	NH_4Cl (1000 mg/dm^3)	(–)** [79]

* – compared to the slurry exposed to potable water
** – compared to the slurry exposed to distilled water
*** – cement-bentonite slurries with the addition of fly ash from fluidized bed combustion of hard coal (H) or lignite (L)
↑ – increase; ↓ – decrease; (–) – no significant change

solutions regardless of the type of ash. The hydraulic permeability of both types of slurries was reduced [12, 37, 49] and there was no 'loss of tightness'; the authors of the study explain this by the formation of complex hydrated sulfate salts and amorphous phases in the form of, for example, hydrated silica gel [85] and colloidal silicic acid [12, 82]. Especially the result of nitric acid filtration is opposite to the effect of acid corrosion usually described in the literature concerning cement concrete and mortar, [74, 75, 86]. The addition of fluidized bed combustion ash can also result in the formation of hydrated carbonate aluminate, which is responsible for increasing the resistance of cementitious binders to sulfate corrosion [75].

4. Carbon footprint of selected self-hardening slurries

Quantification of CFP or EC is extremely difficult, especially when considering all stages of the product life cycle (from *cradle-to-grave*). In the literature on the subject, you can find many methodologies for calculating the carbon footprint, i.e. [87]. It seems that the methods based on a bottom-up (from the detail to the whole) approach [17] will be preferable for building materials. Nevertheless, it is certain that replacing raw materials with anthropogenic materials, particularly waste materials, is beneficial and expected for the climate and the assumption of the Circular Economy (CE).

Within all quantification methods, the underlying principle of carbon footprinting is always the same. Activity data is multiplied by emission factors. These factors convert the emissions associated with each activity into a CO_{2eq} based on their GWP. These factors quantify the carbon content of all the elements needed for the calculation (for example, the carbon content of a ton of concrete or the carbon content of a kilometre travelled by truck). These emission factors come from public or private databases, which can contain thousands of emission factors, and cover a broad range of activities not only from construction-related industries. For construction works, two main types of emission sources can be distinguished [35] (Fig. 3):

a. Primary Emission Sources:
   - Embodied carbon in materials, due to their manufacturing (cement, bentonite).
   - Materials transportation, from factory to construction site (trucks, boats).
   - Energy consumed on the construction site (electricity, fossil fuels).
b. Secondary Emission Sources:
   - People's transportation to the construction site.
   - Equipment's transportation.
   - Equipment's manufacturing (depreciation).
   - Waste management (waste transportation, from construction site to treatment site, waste treatment).

In the area of deep foundation and ground improvement techniques, it was found that the total of the primary emission sources represented more than 90% of the total carbon footprint. In the case of groundworks, where material consumption is high, embodied carbon in materials is the most significant contributor to emissions. In the case of self-hardening slurries used for cut-off walls, the material is responsible for more than 80% of CO_{2eq} emissions [35].

Exemplary calculations of the carbon footprint for individual hardening slurries were performed using the Microsoft Excel based EFFC DFI Carbon Calculator Tool v4.0, which is compatible with the following standards: GHG Protocol Product Life Cycle Accounting

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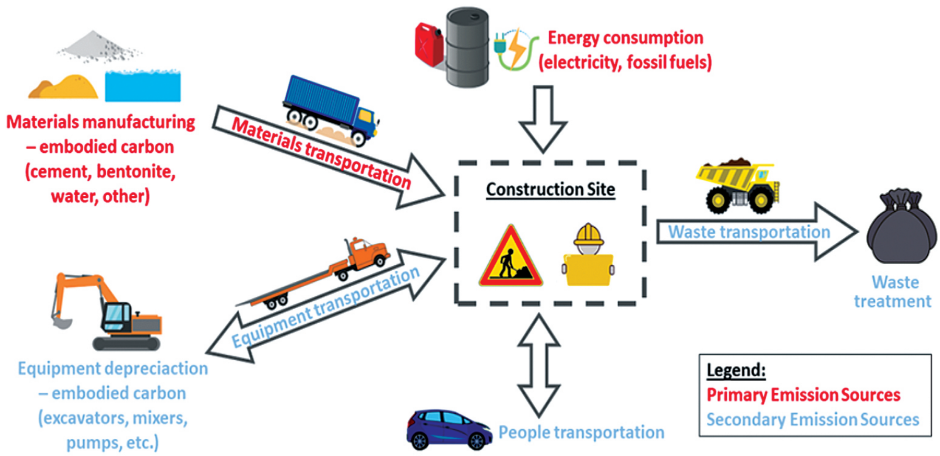

Fig. 3. GHG emission sources in a cut-off wall project. Based on: [35]

and Reporting Standard [88], Bilan Carbone [89], PAS 2050 [90], ISO 14067 [20]. An accurate description of the calculation method can be found in [35]. The calculations were performed for the Primary Emissions Sources, modules A1 to A4. The slurry installation process itself was not included for two reasons. First, because the material (composition of a self-hardening slurry) here is the source with by far the highest emissions. Secondly, because of the possibility of making the cut-off walls with different methods, which have different emission levels (comparing the carbon footprints of the various methods of making cut-off walls requires further exploration outside the framework of this article). For the same reasons, Secondary Emission Sources were not included in the calculations. The calculation assumptions are presented in Table 4.

Table 4. The carbon footprint calculation assumptions

Assumption	Value
Scope of LCA	A1–A4
Data base	EFFC DFI methodology recommended emission factors
Dimensions and volume of the cut-off wall	0.6 m wide, 10 m deep, 1000 m long, 6000 m^3 in volume
Number of recipes considered	4
CEM I	Emission factor – 860 kg CO_{2eq}/Mg Transportation distance – 150 km
CEM III (70% Ground Granulated Blast-Furnace Slag)	Emission factor – 314 kg CO_{2eq}/Mg Transportation distance – 150 km

Continued on next page

Table 4 – *Continued from previous page*

Assumption	Value
Bentonite	Emission factor – 475 kg CO_{2eq}/Mg Transportation distance – 300 km
Fly ash	Emission factor – 0.1 kg CO_{2eq}/Mg Transportation distance – 300 km
Ground Granulated Blast-furnace Slag	Emission factor – 80 kg CO_{2eq}/Mg Transportation distance – 300 km
Water	Emission factor – 0.3 kg CO_{2eq}/Mg Transportation distance – 20 km
Truck load	8 m^3

Table 5 shows the compositions of the slurries analysed, their properties and requirements in terms of the properties of the hardening slurries used for cut-off walls in levees [91, 92] and as hydraulic barriers in landfills [93]. Slurry recipes that meet the requirements for cut-off walls were adopted for the calculations. This made it possible to compare the carbon footprints for specific mixtures with similar properties.

Table 5. The compositions of the hardening slurries, their properties and requirements in terms of the properties of the self-hardening slurries used for cut-off walls

Properties	Recipe 1 [94]	Recipe 2 [31]	Recipe 3 [12]	Recipe 4 [39]	Specific requirements in terms of the properties of the hardening slurries used for cut-off walls
Bulk density [g/cm^3]	1.16	1.33	1.30	1.33	1.15–1.40 [91, 92]
Conventional (Marsh) viscosity [s]	50	50	39	37	$\leq$ 50 [91, 92]
Daily water loss [%]	–	3.9	5.0	4.0	$\leq$ 4.0 [91, 92]
Structural strength after 10 min [kPa]	–	5.2	–	3.8	1.4-10.0 [91, 92]
Compressive strength after 28 days of curing [MPa]	–	1.80	1.38	1.62	0.5–2.0 [91, 92]
Filtration coefficient k10 [m/s] (after 28 days of curing)	$< 10^{-8}$	$9.6{\times}10^{-9}$	$2.5{\times}10^{-8}$	6.35×10^{-10}	$\leq 1,0 \times 10^{-8}$ (levees) [91, 92] $\leq 1,0 \times 10^{-9}$ (landfills) [93]

Continued on next page

Table 5 – *Continued from previous page*

Properties	Recipe 1 [94]	Recipe 2 [31]	Recipe 3 [12]	Recipe 4 [39]	Specific requirements in terms of the properties of the hardening slurries used for cut-off walls
Component [kg/m^3 of the slurry]					
Tap water	900	841	842	869	
Sodium bentonite	40	21	25	35	
CEM I	–	380	143	–	
CEM III	200	–	–	–	
Ground blast furnace slag	–	–	–	413	
Hard coal fly ash-fluidized bed	–	–	–	–	
Brown coal fly ash-fluidized bed	–	–	275	13	
Thermal treatment of municipal sewage sludge fly ash	–	84	–	–	

4.1. Results and discussion

Due to the many assumptions, averages and high level of uncertainty in the data, the calculations' results can only be regarded as a possibly accurate estimate. Table 6 shows a comparison of carbon footprints (modules A1–A4) for four recipes of hardening slurries with different compositions but comparable properties, allowing them to be used as cut-off walls in levees.

Table 6. Carbon footprints for four recipes of self-hardening slurries. Source: own elaboration

Carbon footprint	Recipe 1	Recipe 2	Recipe 3	Recipe 4
Total (6000 m^3 of slurry) [$MgCO_{2eq}$]	650	2210	1010	510
Materials (modules A1–A3) [$MgCO_{2eq}$]	490	2000	810	300
Material Transportation (module A4) [$MgCO_{2eq}$]	160	210	200	210
CF of 1 m^3 of slurry (modules A1–A4) [$MgCO_{2eq}$/m^3 of slurry]	0.11	0.37	0.17	0.09

The calculations presented show that:

– The carbon footprint for self-hardening slurries depends mainly on the amount and type of cement used; the use of blast furnace slag cement or the complete abandonment of the cement binder can significantly reduce the carbon footprint of the slurry.
– As the proportion of non-cementitious components in the slurry increases, the amount of emission associated with the transport of materials increases (a longer transportation distance for combustion by-products than for cement was assumed). In the case of recipe no 4 (without cement), transporting materials to the construction site amounts to more than 40% of the carbon footprint of the slurry. If possible, it is important to use local materials in slurry.
– Depending on the composition of the hardening slurry, its carbon footprint per cubic meter can be several times smaller than the carbon footprint of concrete.
– It is possible to significantly reduce the carbon footprint of the hardening slurry used for cut-off walls while maintaining its desired properties.

Potentially, further reductions in the carbon footprint of cut-off walls will be possible by mixing self-hardening slurries with the native soil, e.g. by using the Continuous Deep Mixing method. The issue requires further research.

5. Leaching and immobilization of contaminants in self-hardening slurries

As structures in contact with groundwater, cut-off walls are crucial to environmental protection. Firstly because of the possibility of its direct contamination – due to the widely used waste materials from thermal processes (e.g. fly ash) in the composition of self-hardening slurries, and secondly, because of their use in landfills as a hydraulic barriers.

The physical transport of contaminants in a water-saturated ground medium can occur by diffusion, advection and surface leaching. Descriptions of these phenomena can be found, e.g. in [56, 95, 96]. The transport of contaminants depends on the magnitude of the hydraulic gradient, material filtration rate, and chemical gradient [97]. In the analysis of contaminant migration in the aquatic environment, sorption processes (sorption is intended as the overall mechanisms which remove the contaminant from solution; these processes include cation exchange, precipitation, adsorption and, in general, any binding between solutes and the solid matrix [94]) are essential, which result in the retention of contaminants in the material [97]. It is also known that bentonites have a significant sorption ability for cations, as well as a low water permeability [72, 98] and their properties are a function of the content of montmorillonite and the nature and number of interlayer cations [99], which is why it is used in cut-off walls to reduce contaminant migration.

Studies on the immobilization (defined as the percentage of residual substance in the slurry after leaching to the original amount of substance in the slurry) of heavy metals in self-hardening slurries derived from slurry components indicate that:

- The level of immobilization is high, at more than 90% [31, 100], also when considering the entire life cycle of the cut-off wall [101].
- The obtained eluates were characterized by heavy metal concentration below the limit value for landfill leachates [31, 100, 102, 103].
- The release mechanism depended on the leaching scenario. So, for example, for the percolation test [104] the predominant release mechanism was dissolution, whereas for the tank test [105] was surface leaching [101].
- Heavy metal release was dependent on the curing time of the slurry [103, 106, 107], L/S (liquid to solid) ratio [101], pH [101] and type of leaching liquid [103, 106–108].

The research indicates that the different types of cut-off walls used to protect landfills are characterised by their sorptive properties towards leachate contaminants. Noted are:

- Sorption of sulphate in CB slurry, resulting in ettringite formation and a reduction in hydraulic conductivity [94].
- Strong sorption of heavy metal solution and phthalate solution in a slurry based on Portland cement, modified bentonite, fly ash and additives as a result of low hydraulic conductivity and physical and chemical adsorption with fly ash and bentonite in hardened slurry [109–111].
- Sorption of nitrogen and potassium compounds occurs in clay soils' hydraulic barriers [97, 112].
- Filtration through the FCB slurry of the ammonium nitrate solution (1%) resulted in a clear and colourless filtrate [12].

The issue of contaminant sorption by hardening slurries requires further research.

6. Summary

Cut-off walls based on self-hardening slurries are an essential environmental engineering tool that can respond to various social and environmental challenges. The drive to reduce environmental and climate degradation requires a good knowledge of the interaction of a structure with the environment during its life cycle, which is also the case for cut-off walls.

Self-hardening slurries are exposed to various corrosion mechanisms resulting from the aggressiveness of the environment and working conditions. Chemical aggression associated with exposure to solutions of different chemical compounds is considered the most dangerous, especially when slurries are used in sealing landfills. Its effect on the properties and durability of the material is complex, depending, among other things, on the slurry's composition and the aggressive solution's type and concentration. Despite numerous studies, it has yet to be possible to outline general recommendations for designing the slurry composition. Hence, a compatibility study is necessary each time to ensure adequate durability of the cut-off wall. Improving the durability of a structure goes a long way to reducing costs and negative environmental impacts during its life cycle.

Self-hardening slurries contain a binder, usually Portland cement, which strongly affects their carbon footprint. However, they also allow for the safe use of significant amounts of SCMs, including those with undesirable properties, for example, from the point of view of cement concrete, thus significantly reducing their carbon footprint, at least in embodied carbon (modules A1–A3). Differences in the carbon footprint of cut-off walls made with different technologies require further investigation. Particularly valuable would be a comparative analysis of cement-bentonite slurry versus soil-cement-bentonite slurry technologies; the differences here relate to the composition of the composite, construction method and the properties of the resulting cut-off wall.

Self-hardening slurries usually show good sorption properties. The risk of contaminants leaching from material containing various industrial by-products is therefore strongly reduced. In addition, slurries have sorptive properties towards contaminants from the leachate with which they come into contact – an issue that requires further research.

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Zawiesiny twardniejące w kontekście współczesnych wyzwań środowiskowych

Słowa kluczowe: immobilizacja, korozja, ślad węglowy, trwałość, uboczne produkty spalania, zawiesiny twardniejące

Streszczenie:

Zawiesiny twardniejące stanowią ważne narzędzie nowoczesnej inżynierii dążącej do realizacji celów Zrównoważonego Rozwoju. Pomimo iż materiał ten stosowany jest zdecydowanie mniej powszechnie od betonu cementowego, to dotyczą go podobne wyzwania związane z rosnącą presją człowieka na środowisko – zmniejszenie negatywnego oddziaływania (np. w wyniku obniżenia śladu węglowego, czy zwiększenia wykorzystania odpadów po-procesowych), zwiększenie trwałości, itd. Pochodną węższego zakresu wykorzystania zawiesin twardniejących w szeroko rozumianym budownictwie jest względnie niewielka baza dostępnych danych literaturowych kompleksowo opisujących ten rodzaj materiału ze środowiskowego punktu widzenia. Niniejszy artykuł stanowi przegląd uzupełniający bieżący stan wiedzy na temat zawiesin twardniejących, ze szczególnym uwzględnieniem trwałości materiału i jego zdolności do immobilizacji zanieczyszczeń. W celu nakreślenia kontekstu krótko scharakteryzowano zawiesinę, warunki jej pracy (w przesłonach przeciw filtracyjnych), właściwości i składniki. Szeroko opisana została odporność zawiesin twardniejących na agresję środowiskową (fizyczną i chemiczną) jako czynnik kluczowy dla trwałości materiału. Wykonano przykładową analizę śladu węglowego zawiesiny twardniejącej w kilku zbliżonych pod względem właściwości technologicznych i użytkowych wariantach recepturowych. Przedstawiono zarys zagadnienia immobilizacji i sorpcji zanieczyszczeń w materiale. Zaproponowano kierunki dalszych badań.

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