(1) Title: Review on Stabilization/Solidification methods and mechanism of heavy metals based on OPC-based binders

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Review on Stabilization/Solidification methods and mechanism of heavy metals based on Ordinary Portland Cement-based binders

Abstract

Stabilization/solidification (S/S) with ordinary portland cement (OPC)-based binders is a suitable method to remediate heavy metal (HM)-contaminated soil and reuse resources of industrial wastes. In industrial wastes, alkaline wastes such as red mud (RM), soda residue (SR), pulverized fly ash (PFA), and alkalinity granulated blast furnace slag (GGBS) can immobilize HM ions (Pb^{2+}, Zn^{2+}, Cd^{2+}, Cr^{3+}, and Cu^{2+}) by precipitation. However, some HM ions (such as AsO_4^{3-}) would redissolve within the strong alkali environment. In this case, PFA, GGBS, metakaolin (MK), and incinerated sewage sludge ash (ISSA) which have low pH, can be used to immobilize HM ions or added to the OPC-based binders to adjust the pH in the soil products. Moreover, the calcium silicate hydrate (CSH), calcium aluminum silicate hydrate (CASH), ettringite (AFt), and calcium monosulfoalumiante hydrates (AFm) generated during the pozzolanic reaction can also immobilize HM ions by adsorption on the surface, ion exchange, and encapsulation. SR and GGBS can be used to immobilize the HMs (such as CrO_4^{2-} and AsO_4^{3-}), which are mainly affected by AFt and AFm. For those not affected by AFt and AFm but related to immobilization by precipitating (such as Mn^{2+}), other wastes except SR and GGBS are suitable for treating contaminated soil. Nevertheless, the formation of AFt is also instrumental for soil product strength. There are several factors affecting soil product strength. In the future, the influence of different hydration products on the S/S effects, competitive adsorption of HM ions, effects on long-term HM stabilization, and novel materials are worth being explored by researchers.

Keywords: soil, industrial wastes, CSH gels, immobilization

1. Introduction

The development of heavily-polluted industrial affiliated enterprises, fossil fuel consumption, mining, smelting, agrochemical use such as Cu-based pesticides, and municipal solid waste discharge over the last decades increased the accumulation of heavy metals (HMs) in the soil (Ji and Pei, 2019; Liu et al., 2018b; Wang et al., 2018b). These toxic HMs with high concentrations not only impair the natural ecosystem services of the human survival environment but also eventually damage human health via the food chain.

Over the years, many remediation techniques have been used to restore HM-polluted soil. In these methods, promising stabilization/solidification (S/S) technology has been extensively developed both in in-situ and ex-situ soil remediation due to its feasibility of the technology, high economic efficiency, and short remediation time (compared with phytoremediation) (Chen et al., 2009; Conner and Hoeffner, 1998b; Fan et al., 2018). The mechanism involved in S/S means that contaminants are converted into the least soluble/toxic and less mobile form by stabilization (Eisa et al., 2020). Besides, solidification can also integrate soil into a monolith mass of high structure involving both chemical interaction and mechanical binding to capsulate hazard contaminants, which further restricts the increase of leaching concentration of HM ions (Reddy et al., 2020; Xia et al., 2020). Under these series of physical (adsorption and encapsulation) and chemical (complexation, precipitate, chelation, etc.) actions, HM is converted into environmentally acceptable speciation for construction materials and land disposal use (Ji and Pei, 2019). Jing et al. (2006) reported that the mixed solidifying binders could immobilize HM ions such as Cr ions by physical encapsulation of cement hydrates and formatting chemical precipitation of hydroxides such as $Ca_2Cr_2O_5 \cdot 6H_2O$ in contaminated soil products.

Traditionally, ordinary portland cement (OPC) has been widely used in S/S as the most prevalent material due to its alkaline properties and strength, whereas cement production increases greenhouse gas emissions and consumption of energy requirements (Singh and Budarayavalasa, 2021). According to the study by Geng et al. (2019) and Benhelal et al. (2013), the production of each ton of cement emits CO_2 2.66-0.82 tons. It is nearly 5-7 wt.% of total global CO_2. However, enormous energy consumption can be conserved by replacing parts of OPC with waste resources obtained without high-temperature processing. In recent studies, the most prevalent additives to bind OPC are soda residue (SR), pulverized fly ash (PFA), granulated blast furnace slag (GGBS), incinerated sewage sludge ash (ISSA), red mud (RM), gypsum, metakaolin (MK), and some other alkaline activators such as $Ca(OH)_2$, NaOH and KOH (Wu et al., 1990). Compared with single OPC, OPC-based binders can recycle waste resources and reduce CO_2 emissions effectively. To identify environmental burdens and impacts in detail, Hossain et al. (2020) assessed different bottom ash management options by applying Life Cycle Assessment (LCA) method and verified the environmental benefit of ash recycling combined with OPC on S/S. Zahmak et al. (2021) also used the LCA method to determine environmental impacts with alkali-activated binders and OPC. They concluded that the soil products treated with wastes and alkali activators outperformed OPC in four of the ten evaluated impact categories, including global warming. Concerning energy consumption and global warming, the superiority of the combination of fly ash and OPC in producing bricks has also been verified (Elahi et al., 2021). Moreover, OPC-based binders have good long-term stability and compatibility (Eisa et al., 2020).

For this purpose, more researchers are focusing on using waste resources as alternative materials for parts of OPC, which are called OPC-based binders, aiming to accomplish CO_2 reduction, low environmental risk, low cost, and environmentally friendly recycling usage and management (Inazumi et al., 2020; Wang et al., 2017; Zha et al., 2020). The process of S/S with OPC-based binders is shown in Figure 1. The treated soil products by S/S could be used in the following fields: repair materials, thermal insulation, sustainable construction materials, and road materials.

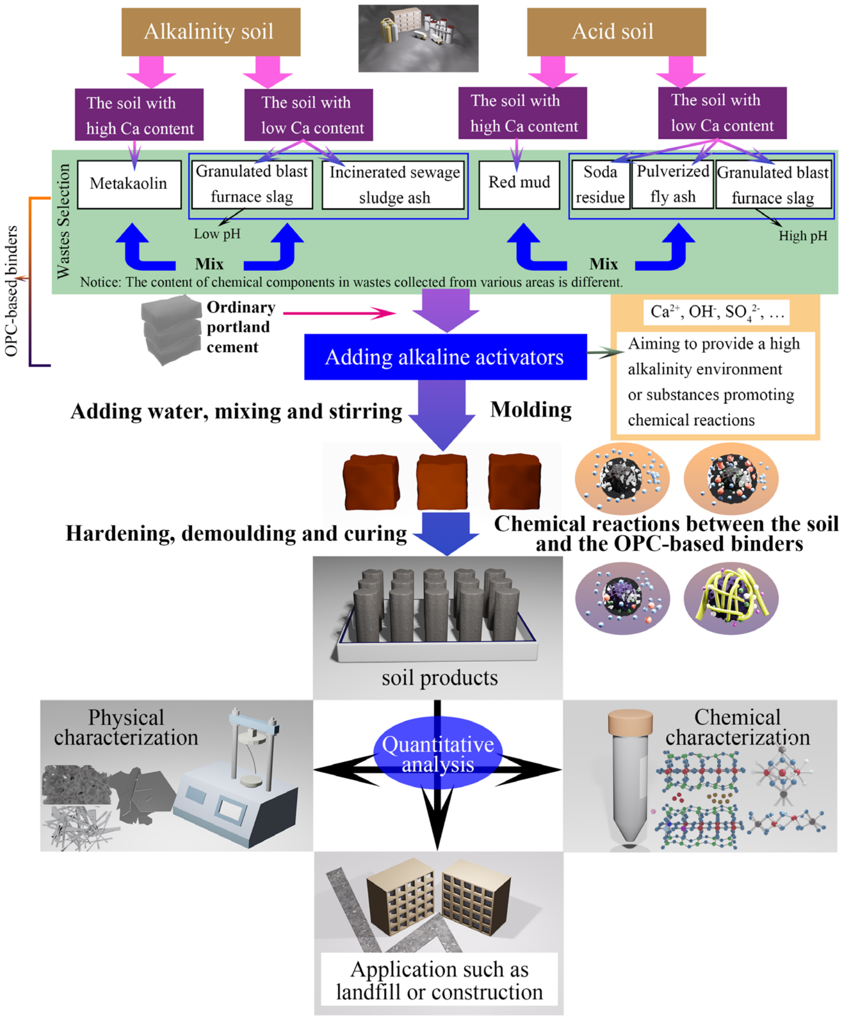

Figure 1 The process of stabilization/solidification with OPC-based binders

Therefore, based on existing research, this study is systematically conducted to review a comparative analysis of the mechanism of immobilizing HMs with different OPC-based binders and summary the representative experimental materials, conditions, effects, and interfering factors in the S/S technology in recent six years. The effects can be determined by the leaching concentration of HM ions that are usually tested by the toxicity characteristic leaching procedure (TCLP) method and the unconfined compressive strength (UCS) of solidified products. The information is expected to assist in selecting appropriate OPC combining materials to deal with HM pollutants, provide a reference for further research and application of materials on OPC-based binders, and contribute to providing a theoretical basis for the resource recovery of industrial wastes. On this base, this review prospects the relevant studies of OPC-based binders and further proposes research direction in the future.

2. OPC-based materials and selection method

2.1 OPC

OPC is a kind of heterogeneous mixture, and it is always made by heating the mixture of clay and limestone, which includes $CaCO_3$, SiO_2, Al_2O_3, Fe_2O_3, and some other residues, to about

1450°C (Chen et al., 2009; Liu et al., 2020). Single OPC has been proven to be an effective material to apply for the S/S technology of immobilizing HMs. Li et al. (2014) studied the leaching effectivity of Pb-contaminated soil products with cement. They found that the leaching concentration of Pb ions declined by 34.9% significantly when the cement-to-dry soil ratio increased from 0.2 to 0.4. In another study, Oluwatuyi et al. (2019) concluded that adding OPC increased the adsorption capacity of the soil products for the Pb contaminants. A similar observation was reported by Fei et al. (2018), who did on-site S/S experiments. It showed an excellent capacity of OPC (8 wt.%) in stabilizing Cd and Zn, of which the immobilization degrees are 99.9% and 99.4%. This research provided a reference for exploiting OPC materials to remediate HM-contaminated soil in practical S/S applications.

2.2 Waste resources combined with OPC

2.2.1 Soda residue

Soda residue (SR) is generated by the ammonia-soda process as a by-product with a pH of 9-11. Its main compositions include aragonite, calcium hydroxide, calcium sulfate, calcite, calcium chloride, calcium oxide, silicon oxide, magnesium oxide, aluminum oxide, and some oxides of iron and silicon, and its excellent characteristic is the performance of durability and porosity because of the composition of very fine particles (<0.074 mm), which made the specific surface area extremely high to adsorb more HM ions (Liu et al., 2018a; Zha et al., 2021). As the compositions of SR are similar to those in OPC (both of them have Ca, Si, and metal compounds), SR could be used as a sustainable material to replace part of OPC.

In terms of HM stability, cement-soda residue (CSR) has been shown to be an effective binder for S/S under acid rain conditions due to its alkalinity. Zha et al. (2021) studied the leaching behavior of Zn-contaminated soil products with CSR under acid rain conditions. The result mentioned that UCS decreased after leaching, whereas the addition of CSR made the Zn concentration in the filtrate accord with the third grade of the applicable standard (<1mg/L). Regarding UCS, the UCS of soil products with adding SR is less than that of adding limestone. Wang et al. (2020c) studied the UCS variation under the condition of different ratios of SR, which are added into materials at 0, 5, 10, and 15 wt.% dividedly. The UCS decreased from 54.6 MPa to 36.0 MPa after 28 days of curing when the contents of SR increased from 0 to 15 wt.%. However, the addition of SR would increase the UCS of soil products, and the soil products treated with SR have higher strength compared to high-calcium FA additives (Liu et al., 2018a; Zha et al., 2020). Adding OPC could increase the UCS of the soil products, and the freezing-thawing resistance could also be improved by adjusting the mass ratio of OPC to SR (Liu et al., 2020).

2.2.2 Pulverized fly ash

Pulverized fly ash (PFA) is derived from power plants and is a waste by-product extracted from the flue gas of a bituminous coal powder furnace (Wang et al., 2020b). The main compositions of PFA are SiO_2, CaO, Al_2O_3, MgO, and some residual carbon which are similar to the compositions of OPC (Liu et al., 2018a). Hence, PFA is endowed with a high capacity for adsorption and hydration activity.

PFA can be classified as the Class-C PFA with high Ca content and the Class-F PFA with low Ca content (Wang et al., 2020b). The usage for the divergence of these two types depends on the need for extra alkaline. Class-C PFA can increase the pH of HM-contaminated soil products, but some HMs would not be immobilized within the high pH condition. For example, As ion is more mobile in an alkaline environment above 8.5 pH. Excess of this value, the concentration of As ion would increase. Li et al. (2018) used cement blended with Class-C coal fly ash (FA) to observe the variation of As leaching concentrations in low-alkalinity conditions. The results show that the leached As ions increased considerably with cement and FA incorporation. In this case, Class-F PFA is an appropriate material to bind to OPC. Contradictory to As, Pb and Cd have a minimum leaching concentration with pH at 11 and 9, respectively. In this case, Class-C PFA is a better choice to immobilize HMs than Class-F PFA. (Liu et al., 2018a) did the S/S experiments on Pb-contaminated soil with high-calcium fly ash (Class-C) and SR. They considered that FA appeared to have a better immobilization potential on S/S than SR.

2.2.3 Granulated blast furnace slag

Granulated blast furnace slag (GGBS) is also one of the by-products made from crushed minerals during steel-making. Zhang et al. (2021b) analyzed the main chemical compositions of GGBS, which showed the maximum component is CaO, with 41.1 wt.% of the total components, and the second one is SiO_2, accounting for 34.11 wt.%. In addition to CaO and SiO_2, the metal-containing components also evidently occupied 11.16 wt.% and 6.57 wt.%, which are Al_2O_3 and MgO. Additionally, some other components have low contents, such as Fe_2O_3, TiO_2, K_2O, Na_2O, and SO_3, of which the contents are lower than 1 wt.%.

Due to the Al_2O_3-rich characteristics, the strength of GGBS-treated soil products shows excellent performance. Zhang et al. (2018) compared the UCS between single OPC-treated soil products and GGBS mixed OPC-treated soil products, which concluded that adding GGBS made UCS arise nearly 2.7 times when cured 56 days later. Furthermore, Wang et al. (2019a) demonstrated that GGBS-OPC showed better results than single OPC from both the leaching out and strength. They suggested that replacing OPC with GGBS could improve the efficiency of immobilizing Pb. Li et al. (2017) similarly concluded that the most feasible percentage of OPC replacement with GGBS for treating As contaminated soil is 20 wt.%. Furthermore, the advantage that GGBS is superior to other binders is the strong resistance to chloride and sulfate erosion (Wang et al., 2020b). More importantly, the possibility of combining GGBS and other materials is various. For example, MgO could be added into GGBS to create more hydrated compounds such as hydrotalcite ($Mg_6Al_2CO_3(OH)_{16}\cdot 4H_2O$) deposited in the available open pores to increase the strength of the soil products (Falciglia et al., 2014; Goodarzi and Movahedrad, 2017; Moon et al., 2010).

2.2.4 Incinerated sewage sludge ash

Incinerated sewage sludge ash (ISSA) is produced as a by-product from the sludge incineration plants. The major compositions of ISSA can be inferred to consist of SiO_2, Fe_2O_3, Al_2O_3, CaO, Na_2O, and MgO by Li et al. (2021a). This research revealed that the ISSA possessed an excellent capacity for adsorbing Pb ions with a 94% removal rate in the 3.0-9.0 pH range. ISSA could remove HMs by adsorbing on the surface due to its large surface area and porosity (Smith et al., 2009; Wang et al., 2020b). However, the UCS of ISSA-treated soil products is lower than GGBS-treated soil products. Wang et al. (2018a) indicated that the soil products of ISSA and GGBS incorporation possessed good mechanical strength, whereas the addition of ISSA would negatively impact the compressive strength compared with the single GGBS.

2.2.5 Red mud

Red mud (RM) is a by-product produced in the production process of aluminum. It is also a kind of industrial waste in which a large number of iron oxides exist inside, resulting in red color. The apparent characteristic of RM is its alkalinity, which can increase the pH of soil products

(Korznikov, 2015). The composition of RM can be seen from the study by Wang et al. (2020a) that the main parts are Fe_2O_3, Al_2O_3, and SiO_2. Besides combining with OPC, the research stated that $R_4P_1PC_3$ and $R_4P_1PC_5$ (R: RM, P: phosphorus gypsum, PC: cement, the numbers represent proportions), which are the RM mostly contained groups, showed the lowest leaching concentration. Fe_2O_3 also contributes to the adsorption of HMs. Because of the performance of solidified soil products on leaching concentration and UCS, the binders with RM could be used for low-carbon and low-cost treatment of S/S (Wang et al., 2019c).

2.2.6 Metakaolin

Metakaolin (MK) is a pozzolanic material made from kaolinitic clay at the temperature of 500°C-800°C (Siddique and Klaus, 2009). The significant oxide components are Al_2O_3 and SiO_2. MK possesses a volcanic ash effect to accelerate the speed of hydration reaction in cement and fill the pores of hydration production to strengthen the UCS of soil products (Xing et al., 2018). Xing et al. (2020) cured the FA-MK blended soil-cement concrete blocks for 7 days, and they concluded that the increase in the ratio between MK and FA could significantly improve the structural compactness of soil-polymer concrete. Another research also came to the same conclusion. Zhang et al. (2014a) focused on the macro strength and microstructure of cement-stabilized Lianyungang marine clay mixed with MK, and they stated that the UCS of soil products containing 3 wt.% and 5 wt.% MK could be improved 2-3 times compared with that of materials without MK.

2.2.7 Alkaline activators

The early strength of the soil products treated by OPC grows slowly, and it is greatly limited during the application of OPC. Nevertheless, adding some alkaline activators, such as alkali hydroxides, sulfate additions, and sodium silicate can ameliorate this problem (Xu et al., 2018). Sodium silicate can supply Si elements to promote the reaction ultimately between OPC and soil. Sulfate additions such as calcium sulfate and sodium sulfate can promote the process of hardening cemented soil products. Nevertheless, Rica et al. (2016) tested the mechanical properties of soil products treated with chloride, phosphate, and nitrate salts. They concluded that the soil products presented swelling and heaving, inducing volumetric stability failures on the soil products treated with sulfate salts. Therefore, the volume change of the contaminated soil products containing sulfate salts should be paid attention to. At this point, some researchers have been focused on exploring the stabilizer to deal with sulfate-rich soil. For instance, Zhang et al. (2015) used metakaolin-based geopolymer (MKG) to stabilize a synthetic lean clay and found that MKG could improve the mechanical properties of sulfate-rich contained soil products.

Considering the results above, alkali hydroxides such as $Ca(OH)_2$, NaOH, and KOH may be the best choice to combine with OPC without introducing other additives to inhibit the expansion of the soil products. The difference among these three alkali hydroxides is cations, which make the effects of UCS vary. In the studies of alkali hydroxide usage for S/S, K ions bounded to $Al(OH)_4^-$ favored forming large silicate oligomers, resulting in faster hardening of cementitious materials (Bondar et al., 2011; Xu and Van Deventer, 2000). On the other hand, Na and Ca ions showed potential for binder strength, while adding Ca ions further promoted the system strength (Xu et al., 2018). Moreover, adding Na-based alkaline activators could generate sodium aluminosilicate hydrate (NASH) gel coexisting with CASH gel to enhance the bonding of soil product constituents (Saloni et al., 2021). However, the content of Ca significantly influences the existence of NASH gel by replacing Na with Ca to form calcium sodium aluminate silicate hydrate (CNASH) gel (Saloni et al., 2021). The formation of CASH gel has precedence over NASH gel in a calcium-rich environment (pH over 12) (Garcia-Lodeiro et al., 2011).

2.3 Selection of materials combining OPC

Adding some alkaline activators can generate extra hydration products to improve the stabilization efficiency of HMs. For example, extra $Ca(OH)_2$ can be combined with RM and MK, which lack Ca content to produce more CSH gels. Adding waste resources with rich mineral content can not only make full use of waste resources but also improve the strength of soil products. How to choose the appropriate waste resources mentioned above has been a concern. Numerous studies mentioned above have discussed that alkaline waste resources such as RM, SR, PFA, and GGBS could immobilize HM ions by forming alkaline hydroxide precipitation. Thus, HM ions, such as Pb^{2+}, Zn^{2+}, Cd^{2+}, Cr^{3+}, and Cu^{2+}, could be well immobilized by precipitation synthesis. Moreover, SR and GGBS with SO_4^{2-} can synthesize more AFt and AFm, so they are theoretically suitable for HM ions (such as CrO_4^{2-} and AsO_4^{3-}) which AFt and AFm adsorb. For HM ions (such as Mn^{2+}) not affected by AFt and AFm in the S/S process, other wastes (except for SR and GGBS) are more suitable to be selected in S/S technology.

In contrast, high pH is not conducive to immobilizing HMs such as AsO_4^{3-}, and re-dissolution of HM precipitation would happen. In this case, PFA, GGBS, MK, and ISSA which have low pH, can be used to immobilize HM ions or added to the OPC-based binders to appropriately reduce pH without affecting the pozzolanic reaction. GGBS has a wide range of pH values according to different collection sites. In addition, GGBS has superior performance in resistance from freeze-thaw, chloride, and sulfate attack (promotion of $Mn_6(CO_3)_2(OH)_7(Cl, OH)$ and $Mn_2(OH)_3Cl$), so it is better to be applied to areas affected by acid rain and temperature.

Nevertheless, some noticeable problems should be solved before applying these alkaline wastes to contaminated soil. For example, besides the main components mentioned in 2.1.1, some other toxic HMs (As, Pb, Cd, Ni, Cu, Zn), non-metallic elements (F, Se), and soluble salts also exist in the SR, which result in the increased risk of extra HM leaching and soil salinization (He et al., 2020; Wang et al., 2020e; Zhao et al., 2020). Likewise, the existence of a massive amount of Al (exist as the toxic form $[Al(OH)_4]^-$) in RM has the risk of releasing into the soil products. In addition, complex mixture precipitation may accumulate and form an impervious coating resulting in the interference of the hydration to reduce the generation of gels (Wang et al., 2015; Wang et al., 2018c). Moreover, persistent organic pollutants contained by PFA (such as dioxins, polycyclic aromatic hydrocarbons, furans, and sulfates) need to be overcome in the treatment before using it in the S/S process (Zhang et al., 2021c).

3. The mechanism adsorbing HMs in soil products

3.1 Soil and OPC-based binders

The soil composition resembles OPC-based binders, which were investigated by Liu et al. (2020). They exhibited that the main compositions in soil are 77.9 wt.% SiO_2, 16.5 wt.% Al_2O_3, 6.7 wt.% Fe_2O_3, and other metallic oxides less than 2.5 wt.%. However, OPC and soil are complex systems making the efficiency of S/S different. Guo et al. (2017) asserted that the efficiency of S/S is associated with the phase transformation of HMs, variability of the soil, and the equilibrium of the solution.

3.2 Mechanism of pozzolanic products synthesis

3.2.1 CSH, CAH, and CASH

The hydration product gel from OPC is a mixture of crystalline minerals in disordered phases

(Wang et al., 2020b). With the reaction between soil and OPC-based binders, the spherical particles mainly composed of silicon dioxide in soil products and OPC-based binders dissolve gradually, increasing the concentration of free silicon dioxide (Liu et al., 2018a). Meanwhile, calcium-based compounds dissolve in bulk H_2O, which generates a large number of Ca ions. This hydration reaction makes the cement particle's surface form a C-S-H membrane (Yousuf et al., 1995). The membrane inside exists cement grains, lots of Ca ions and silicate ions, and exists an amount of water outside. This makes the osmotic potential on both sides of the membrane different, causing the outward migration of Ca ions and silicate ions and the inward migration of bulk water molecules (Yousuf et al., 1995). In this proceeding, $Ca(OH)_2$ is formed inside the membrane, and more SiO_4^{4-} generated by cement particles adheres to spherical particles. As the reaction time goes by, the exchange of internal and external ions generates pressure to rupture and reform the membrane (Yousuf et al., 1995). Zha et al. (2019) and Zha et al. (2020) stated that the pozzolanic reactions between Ca^{2+}, SiO_2, and Al_2O_3 occurred in an alkaline environment generating CSH, CAH and CASH, and the reactions developed fully with increasing curing time. The formulas for the hydration reaction producing CH were studied by Wang et al. (2020d), and they are shown in formulas (1) and (2).

$$3CaO\cdot SiO_2+nH_2O=xCaO\cdot SiO_2\cdot(n-3+x)H_2O+(3-x)Ca(OH)_2 \tag{1}$$

$$2CaO\cdot SiO_2+nH_2O=xCaO\cdot SiO_2\cdot(n-2+x)H_2O+(2-x)Ca(OH)_2 \tag{2}$$

This process increases the pH of the soil products combined with OPC-based binders interaction as follows formula (3).

$$Ca(OH)_2\rightarrow Ca^{2+}+2(OH)^- \tag{3}$$

The high pH causes the reaction bounding Si and Al compounds with hydroxide ions with the following equations (4) and (5) (Ouhadi et al., 2021).

$$Al_2Si_4O_{10}(OH)_2\cdot nH_2O+2(OH)^-+10H_2O\rightarrow 2\{2Al(OH)_4^-+4H_4SiO_4\}+nH_2O \tag{4}$$

$$2H_4SiO_4\rightarrow 2H_3SiO_4^-+2H^+\rightarrow 2H_2SiO_4^{2-}+2H^+ \tag{5}$$

Then the ions of Si and Al compounds interact with Ca ions released from CH in the alkaline environment as follows in formulas (6) and (7) (Ouhadi et al., 2021).

$$H_2SiO_4^{2-}+Ca^{2+}+2OH^-\rightarrow CSH\{3CaO\cdot 2SiO_2\cdot 3H_2O\} \tag{6}$$

$$Al(OH)_4^-+Ca^{2+}+2OH^-\rightarrow CAH\{3CaO\cdot Al_2O_3\cdot Ca(OH)_2\cdot 12H_2O\} \tag{7}$$

It is reasonable to hypothesize that the formation of CASH is produced by silica ions, alumina ions, and CH as follows in formula (8) (Wang et al., 2020d).

$$H_2SiO_4^{2-}+Al(OH)_4^-+Ca^{2+}+2OH^-\rightarrow CASH\{CaO\cdot Al_2O_3\cdot 2SiO_2\cdot 4H_2O\} \tag{8}$$

In light of these formulas, sufficient Si, Ca, Al element, and alkaline activators are necessary to develop pozzolanic reaction systems. Liang et al. (2020) used municipal solid waste incineration fly ash (MSWIFA) with ratios of 5 wt.% and 10 wt.% to blend cement-stabilized soil products. They found that PFA, which MSWIFA treated, contained abundant silicon elements. Within these silicon elements, the active silica of PFA intensifying the pozzolanic reaction could produce more hydration products CSH. In addition, the previous study by Wu et al. (1990) and Zhang et al. (2018) showed that alkaline activators such as $Ca(OH)_2$, KOH, and NaOH could break the chemical bonds of Si-O and Al-O, which resulted in the formation of low-solubility calcium silicate and calcium aluminate hydrates by increasing the ionic concentrations in the liquid phase. Similarly, Yip and Van Deventer (2003) and Xu et al. (2018) mentioned the dissolved OH^- could break the covalent bonds such as Al-O-Si, Si-O-Si, and Al-O-Al either, which further formatted the amorphous gel by consolidating the binders and soil products.

3.2.2 AFt and AFm

There is much SO_4^{2-} in the soil and some OPC-based binders (Zhang et al., 2021b). These SO_4^{2-} can react with CAH mentioned above to form acicular AFt with the hydrated reaction. AFt is an early hydrated product, and it can solidify HMs in chemical adsorption ways (substitute Al element with As element) (Wang and Wang, 2022). In addition, AFt could directly react with CO_2 to produce calcite to fill pore spaces and improve structural integrity (Malviya and Chaudhary, 2006). However, as the hydrated time increases, the concentration of sulfate decreases, and too much AFt gather with each other, resulting in the gradual transformation to calcium monosulfoalumiante hydrates (AFm), which may decline the strength of soil products as a consequence of less content of insoluble $CaSO_4$ contained as follow formulas (9) and (10). The hydration reaction process is shown in Table 1.

$$3CaO\cdot Al_2O_3\cdot 6H_2O+3(CaSO_4\cdot 2H_2O)+6H_2O\rightarrow 3CaO\cdot Al_2O_3\cdot 3CaSO_4\cdot 32H_2O \quad (9)$$

$$3CaO\cdot Al_2O_3\cdot 3CaSO_4\cdot 32H_2O+2(3CaO\cdot Al_2O_3\cdot 6H_2O) \rightarrow 3(3CaO\cdot Al_2O_3\cdot CaSO_4\cdot 12H_2O) \quad (10)$$

Table 1 Hydration reaction process of cement (Chen et al., 2009; Wang et al., 2020d)

Stages	Reactions
Rapid hydration period	Cement begins to dissolve and produce the hexagonal $Ca(OH)_2$ due to the hydration reaction. CSH, CAH, CASH, and acicular AFt are produced, which fill pores in soil products to strengthen soil products structure
Induction period	AFt, CSH gels, and $CaCO_3$ cover the surface of cement to prevent hydration of cemented material
Accelerated hydration period	OH^- and Ca^{2+} pass through the film due to the membrane rupture with osmotic pressure. The hydration reaction can continue. The cementitious materials begin to set
Slow hydration period	The remaining substances in the system continue to hydrate. At the same time, AFt converse into AFm results in the slow growth of soil product strength

In addition, the formation of the AFt always needs high pH. Within the alkaline environment, Ca ions exist in the form of $Ca(OH)_2(aq)$ combined with $Al(OH)_4(aq)^-$ provided by calcium aluminum sulfate and react with $SO_4(aq)^{2-}$ from gypsum to form stable AFt (Wang et al., 2020a). The formation of AFt can solidify HMs further. Wang et al. (2020a) assessed the potential application of AFt for hazardous waste stabilization. They considered that AFt could stabilize HMs such as Cu, Cr, Cd, Pb, Zn, and Fe by cationic substitution in the lattice. Albino et al. (1996) and Liang et al. (2020) aimed at Cd and Pb to study the effects of the variation of the content of $FeSO_4$ on S/S. They found that the increase of $FeSO_4$ made the leaching concentration of Cr show a downward trend and significantly reduced the leaching concentration of Pb by 67%. However, Shi and Spence (2004) reported that the high sulfate content would deleteriously affect the mechanical properties and immobilization. The process of pozzolanic reaction generating CSH and AFt is shown in Figure 2.

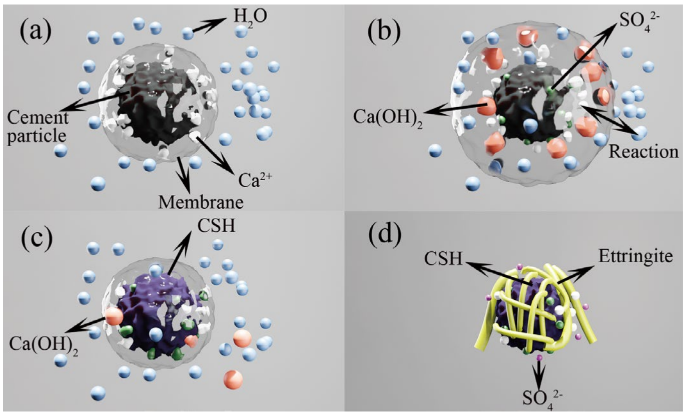

Figure 2 The process of pozzolanic reaction generating CSH and AFt. (a) The formation of the membrane around the cement particle. (b) The synthesis of CH. (c) The generation of CSH. (d) The formation of AFt (Liu et al., 2018a; Liu et al., 2020; Ouhadi et al., 2021; Wang et al., 2020a; Yousuf et al., 1995)

Thus, how to prevent AFt from the formation of AFm in an appropriate proportion is a challenge. Lothenbach et al. (2008) and Liu et al. (2020) suggested that the presence of limestone, which had much calcium carbonate, could form stable monocarbonate instead of monosulfate and inhibit the transformation from AFt to AFm.

3.3 Mechanism of S/S for HMs

The S/S mechanism for HMs mainly include (1) adsorption on the surface, (2) chemical bonding, and (3) physical encapsulation (filling effect), which is shown in Figure 3 (Guo et al., 2017; Wang et al., 2020b). Adsorption means HM ions stick to the surface structure and the pore structure of CSH, CAH, and CASH gels without any chemical reaction. Chemical bonding implies that chemical reactions between HM ions and gels will occur, such as precipitation in the alkaline environment, ion exchange with hydration products, and surface complexation. Physical encapsulation refers to precipitation or complexes filling the pores of soil products and then encased by gels and soil particles. HMs can be classified as amphoteric-type, precipitation-type, and anion-type (Wang and Wang, 2022). Phair and Van Deventer (2001) stated that the primary forms of Pb and Cu in aqueous solutions are $Pb(OH)_2$, $Pb(OH)_3^-$, $Cu(OH)_2$, and $Cu(OH)_3^-$ in the alkaline condition, and the immobilization is mainly dependent on the physical encapsulation. However, the main mechanism of different HM ions with different binders varies. For instance, Conner and Hoeffner (1998a), Liu et al. (2017), Zha et al. (2019), and Wang et al. (2021b) stated that the S/S mechanism for Pb^{2+} in contaminated soil products is mainly dependent on adsorption onto hydrated gels, whereas the S/S mechanism for Zn^{2+} could be precipitated or complexed in the alkaline environment as $Zn(OH)_2$, $ZnCO_3$, ZnO, $Zn_3(PO_4)_2{\cdot}4H_2O$ and $CaZn_2(PO_4)_2{\cdot}2H_2O$. Similarly, Cr^{3+} could be stabilized in the soil products by forming insoluble Ca-Cr oxide and hydroxide nitrate complex precipitation such as $Ca_2Cr(OH)_7{\cdot}3H_2O$ and $Ca_2Cr_2O_5{\cdot}6H_2O$ (Dermatas and Meng, 2003; Jing et al., 2006; Kindness et al., 1994; Omotoso et al., 1998).

Simultaneously, these precipitations could be encapsulated during the hydration process or adsorbed onto the hydrated gels to degrade mobility. During the S/S process, water-insoluble is the primary substance encapsulated by OPC-based binders (Ji and Pei, 2019). In contrast, adsorption and chemical bonding process are the main mechanism for stabilizing dissolved HM ions. Generally, the synergistic effects always appear in the immobilizing HMs process. That is, multiple S/S mechanism occur in the pozzolanic process at the same time.

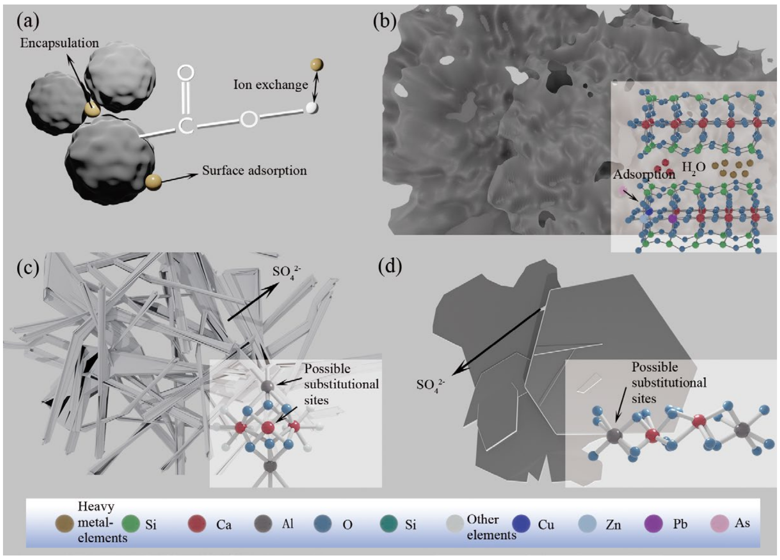

Figure 3 The mechanism of immobilizing heavy metal ions and the structure of gels. (a) Muti-mechanisms. (b) CSH. (c) AFt. (d) AFm. (Guo et al., 2017; Moon et al., 2008; Phair and Van Deventer, 2001; Urushihara et al., 2022; Wang et al., 2020b)

In addition, not only hydrated products (CSH, CAH, CASH, and CH) could immobilize the HMs, but also the iron oxides/hydroxides contained in OPC-based binders could adsorb or co-precipitate the HMs. Dixit and Hering (2003) and Li et al. (2018) compared As(III) and As(V) sorption onto amorphous iron oxide (HFO), goethite, and magnetite in various pH environments. They found that when pH>10, the adsorbed As(V) had high immobilization properties on these iron oxides/hydroxides. Similar research was also presented by Wang et al. (2020a), and they concluded that the binders which contained RM and OPC at the portion of 3:1 have more excellent immobilization ability on Pb compared to Cu and Zn due to the active adsorption sites on the surface of aluminum and iron oxides combining with soluble and exchangeable Pb.

The immobilization of HM ions is influenced by the hydrated reaction above. In contrast, HM ions can also impact the hydrated reaction. When Ni^{2+} and Cu^{2+} existed in the contaminated soil, they would react with CH to form amorphous or insoluble compounds such as $CaNi_2(OH)_6 \cdot 2H_2O$ and $Cu(OH)_2$, which generated precipitation encapsulating reaction substances to hinder the hydration process (Wang et al., 2020d). On the other hand, the reaction between HMs and CH would reduce the alkalinity of soil products, whereas alkalinity plays an essential role in gel synthesis. Table 2 summarizes the S/S mechanism of various HMs during the hydration process.

Table 2 Mechanism of Solidification/Stabilization (S/S) for heavy metals (HMs) during hydration process through semi-quantitative analysis (Wang and Wang, 2022; Wang et al., 2019b; Wang et al., 2019d)

Types of HMs	HM ions	Main S/S mechanism
Amphoteric-type	Pb^{2+}	At intermediate pH (pH=9), HM ions precipitate as

Precipitation-type		hydroxides, such as $Pb(OH)_2$, $Pb_2(OH)_2CO_3$, and $PbCO_3$. Furthermore, the hydroxides such as $Pb(OH)Cl$ hardly appear in the AFm but weakly exist in the AFt. However, the precipitates partially redissolve to form $Pb(OH)_3^-$ at higher pH and Pb^{2+} directly sorbed by CSH through physical adsorption by its high specific surface area and its layered structure and chemical adsorption by the formation of Si-O-Pb. It is noteworthy that Pb^{2+} has no effects on the structure of AFt and AFm, so it can be inferred that only physical adsorption exists between Pb^{2+} and AFt.
	Zn^{2+}	Zn^{2+} precipitates as $Zn(OH)_2$, $ZnCO_3$, ZnO, $Zn_3(PO_4)_2\cdot 4H_2O$ and $CaZn_2(PO_2)_2\cdot 2H_2O$. Furthermore, Zn^{2+} exchanges with the Ca^{2+} in C-S-H to form Si-O-Zn.
	Cr^{3+}	Cr^{3+} precipitates as $Ca_2Cr(OH)_7\cdot 3H_2O$ and $Ca_2Cr_2O_5\cdot 6H_2O$. Then these precipitates are encapsulated by CSH.
	Mn^{2+}	Mn^{2+} precipitates as $Mn(OH)_2$ in the alkaline environment. $Mn_6(CO_3)_2(OH)_7(Cl, OH)$ and $Mn_2(OH)_3Cl$ are generated due to the existence of Cl^- and carbonation. The S/S of Mn^{2+} is irrelevant to the AFt and AFm.
	Cd^{2+}	Cd^{2+} precipitates as $CdCO_3$, CdO, and $Cd(OH)_2$.
	Cu^{2+}	Cu^{2+} exchanges with the Ca^{2+} in C-S-H to form Si-O-Cu.
Anion-type	CrO_4^{2-}	CrO_4^{2-} precipitates as $Ca_4Fe_2CrO_{10}\cdot 14H_2O$ and $Ca_5(CrO_4)_3OH$. Cr(VI) is mainly chemically sorbed by AFm and hardly sorbed by AFt and CSH.
	AsO_4^{3-}	AsO_4^{3-} precipitates as $Ca_3(AsO_4)_2$, $CaHAsO_3$, $Ca_4(OH)_2(AsO_4)_2\cdot 4H_2O$ and $NaCaAsO_4\cdot 7.5H_2O$ (main mechanism). Physical adsorption happens on the surface of C-S-H, and chemical adsorption happens on AFt (As substitute S) and AFm (As substitute Al).

Although OPC-based binders can utilize wastes, the effects on the leaching concentration and strength of soil products differ with various OPC-based binders. To evaluate the degree of S/S performance, the standards of different countries in physics and chemistry are given in Table 3. Table 4 summarizes recent studies on leaching concentration and UCS effects of HMs in S/S.

Table 3 Physico-chemical quality standards in different countries for Solidification/Stabilization (EPA, 1992; Epd, 2011; Malviya and Chaudhary, 2006)

Physical properties	
Hardening time	2-5 days
Compressive strength	
US EPA 2013 landfill disposal standard	350 kPa
The strength requirement of the environment Canada WTC	440 kPa
Criterion for on-site fill material use in Hong Kong	1.0 MPa
UK environmental agency for land-filling	1.0 MPa

UKEA limit	1.0 MPa
France’s criteria for the deposit of stabilized waste in hazardous waste	1.0 MPa
ACI criteria of low strength filling materials	1.15 MPa

Chemical properties (TCLP limits)

Parameters	Universal treatment criteria for on-site reuse (mg/L)	Hazardous waste criteria (mg/L)	Landfill disposal criteria (mg/L)
Pb	0.75	5	50
Zn	4.3	5	250
Cd	0.11	1	10
Cr	0.6	5	50
Cu	-	-	250
As	5	5	50
Ni	11	-	250

Table 4 The recent research about experiments of OPC-based binders in heavy metal immobilization

Heavy metal species (initial concentration)	Proportion of binders	Curing condition	Leaching concentration	Summary of research results	Ref.
Zn (1000, 5000, 10000 mg/kg)	Cement 50 wt.%, soda residue 50 wt.%	Cured at 20°C 28d	0.233-0.642 mg/L	The acidic environment made hydrogen ions react with the internal substances of filled soil products, which led to the integrity loss of structural	(Zha et al., 2021)
Pb, Zn, Cr (10000 mg/kg)	Cement 25 wt.%, fly ash 75 wt.%	Cured at 22±1°C 0, 7, 28, 90d	Pb: 15-20 mg/L Zn: 10-15 mg/L Cr: 0.15-0.3 mg/L	For the strength characteristic, the most notable detrimental effect on remediation was induced by Cr^{3+}, whereas the least was induced by Pb^{2+}. For the leaching characteristic, the trend was reversed	(Zha et al., 2019)
As (>400 mg/kg)	Cement 80 wt.% and 50 wt.%, fuel ash (FA), furnace bottom ash (FBA) and ground granulated blast furnace slag (GGBS) 20 wt.% and 50 wt.%, respectively	Cured at 60±2°C 7d	0.02-0.1 mg/L	Partial OPC replacement by FA, GGBS, and FBA showed the corresponding decline in the compressive strength	(Li et al., 2017)
Cu, Zn (Cu: 74.04 mg/kg, Zn: 154.72 mg/kg)	Sewage sludge ash (ISSA) at 0.2 mass ratio and cement/ lime at 0.05/ 0.1 mass ratios	Cured at 23±2°C 3, 7, 14, 28d	Cu:0.075-0.3 mg/L Zn:0.01-4.00 mg/L	Lime can be used to replace ordinary portland cement (OPC) for better HM immobilization and carbon emission reduction	(Li et al., 2021b)
Pb, Zn, Cu (2500, 5000,10000, mg/kg)	Cement 10-100 wt.%, red mud 40-100 wt.%, phosphogypsum 10-70 wt.%, fly ash 30-70 wt.%	Cured at 20±0.5°C 7, 14, 28d	Cu: 1.5mg/L Pb: 1mg/L Zn: 3mg/L (minimum)	The addition phosphogypsum significantly imcreases the strength of the soil products and the main mechanism of stabilizing HMs is the adsorption of red mud and ettringite, and ion exchange by Ca or Al in ettringite	(Wang et al., 2020a)
As	Lime dust 5-15 wt.%, cement kiln	Cured	0.0222-0.999	20 wt.% CKD and 5 wt.% lime dust have the optimized	(Eisa et

(6.3 ppm)	dust (CKD) 15-30 wt.%	temperature is not given, 28d	ppm	correlation between strength and acceptable level of arsenic leakage in the TCLP test	al., 2020)
Cr (1000, 5000, 10000 mg/kg)	Cement 20 wt.%, 30 wt.%, 40 wt.%, 50 wt.%, soda residue (SR) 80 wt.%, 70 wt.%, 60 wt.%, 50 wt.%, respectively	Cured at 22±1°C 7, 14, 28, 56, 90d	0.10-11.9 mg/L	The increase of the SR could improve soil product strength and decrease the concentration of Cr^{3+}	(Zha et al., 2020)
As (201 mg/kg)	Cement 80 wt.%, red mud (RM), blast furnace slag (BS), metakaolin(MK) 20 wt.%	Cured at 23±1°C 7, 28d	0.1-0.5 mg/L	The RM-incorporated binder can be used for low-cost and low-carbon S/S treatment, whereas MK-incorporated binder is superior in mechanical strength	(Wang et al., 2019c)
Ni, Cu, Zn, Pb (Ni:1200 mg/kg Cu: 1500 mg/kg Zn: 1600 mg/kg Pb:2500 mg/k)	Cement 100 wt.%, 50 wt.%, 33 wt.%, ground granulated blastfurnace slag (GGBS) 0 wt.%, 50 wt.%, 0 wt.%, pulverised fly ash(PFA) 0 wt.%, 0 wt.%, 67 wt.%, respectively	Cured at 20±2°C 28d	Ni: <0.003 mg/L Cu: 0.01-0.02 mg/L Zn: 0.01-0.02 mg/L Pb: 0.01-0.03 mg/L	The addition of GGBS and PFA can decrease both of strength of soil products and the concentration of HMs	(Wang et al., 2019a)
Zn (0, 1000, 3000, 5000 mg/kg)	Cement 75 wt.%, 60 wt.%, slag 25 wt.%, 40 wt.%, respectively	Cured temperature is not given, 7, 14, 28d	16-296.4 μg/L	15 wt.% cement + 10 wt.% slag has a better effect on solidification/stabilization	(Liang et al., 2018)
Pb, Zn, Cd (5000 mg/kg)	Red mud(R), phosphogypsum(P) and Portland Cement (PC) with ratio of 4:1:2	Cured at 20±0.5°C 7, 28d	Pb: 0.12-8 mg/L Zn: 0.5-7 mg/L Cd: 0.8-9 mg/L	At least 15 wt.% of RPPC binder should be added for the treatment of soil products with 5000 mg/kg of Pb, Zn, and Cd co-contamination to limit the metal leachability	(Wang et al., 2021a)
Cr, Pb	Cement 50 wt.%, 66.6 wt.%, 60	Cured at	Cr: 0.2325-3.619	The strength of cement-stabilized soil products can be	(Liang

(unmentioned)	wt.%, 75 wt.%, 80 wt.%, municipal solid waste incineration fly ash(MSWIFA) 50 wt.%, 33.4 wt.%, 40 wt.%, 25 wt.%, 20 wt.%, respectively	25±2°C 7, 14, 28d	mg/L Pb: 0.0002-2.995 mg/L	maintained by adding PFA as a substitute for OPC when reducing the content of OPC. Moreover, an apparent effect of strength improvement could be achieved only when the contents of OPC is similar to that of PFA (within 5 wt.%)	et al., 2020)
Pb (800, 5000, 50000 mg/kg)	10 wt.%, 20 wt.%, 30 wt.%, and 40 wt.% soda residue (SR) / cement rates with total contents of 7.5 wt.%, 10 wt.%, 15 wt.%, and 20 wt.%	Cured at 20°C 1, 3, 7, 14, 28, 56, 90d	Pb: 1.87-4.16 mg/L	The addition of SR reduced Pb concentration by 3.28 times that in single cement. The addition of SR improved the USC within 20 days. However, it declined the later UCS of the cement-cured soil products.	(An et al., 2022)

TCLP: Acetic acid, pH = (2.88±0.05), temperature = (23±2) ℃, vibration time = (18±2) h, vibration velocity = (30±2) rpm, liquid-solid ratio = 20:1 (L/kg)

4. Factors affecting UCS

4.1 Concentration of HM ions

The existence of HM ions not only influences the leaching concentration but also has effects on the strength of soil products. Wang et al. (2020d) found that Ni and Cu ions were relative to the UCS and impermeability of the soil products, and they considered that the formation of amorphous or insoluble substances such as $CaNi_2(OH)_6 \cdot 2H_2O$ and $Cu(OH)_2$, which encapsulated binders to hinder hydrated reaction, is responsible for the strength lag of growth of the soil products. On the other hand, the alkalinity of soil products would also decrease due to the consumption of OH^- between HM ions and CH. The alkalinity is closely related to the production of gels, which can bind with soil products, so the decline of pH would decrease soil product strength. HM ions could also affect the integrity of soil products through the replacement chemical reaction.

Concerning the soil contaminated by Cr, Zha et al. (2019) and Sophia et al. (2010) concluded that the Si^{4+} ions and Ca^{2+} ions in CSH, Al^{3+} ions in CAH, and Al^{3+} ions in AFt, which were the main elements in the substances supporting the microscopic structure of the soil products, could be replaced by HM Cr^{3+} ions deteriorating the complete soil products structure. However, HM ions are not always having negative effects on UCS. Liu et al. (2018a) stated that the UCS of solidified soil products would increase in low concentration of HM ions and decrease in the high concentration of HM ions. At the Zn^{2+} concentration of 1000 mg/kg, Zha et al. (2021) observed a small amount of rhomboid calcium hydroxy-zincate crystals of which the chemical formula is $Ca[Zn(OH)_3]_2 \cdot 2H_2O$ formed. The reaction formulas are as follows (11), (12), (13), (14), and (15) (Lasheras-Zubiate et al., 2012; Yousuf et al., 1995; Zha et al., 2021)

$$Zn^{2+}+2OH^- \rightarrow Zn(OH)_2 \tag{11}$$

$$Zn(OH)_2+OH^- \rightarrow Zn(OH)_3^- \tag{12}$$

$$Zn(OH)_2+2OH^- \rightarrow Zn(OH)_4^{2-} \tag{13}$$

$$Zn(OH)_3^-+Ca^{2+}+H_2O \rightarrow Ca[Zn(OH)_3]_2 \cdot 2H_2O \tag{14}$$

$$Zn(OH)_4^{2-}+Ca^{2+}+H_2O \rightarrow Ca[Zn(OH)_3]_2 \cdot 2H_2O \tag{15}$$

At the same time, the structure of the soil products became denser with tiny pores, which increased the soil product strength (Zha et al., 2021). In another study, within the low concentration of HM ions, Pb^{2+} could substitute Ca^{2+} from hydrated products and accelerate the hardening of soil particles to increase strength (Liu et al., 2018a). This could also explain the reason for the increase of UCS at the initial time. There is a similar report suggesting that the formation of $CaZn_2(OH)_6 \cdot H_2O$ could be produced under the low concentration of Zn^{2+} (<1000 mg/kg) (Ouhadi et al., 2021). Conversely, with the high Zn^{2+} concentration from 5000 mg/kg to 15000 mg/kg, the destruction of the structure of the soil products due to excess internal cracks appeared, accounting for the decrease of UCS (Liu et al., 2017; Mollah et al., 1992; Yousuf et al., 1992).

Overall, with the increase in curing time, the strength of soil products tends to increase first and then appears to decrease (Wang et al., 2020a). The change in soil product strength probably comes from hydration resistance, causing the strength lag of growth of soil products in the hydrated reaction (Wang et al., 2020d; Xu et al., 2018). Another reason for reducing the UCS may be the transition from AFt to AFm, causing the intensity to increase in the first 7 days and decrease after 14 days.

4.2 Proportion of OPC-based binders

Higher OPC blended with mineral admixtures could make soil products form a more compact structure and increase soil product strength (Subalakshmi et al., 2012). Li et al. (2017) used FA, GGBS, and FBA to replace the partial OPC and found a corresponding decline in the UCS, which showed the inferiority of the OPC-based binders to single OPC on UCS. Nevertheless, the addition of OPC-based binders is not harmful to the strength of soil products. Liu et al. (2018a) used FA and SR to solidify the soil, and they found that adding these binders could improve soil product strength. The other study also observed that the strength of the soil products increased gradually as the amount of lime increased (Wang et al., 2020a). Nwaubani (2018) stated that superior quality performance appeared when replacing OPC with waste in less than 10 wt.%.

Moreover, the substances with small particle sizes in soil products, such as silica fumes, could fill the pores created by free water and then refine the microstructure of soil products to increase the strength for engineering applications (Chen et al., 2021; Wang et al., 2020b). In addition, some complexes and gel cementation generated by complex physicochemical reactions, such as $CaSiO_3 \cdot CaCO_3 \cdot Ca(OH)_2 \cdot nH_2O$ complex, could also aggregate to improve soil product strength (Zha et al., 2021). The reaction formulas are as follows (16) and (17) (Sun et al., 2012; Zha et al., 2021)

$$CaCO_3 + Ca(OH)_2 \rightarrow CaCO_3 \cdot Ca(OH)_2 \quad (16)$$

$$CaCO_3 \cdot Ca(OH)_2 + SiO_2 + 2OH^- + H_2O \rightarrow CaSiO_3 \cdot CaCO_3 \cdot Ca(OH)_2 \cdot nH_2O \quad (17)$$

Figure 4 summarizes the relevant research on the highest strength performance with different addition contents of OPC-based binders of S/S products (the UCS value is also related to the strength grade of the cement used in the experiment).

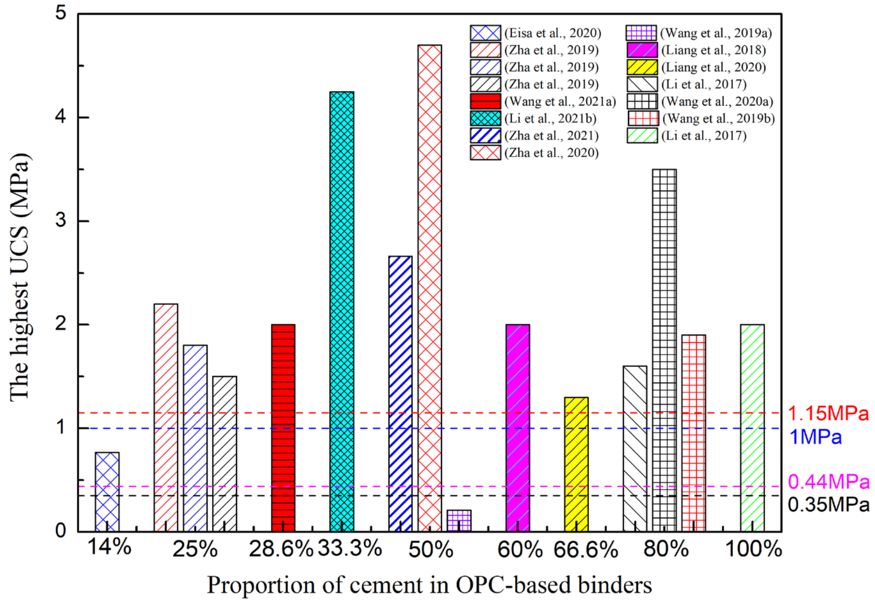

Figure 4 The highest strength performance with different addition contents of OPC-based binders of Solidification/Stabilization specimens

4.3 pH

The reaction between OPC-based binders and soil would influence the alkaline environment in the soil products. The low pH often fails to meet the demand for forming AFt (Wang et al., 2020a). The alkaline environment with high pH could make more Al^{3+} and Si^{4+} ions in the pore solution to participate in the pozzolanic reaction, producing more additional CSH, CASH gel, and AFt, propping up the skeleton structure of soil products (Liu et al., 2017; Shi and

Fernandez-Jimenez, 2006; Slaty et al., 2015; Wianglor et al., 2017). Liu et al. (2017) used 1.0 mol/L NaOH solution to immerse the soil products, where the masses of the solution were three times higher than the content of soil products, and they found the UCS of the soil products increased significantly when the soil pH value remained above 10.5 after soaking in the NaOH solution. Zha et al. (2013) found that the pozzolanic reactions would be strengthened in the alkaline environment, and the formation of precipitation products, such as Ca-Cr oxide and hydroxide nitrate complexes, would influence the soil product strength (Dermatas and Meng, 2003; Omotoso et al., 1998; Zha et al., 2020). Adding RM could increase the pH of the soil products, while some HMs would increase the mobility in the high pH condition. Thus, adjusting the pH when adding activators to the soil products is crucial. Wang et al. (2020a) mixed RM and phosphogypsum in different ratios as binders to solidify soil, and they found that incorporating phosphogypsum could reduce the pH of RM. In addition, when the amount of phosphogypsum reached 50 wt.%, the pH of the RM dropped from 12 to less than 8 (Wang et al., 2020a).

4.4 Chemical compounds in the soil texture

The high concentration of Na^+ and K^+ ions in soil products could decrease the thickness of the electric layer by exchanging with Ca^{2+}, which could shorten the distance between the particles to form a dense structure (Zhang et al., 2018). Cl^- ions would impede the S/S treatment by combining H^+ from acid rain to form HCl. The content of hydrocarbons would also affect the UCS (Zha et al., 2021).

4.5 Moisture content and curing time

The strength of the soil products would increase as the curing time increases due to the formation of more gels and other chemical compounds mentioned above. In addition, the ratio of water/cement (w/c) would also affect the fluidity of the S/S system. The difference in w/c affects UCS from volume shrinkage, bulk density, changes in soil pH, and curing time (Zhang et al., 2021a). Wang et al. (2019a) set the two ratios of w/c, which were 0.5:1 and 1:1. The 0.5:1 w/c group showed better performance on the strength of the soil products. Previous findings demonstrated that the key to solidifying soil is controlling the moisture content. When adding FA to OPC, the water demand of the mix increased, and a decrease in the soil product strength was observed with the increases in the FA to OPC ratio (Lombardi et al., 1998). The reason could be explained that the position occupied by the original water changed to voids and pores.

With the increase of water, the position of pores increased, which resulted in the sparse soil products structure decreasing the strength of the soil (Cao et al., 2020; Ma et al., 2014). Another research indicated that the higher w/c ratio promoted the dissolution of minerals and accelerated the release of HMs, decreasing soil product strength (Quina et al., 2011). In contrast, the low moisture content would cause incomplete dissolution of Ca ions and weaken the degree of hydration reaction due to undissolved reactants. Generally, the waste/binder ratio of 0.4-0.5 is used with a water/solid ratio of 0.4-0.6 (Malviya and Chaudhary, 2006). In addtion, solidified specimens are expected to harden during 2-5 days (Malviya and Chaudhary, 2006).

5. Prospect

5.1 Unification of standard of using waste materials on S/S

The compositions of industrial wastes in different places significantly vary from treatment methods (Zhu et al., 2020). It is possible that the environmental toxicity and the mechanical properties may not be stable even under the same experimental synthesis process conditions.

Moreover, there are wide variations in physical properties, such as inter-particle and intra-particle speciation, which cause heterogeneous distribution on the surface (Rickard et al., 2011; Zhang et al., 2014b).

Until now, there is no reliable quality reference standard for adding waste resources due to the diversity of raw waste materials. The reaction between diversity compositions in waste materials and soil is also unknown. Therefore, it is challenging to apply experimental results to the promotion and application with standardizing the quality control technology in reality. The influence of different hydration products on the S/S effects and how to control the quality of waste materials with different compositions to achieve the same effects is also worth exploring.

5.2 Effects on long-term HM stability

The variability and the complexity of the environmental system, primarily caused by the freeze-thaw (F-T) and dry-wet (D-W) cycle, acid rain, and alkaline groundwater, greatly influence the long-term stability of HMs (Wang et al., 2016; Wardhono et al., 2017; Yang et al., 2020). Previous studies stated that the combination of various raw waste materials did not show better performance in apparent porosity after 250-450 times freeze-thaw cycles (Borosnyoi, 2016). Yang (1993) studied the F-T and W-D durability with blast furnace slag and cement. The report stated that the performance of 20 wt.% replacement of the cement was better than that of 40 wt.% replacement and the F-T condition affected UCS more adversely than the D-W condition (Yang, 1993). The F-T cycles could cause a decrease in the stable form content of HMs in the soil products. The main decreased stable form is the carbonate-bound state content (Yang et al., 2022).

In addition, acid rain and alkaline groundwater could result in pH changes. The environmental pH tremendously influences long-term HMs stability. Chemical resistance (NaOH, NaCl, H_2SO_4, and HCl) could be considered in the environment to simulate various environmental conditions, and some indexes such as effective diffusion coefficient (De), cumulative fraction of leached substance (CFL), and leachability index (LX) could be used to reflect the long-term stability of HMs (Al Menhosh et al., 2018). Du et al. (2014) performed semi-dynamic leaching tests by simulated acid rain with Pb-contaminated kaolin soil and cement. Part of the CH and CSH gradually dissolved when the soil products were exposed to the acid environment, which caused the acid buffering capacity of the soil matrix to decrease. The change of De was small as the pH ranged from 4.0 to 10.0. However, the De increased rapidly when the pH decreased from 4.0 to 2.0. They considered that a threshold pH existed between 2.0 and 4.0 (Du et al., 2014). Wang et al. (2016) came to a similar conclusion. There was no noticeable CFL change of Pb within the pH range from 5.00 to 10.00. However, the strong acid condition significantly influenced the leaching behaviors of Pb. De and LX indicated that S/S disposal could be considered appropriate for "controlled utilization" in the environment with a pH ranging from 3.65 to 10.00 (Wang et al., 2016).

Regarding long-term stability evaluation, the mathematical model (nonlinear regression analysis and kinetic leaching) can be constructed by analysis software such as Python or Matlab (Zhang et al., 2021b). A feedback mechanism is advised to be established to optimize the environmental conditions and improve the long-term HMs stability.

5.3 Competitive adsorption of HM ions

Competitive adsorption exists among various HM ions. Andrejkovicova et al. (2016) stated that the adsorption of geopolymer for HM has a sequence, which showed the order $Pb^{2+}>Cd^{2+}>Cu^{2+}>Zn^{2+}>Cr^{3+}$. Ji and Pei (2019) thought the reason for this regularity might be from variation in free bonds available for complexation, the activity of HM ions, pore size distribution on geopolymer surface and hydrated ions, and free energy of hydration. Another research discussed by Jin et al. (2011) considered that the cation substitution might be related to the ionic radius. For instance, the ionic radius of Na^+, Pb^{2+}, and K^+ are 0.095, 0.119, and 0.138 nm, respectively. The Pb^{2+} can easily substitute the position of Na^+ and K^+ due to its medium ionic radius, which causes an excellent immobilization effect of Pb^{2+}. However, Ji and Pei (2019) indicated that analogous ions could substitute the Al^{3+} position with a close ionic radius to Si, such as Cr, Mn, and Fe.

The competitive adsorption sequence and selectivity mechanism remain unclear in the case of soil products contaminated by multiple HMs so far. For active adsorption sites on the soil product surface, which S/S agents add, the mathematical models (such as Langmuir, Freundlich isotherm equation, and kinetic equation) can be applied to the adsorption process to judge the types and priority of adsorption reaction.

5.4 Novel effective materials applied in the field

As so far, phosphorous materials (mainly for Pb), ferrous materials (mainly for As), clay, and oxidant and reductant (mainly for As(III) and Cr(VI)) are still the mainstream materials to improve OPC. Wang et al. (2018c) used magnesium potassium phosphate cement (MKPC) which was compared with OPC to immobilize Pb of contaminated soil. They found that the final setting time of the MKPC (16 min) was significantly less than that of the OPC (1278 min) due to exothermic reactions between phosphates and free limes. Because the final setting time is required to be less than 360 min for practical use, phosphates materials can be combined with OPC to shorten the setting time. Moreover, the chemical precipitation of pyromorphite and lead phosphate also showed excellent immobilization performance on the leachability of Pb. However, pure reagents related to phosphate are expensive, so other waste resources (after the detoxification process) containing phosphate can be considered for S/S utilization combined with the wastes introduced in this paper.

Besides, for the disadvantage of interfering with OPC hydration mentioned in section 2.3 and carbon emission, biochar incorporation can be considered to add to OPC-based binders due to the mesoporous structure and adsorption capacity of biochar to control interference. Chen et al. (2022) added carbon-negative rice husk biochar (RBC) and yard waste biochar (YBC) into municipal solid waste incineration fly ash (MSWIFA) and OPC. They used the semi-quantitative analysis method (due to the approximately 5% methodical error from quantitative X-ray diffraction (Q-XRD)) to calculate the content of the respective phases during the S/S process. The conclusion indicated that adding the biochar could strengthen the process of pozzolanic reaction (due to the abundant Si element in RBC) and increase the area of CSH. The increase of the CSH area is the porous property of biochar, which could reduce the influence of HM precipitation on hydration. Therefore, wastes with porous performance, such as biochar, are also worth exploring to be combined with OPC-based binders in the future.

6. Summary and conclusions

Ordinary Portland Cement-based binders have been used as a common material in the Solidification/Stabilization technology for treating heavy metals. However, cement production increases greenhouse gas emissions and consumption of energy requirements. Combining waste resources and Ordinary Portland Cement-based binders to apply into Solidification/Stabilization treatment remediating heavy metal-contaminated soil can not only accomplish CO_2 reduction but also reuse wastes from industries.

Most heavy metal ions can be immobilized by forming precipitation. However, the calcium silicate hydrate, calcium aluminum silicate hydrate, ettringite, and calcium monosulfoalumiante hydrates generated during the pozzolanic reaction can also immobilize heavy metal ions by adsorption on the surface, ion exchange, and encapsulation.

Soda residue, red mud, and alkaline granulated blast furnace slag are suitable for heavy metals, which are mainly immobilized as precipitation in an alkaline environment. Pulverized fly ash, granulated blast furnace slag, metakaolin, and incinerated sewage sludge ash which have low pH, can be used to immobilize heavy metal ions dissolving in the high pH or added to the OPC-based binders to reduce pH appropriately. Al-rich red mud and granulated blast furnace slag can synthesize more AFt and AFm in the environment containing SO_4^{2-}, and they are suitable to treat those heavy metals influenced by AFt and AFm.

The factors affecting the strength of the soil products are: (1) the concentration of heavy metal ions, (2) the proportion of OPC-based binders, (3) pH, (4) chemical compounds in the soil and soil texture, (5) moisture content and curing time.

Unification of the standard of using waste materials on S/S, effects on long-term HM stabilization, competitive adsorption of HM ions, and novel effective material additives (such as phosphorous materials and biochar) deserve further study in the future.

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