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Abdellatif Elghali, Mostafa Benzaazoua, Julien Couvidat, Yassine Taha, Carmen Mihaela Neculita, et al.. Stabilization/solidification of contaminated sediments: challenges and novelties. Low Carbon Stabilization and Solidification of Hazardous Wastes, Elsevier, pp.93-112, 2022, 10.1016/b978-0-12-824004-5.00023-2 . hal-03654380

HAL Id: hal-03654380
https://hal.science/hal-03654380

Submitted on 28 Apr 2022

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Stabilization/solidification of contaminated sediments: challenges and novelties

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Abstract:

Sediments contamination by organic and inorganic pollutants presents a serious risk to human health, fauna and flora. Stabilization/solidification (S/S) is one of the most used technics for the management of contaminated sediments. It aims to improve the mechanical and geochemical properties of contaminated sediments by adding cementitious materials. Cement hydration enhances workability, mechanical resistance, cohesion and decrease the permeability of contaminated sediments. Consequently, the inorganic contaminants (e.g. heavy metals) are physically immobilized by decreasing: i) the available surface area of reactive grains and ii) ~~decreasing~~ the contact time between oxidizing agents and reactive grains (enhancing surface runoff). Moreover, the contaminants are also chemically immobilized by: i) their uptake during cement hydration and ii) their precipitation as low-solubility secondary products (e.g. oxy-hydroxides). However, contaminants can be remobilized if: i) the physical properties of stabilized sediments are altered due to climatic conditions and/or ii) the geochemical conditions become acidic which may lead to the dissolution of the precipitated contaminants-bearing phases. The efficiency of S/S technology may vary from laboratory to field scale.

Key Words:

Contaminated sediments, stabilization/solidification, cement, physical trapping, chemical fixation

1. Introduction

The release of organic and inorganic contaminants into aquatic eco-systems increased is a challenging issue to the environment (human, fauna, flora and water resources). Contaminants may result from human activities (e.g. mining industries) and/or natural rocks weathering [1]. In aquatic ecosystems (e.g. river, lake); sediments are the host of various contaminants depending on their mineralogical properties. Heavy metals release into aquatic ecosystems presents a serious risk to human health through food chain. Heavy metals are characterized by their long term persistence and bioaccumulation causing harmful effects at pints for from their

sources [2]. In aquatic eco-systems, the reactivity of certain minerals, such as sulfides, is limited due to low concentrations of soluble oxygen which is around 8 mg/L. However, contaminants may be released due to dissolution mechanisms. Due to wind and waves energy the sediments, especially their fine fractions, are resuspended. Consequently, the contaminants are released in water. For ~~this~~ these reasons, contaminated sediments need to be properly managed to eliminate risk of contamination. Sediments dredging before their stabilization and/or revalorization is widely used for contaminated sediments management [3-7]. However, sediments dredging changes their environmental conditions in terms of temperature, humidity, redox potential, grain accessibility by oxidizing agents (e.g. oxygen, micro-organisms) and stability of organic matter. Therefore, the potential of contaminants release and remobilization is enhanced. In oxidizing conditions, sulfide oxidation by water and oxygen takes place to generate acidic leachates and enhances metal(oid)s mobility [8]. Then, the kinetic of sulfide oxidation is accelerated by bacterial activity (e.g. *Acidithiobacillus ferrooxidans*) especially in acidic conditions [9, 10]. It is known that sulfide (e.g. pyrite) oxidation and carbonate/silicates dissolution can be a source of various metal(oid)s such as As, Cd, Cu, Zn, Pb, Ni and Bi [11]. In general, the contaminants can present as soluble ions within porewater and/or associated to solid matrix as oxides or organic/inorganic complexes or associated to structural composition of minerals. Consequently, if the sediments were left without and management scenario the contamination will be stopped by the complete depletion of contaminants-bearing minerals or complexes. At the long-term scale, the contamination is provided by the reactivity of solid phases because soluble form can be easily washed by water. The contamination is generated by the interaction between solid matrix (contaminants-bearing minerals) and oxidized agent (e.g. water, oxygen).

Previously, ocean dumping was a major scenario for disposal of contaminated dredged sediments, but this method was abandoned since 1972 [7, 12]. Recently, solidification/stabilization (S/S) using cementitious materials was widely used as a viable remediation technique for contaminated sediments. USEPA [13] stated that, between 2002 and 2005, around 24% of contaminated materials were treated using S/S (Ex-situ and in situ) in the United States. The S/S technology aims to modify geo-mechanical, hydrogeological and geochemical properties of contaminated materials (e.g. sediments, soils) in order to reduce the release of organic and inorganic. This chapter will discuss: i) the main characteristics of sediments, ii) the main materials used for S/S of contaminated sediments, iii) the main physical and chemical mechanisms responsible of contaminants fixation during S/S as well as the main tests used for chemical effectiveness of S/S and iv) how contaminated sediments can be integrated in a circular economy (e.g. valorization).

2. Sediments genesis and their main characteristics

2.1. Sediments genesis

From a general point of view, a sediment is a deposit of mineral or organic clastic materials. Clastic particles may have an allochthonous origin (from the catchment basin) or an autochthonous origin (from immediate distance of the sedimentation place) [14].

Geologically, the breakdown of earth's rocks generates the mineral particles which, carried by wind and/or water, will accumulate on the streambed or seabed. This process, calls *weathering*, may act along two pathways: one physical, by cracking caused by expansion-contraction of the parent material, or erosion through the action of water, ice or wind, and one biogeochemical, through the action of chemical processes (hydration, hydrolysis, dissolution, and oxidation-reduction) and biological processes.

Diagenetic processes will transform sediment particles into sedimentary rocks. Four different sources may generate sediment particles (Insert Figure *1*~~Figure 1~~):

- A terrigenous source, which particles originate from the erosion of emerged surfaces, and may be carried by runoff, aerial way, or fluvial water stream;
- An endogenous source, where particles are produced by the environment where the sedimentation takes place, such as organic or inorganic debris of the autochthonous flora and fauna;
- Neoformed particles, either by alteration, transfer, or precipitation in the sedimentary basin, or inside the sediment;
- An anthropic source concerning industrial, urban or agricultural discharge, rich of organic matter (OM), suspended matter, micropollutants, metals and metalloids.

A sediment composition is complex and usually heavily depends on the catchment basin, indigenous contributions and local hydrodynamics able to mix or transport particles from a close basin. Mineral part of autochthonous sources is mainly composed of carbonates and evaporates.

2.2. Main sediments characteristics

For simplification, two categories may divide dredged sediments:

- Marine sediments, usually dredged into harbors or estuaries;
- Continental (freshwater) sediments dredged into rivers, waterways, channels, reservoirs, lakes or dams.

In this chapter, continental sediments also include urban sediments issued from stormwater management structures/facilities (retention/detention or infiltration ponds) in urban areas or alongside roads and highways. Despite a growing interest, the volumes of dredged urban sediments are intermittent and disparate and data in literature upon their use of stabilization/solidification processes (S/SP) for treatment are scarce, and the present chapter will not address them.

Most water structures require regular maintenance to ensure their proper functioning. Usually, restoring the water level is the main consideration of dredging operations, however others may apply, especially environmental considerations. The diversity of sedimentation beds induces a wide variety of sediment's characteristics, in line with both the influence of the catchment basin, and local physicochemical and environmental conditions.

Among the main physicochemical characteristics to describe sediments, particle-size distribution (PSD) is essential as it may influence S/SP and further affect the characteristics of considered reuse product. Sediments' constituents may be sorted according to their size: in descending order, block, stone, pebbles, gravel, sand, silt and clay, added to colloidal molecules able to flocculate reversibly [14]. PSD is highly dependent of generation and transport processes of sediments, mainly erosion, hydrodynamics conditions, transportation and deposition (local diagenetic processes, aggregation, etc.) [15, 16]. Continental and marine sediments show some slight differences (Insert Table *1*~~Table 1~~). Continental sediments have a higher clay content (particles <2 µm) with a median value of 11.9 against 5.71 wt.% for marine sediments, but a lower silt content (52.3 vs 63.0 wt.%). Sand content is approximately close. From a given site's perspective, local hydrodynamics also may influence PSD of accumulated sediments, as in a reservoir where the inlet is enriched in coarse particles and the area near the dam is enriched with fine particles [17, 18].

From Insert Table *1*~~Table 1~~, we observe that continental sediments tend to a higher total organic carbon (TOC) content, with a mean content of 4.21 wt.% versus 2.60 wt.% for marine sediments, and a median of 3.40 vs 2.50 wt.%. Organic matter content might be deduced from TOC, considering that OM (wt.%) = 2 × TOC (wt.%). We can deduct from TOC values that fluvial sediments have a median organic matter content of about 6.8 wt.% versus 5 wt.% for

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marine sediments. However, dispersion of the results shows a high variability of this parameter depending on many physical and geochemical processes. Organic matter, either natural or anthropic, is of great importance in sediments both from a biogeochemical perspective as a carbon source, electron donor, adsorption surfaces, chelating agent, or particles textural component; and from geotechnical perspective where OM hampers with hydration processes of hydraulic and pozzolanic binders [19-22]. OM is mainly composed of humic acids, fulvic acids, proteins, polysaccharides, and lipids [23]. Hydrogen bonds and van der Walls forces allow OM to form supramolecules, and to interact with trace metals and metalloids and mineral particles [24-27].

As sink of most dissolved and particulate compounds, sediments often exhibit anthropic or natural contamination to varying degrees with trace metals and metalloids (As, Cd, Cr, Cu, Hg, Ni, Pb, Sn, Zn, etc.) or organic substances (Polycyclic Aromatic Hydrocarbons (PAH), polychlorinated biphenyl (PCB), hydrocarbons, pesticides, etc.). For contaminants, Cd suggest being much highly concentrated in fluvial sediments (median of 0.65 vs 0.33 wt.%), conversely with marine sediments for Cu (median of 41.3 wt.% vs 21.7 wt.%) (Insert Table *1*~~Table 1~~). Most of the median results are close between both sediments and sometimes have a spread mean because of high extreme points – e.g. Hg and ∑16PAH for both sediments, and Cd, Pb, Zn and ∑7PCB for fluvial sediments.

Dissolved ionic species represent an additional distinction between freshwater and marine sediments. More concentrated in seawater than in freshwater, dissolved ionic species – and chlorides in particular – are present in marine sediments porewater, or as precipitates when the sediments dry. This difference induces a higher ionic strength for seawater, a slightly higher pH and lower dissolved oxygen, which will in return influence sedimentation processes, or contaminants solubility. Salinity is also a key parameter of numerous valorization processes, in particular in S/SP where chlorides interfere with cements components or steel bars in the case or reinforced concrete [28, 29].

More generally, other physicochemical parameters will vary drastically even in a given type of sediments, often intercorrelated to each other: pH, Eh, cation-exchange capacity (CEC), carbonates content, C/N ratio, available nutrients, major and trace metals and metalloids speciation… Water content for example may vary in a wide range (20-90%) and depends on the fineness of the texture and content in organic matter.

For mineralogical composition, freshwater and marine sediments share commonly found minerals detailed in **Erreur ! Source du renvoi introuvable.**~~Table 2~~, mainly silicates and aluminosilicates, carbonates, metallic oxides and hydroxides, phosphates, sulfides, evaporites and clays [5, 30, 31]. We will address specific issues and problematics of several classes of minerals, or particular minerals. Clays exhibit a specific affinity for trace metals and metalloids and interact with their mobility, as well as with organic matter to form clay-humic complex [32-34] . Carbonates regulate buffering power of sediments, influencing the local pH and ionic strength, as well as the mobility of trace metals and metalloids through various mechanisms (precipitation, co-precipitation, adsorption, absorption and substitution) [31, 33, 35]. Metallic (oxi)hydroxides – mainly of Fe and Mn – also interact with trace metals and metalloids through adsorption and co-precipitation (Alloway, 2012; Dijkstra et al., 2004; Lions et al., 2010), as well as phosphates minerals used to immobilize trace metals and metalloids by adsorption or precipitation [36-38]. Finally, sulfides are a well-known mineral class with close interaction to trace metals and metalloids, such as Cd, Cu, Ni, Pb, or Zn. Metallic sulfides are insoluble in water and readily precipitate when formed from water column [32, 39]. As reduced species of sulfur, metallic sulfides are highly sensitive to oxidation, susceptible to release associated trace metals and metalloids [30, 33, 40]. Sulfides oxidation and sulfates reduction highly depends on environmental geochemical conditions and bacterial activity [41]

Insert Figure 1.PNG here: Conceptual model illustrating sediments genesis

Insert Table 1 here

Insert Table 2 here

3. Stabilization/solidification technics

3.1.Definition of the technology

Solidification/stabilization of a contaminated material implies the addition of cementitious additives which improves the physical and chemical properties of the contaminated material to produce an environmentally acceptable mixture. Solidification and stabilization have different objectives: solidification which refers to the hardening of matrix which makes it easy to handle and ii) stabilization which refers to the immobilization of one or several chemical species of potential concern (e.g. organic contaminants, heavy metals). Within S/S technologies, the contaminants are not removed but transformer from leachable form to stable and low-solubility form. This technology can be used in-situ and ex-situ. Moreover, S/S technology is considered is time and cost-efficient [42]. Solidification of contaminated sediments improves their workability and may create an opportunity for their valorization [43]. However, there is no 'magical formulation' that can be used successfully for all types of contaminants; S/S technology is site-specific [44]. The choice of cementing materials must consider the characteristics of contaminated sediments and the compatibility of cement and sediments [45].

3.2.Most used materials for S/S technology

Ordinary Portland cement is the most used material for S/S of contaminated sediments due to its demonstrated effectiveness [46, 47]. Cement-based stabilization/solidification was used successfully to immobilize heavy metals and ~~metaloids~~metalloids (Insert Table *3*~~Table 1~~). Recently, several researches were conducted to ~~substitute ,at~~substitute, at least, partially cement in S/S technology [47, 48]. The recent trends aim to reduce cost related to cement and valorize by-products of other industries, which participate in the principal of circular economy. Moreover, in some cases cement substitution by industrial by-products (e.g. cement kiln dust, fly ashes) improves the mechanical properties of solidified materials and increases contaminants fixation. Insert Table *3*~~Table 1~~ presents a summary the most used materials for S/S of contaminated materials. The main conclusion is that the choice of the materials and their dosages is case-specific. In general, cement is substituted by materials providing additional acid neutralization capacity or pozzolanic materials.

Park (200) [49] substituted 20 wt.% of ordinary Portland cement by CKD to stabilize hazardous wastes containing heavy metals. CKD is a by-product of Portland cement manufacture and characterized by an uniform particle size distribution. CKD contains high CaO and SiO_2 contents (around 40 and 10 wt.%, respectively). The use of CKD provided additional alkalinity which accelerated cement setting and hydration. Additionally, the formulation containing CKD showed the highest compressive strength and consequently the lowest metals release. CKD can be combined with other industrial by-products such as lime dust. Eisa et al (2020) [50] showed that the use of lime dust improves arsenic stabilization within contaminated soils. However, if the levels of ~~alkalies~~alkalis within CKD are low, contaminants fixation and compressive strength of stabilized material are negatively affected [51]. Metal leaching can be significantly reduced when CKD is combined with red mud bauxite (RMB) which is characterized by a high affinity to metals. Indeed, Doye and Duschene (2003) [52] stabilized high reactivity materials (sulfides) using formulations based on CKD and RMB (10 wt.%).

Recently, the use of low carbon cements for the stabilization/solidification of sediments is progressively needed due to their advantageous economic and environmental spinoffs. Unlike

conventional cements, low carbon cements allow to reduce the GHGs emissions and production costs either via the decrease of clinkerization temperature or via the substitution of clinker by supplementary cementitious materials (SCMs). Sulfoaluminate and reactive MgO cements have been used as green alternatives to stabilize hazardous wastes due to their capacity to immobilize toxic contaminants in the hydration products; ettringite in Sulfoaluminate cements [53] and hydrated brucite (Mg(OH)2) in reactive MgO cements [54]. Otherwise, many alternative SCMs were tested worldwide going from waste streams (i.e. silica fume, fly ashes, blast furnace slag, etc.) to thermally activated clays (i.e. metakaolin, calcined clays, etc.). Recently, researchers have started to evaluate the performance of limestone calcined clay cement (LC3) to immobilize pollutants [55]. This kind of binders is mainly composed of calcined clays at low temperatures mixed with limestone and clinker. It allows the development of materials with high durability, low water absorption and high chloride resistance [56]. Also, alkali activated cements are promising binders for the S/S of sediments. They allow to obtain stabilized matrices with low permeability, better acid and sulfate resistance, better heavy metals stabilization [57].

Insert Table 3

3.3. *Discussion of the main chemical and physical mechanisms responsible for contaminant stabilization during S/S technology*

Cementitious additives alter the physical and hydrogeochemical properties of contaminated materials. The main objective of S/S technology is to produce a stabilized material environmentally acceptable by decreasing its reactivity and chemical species release. In general, fixation and immobilization of contaminants is ensured by mechanisms related to i) cement hydration, ii) precipitation of secondary minerals at neutral to alkaline environment and iii) hardening of contaminated materials. Two main categories of mechanisms are responsible of contaminants immobilization: i) chemical and physical mechanisms [45]. However, quantifying the share of each of these two categories is difficult because of their interaction. The major mechanisms responsible for contaminants stabilization are summarized in Insert Figure 2~~Figure 2~~.

3.3.1. Physical mechanisms

Basically, solidification of contaminated sediments alters their mechanical properties. Cement hydration products formation is translated by the hardening of the matrix such as the increase of its mechanical resistance. Solidification of contaminated sediments by cement hydration alters its microstructure. Indeed, the porosity of the stabilized contaminated sediments is considerably reduced [46]. Basically, reduction of contaminants release by stabilized sediments is ensured by reduction of liberation degree (i.e exposure rate) of reactive minerals. It is demonstrated that reactivity of minerals is correlated to their liberation degree [58-60]. Indeed, during sediments hardening by cement hydration, several mechanisms are responsible of the decrease of the available surface area of reactive minerals such as:

i) Precipitation of cement products at the surface of grains. Indeed, reaction of hydration of cement compounds such as alite and belite leads to the formation of CSH which is responsible of cementing action of Portland cement [61]. In general, cement hydration products are precipitated at the porosity of sediments and at the surface of grains. Additionally, cement hydration removes the free sediments pore water which is generally highly charged in soluble ions.

ii) Reduction of contact surface and time contact between oxidizing agent and reactive grains. Indeed, sediments cementation improves their geo-mechanical performances (e.g. UCS). Consequently, the porosity and saturated hydraulic conductivity are

decreased, which affect the hydrogeological behavior stabilized sediments. There are few papers that studied the effect of cement on the water balance within stabilized materials. Elghali et al (2020) [47] studied the effect of cementitious additives on water content and infiltration within contaminated tailings using volume water probes. The authors demonstrated that contaminated materials cementation decreases water vertical infiltration rate and favors surface and sub-surface runoff, which is explained by reduction of hydraulic conductivity. Spence and Shi [45] stated that hydraulic conductivity of mature Portland cement can reach 10^{-13} m/s. When water vertical infiltration is limited, both contact time and surface contact between reactive grains and oxidizing agents are limited, which reduces the overall reactivity of contaminated sediments. It is known that the chemical species release from reactive grains depends on their accessibility by oxidizing agent (e.g. water, oxygen) [62].

3.3.2. Chemical mechanisms

Cementitious additives allow fixation of contaminants by several chemical mechanisms including: i) precipitation of metal(loid)s as oxy-hydroxides, ii) adsorption of contaminants on the surface of cement hydration products (e.g. CSH) and iii) fixation of contaminants by secondary oxy-hydroxides.

i) Cements are characterized by their high acid neutralization capacity. This means that pH of sediments pore water is increased to reach circumneutral and alkaline values. In these environments, the metal(loid)s are characterized by low solubility, which means that soluble ions will precipitate as hydroxides [63]. Benzaazoua et al (2002) [64] demonstrated that arsenic mobility is controlled by calcium content within binding agent. Indeed, in lime rich mixtures, arsenic is immobilized by portlandite dissolution. The arsenic is precipitated as $CaAs(OH)_4$. Other authors demonstrated the same conclusions regarding other heavy metals such as Cd [43]. Moreover, depending on iron activity in the pore water secondary iron oxy-hydroxides precipitate in circumneutral/alkaline conditions (Figure 2). These secondary phases are known for their affinity to several chemical species (e.g. Cd, As). The precipitation of iron-oxy-hydroxides is often associated with fixation of contaminants [65-67]. The mechanisms by which the contaminants are fixed by secondary oxyhydroxides are commonly called 'sorption' which includes adsorption on the surface, complexes formation, physical adsorption and eventually substitution [68]. Fixation of contaminants by secondary phases may be reversible depending on the fixation mechanism and the crystallinity of the phase. Indeed, the release of a substituted contaminated from well crystallized is negligible compared to a contaminant adsorbed in an amorphous phase [69]. The durability of chemical fixation of contaminants by secondary phases can be altered if the geochemical conditions change (~~e.g~~e.g. pH, redox potential).

ii) During cement hydration results in the formation of several products including calcium hydroxide, calcium sufoaluminate and calcium silicate hydrate (CSH) [64]. Formation of CSH gel-like, which is characterized by high specific surface area, is responsible of the adsorption of several contaminants. Contessi et al (2020) [70] highlighted the formation layers of CHS containing high concentrations of Pb during the stabilization of a Pb-contaminated materials stabilization. Fixation of contaminants by CSH phase is pH dependent. If the pH of stabilized material is suitable for CSH phase stability, the contaminants release will be negligible. Similarly, at acidic conditions fixation of contaminants by CSH is reversible. Moreover, Yakubi et al (2018) [71] stated that stabilization of metals (Cu, Pb, Zn, Cr, Cd, As and Mn) is possible within cementitious materials at a pH values ranging between 5 and 11 except Zn

which is was found at its soluble form at all pH ranges. Additionally, As the chemical composition of CSH is variable depending on cement composition, its capacity to immobilize metals is variable. Indeed, retention of metals by CSH depends on its Ca/Si ratio

3.3.3. Biological mechanisms

Biological processes are not directly involved in S/S classical technology. However, unlike organic contaminants, which are bio-chemically degradable, heavy metals are not, but they can change speciation, solubility and toxicity depending on environmental conditions (e.g. pH, redox potential, presence of organic material, microbial activity). As a result, in metal contaminated solids, remediation strategies can either enhance metal mobilization (increased solubility via biohydrometallurgy for example) or immobilization (decreased bioavailability). Biological-based processes (e.g. biosorption, bioaccumulation) are therefore promising for the treatment of contaminated sediments [72]. The S/S popularity is based on the rapid, versatile (in situ or ex situ), broad range of contaminants immobilized and low-cost achievement of treatment objectives. However, stabilization of contaminants without matrix solidification could also sometimes prove sustainable, especially when coupled to a controlled release of reactants/microbes for enhanced remediation in the long term [73] or to green/alternative binders for diverting hazardous wastes from disposal following an life-cycle assessment [74]. In this sense, the literature shows a slow but steady evolution towards environmentally sustainable solutions: from high impact (thermal treatment and cementification) to low impact (chemical and biological) processes [72]. Moreover, in some countries (Denmark, South Korea) the S/S technology is prohibited because removal of contaminants from soils is required over their stabilization [73].

Insert Figure 2.JPG here: Conceptual model explaining the main mechanisms responsible for contaminants immobilization during S/S technology

4. Main tests used for assessment of the effectiveness of S/S technology

The main objective of S/S technology is to reduce the risk of organic and inorganic contamination of environment by the reactivity of the contaminated material. Despite hydro-geotechnical properties of stabilized matrix, the reactivity of stabilized matrix is crucial to evaluate the effectiveness of the used S/S technology. After stabilization/solidification of contaminated sediments, interaction with oxidizing agents (e.g. water, oxygen, microorganism) took place [75]. The chemical quality analysis of the leachates can indicate the effectiveness of S/S technology to immobilize contaminants. During last decades, several procedures were developed to evaluate chemical species release by solid materials (e.g. stabilized matrix). The most used tests for the leaching of stabilized materials are tank leaching tests [76], toxicity characteristics leaching procedure (TCLP), 1313, 1315 and 1316 methods, column tests and field cells. All these tests consist of submitting a monolith or crushed sample to weathering by deionized water or an acid with different L/S ratio and time duration. Within each test, the leachates are collected and analyzed for their physio-chemical characteristics (e.g. pH, electrical conductivity, chemical composition). The leachates analysis allows quantifying chemical species release from the stabilized matrix. This way, the chemical effectiveness of S/S technology to immobilize contaminants is studied.

Thank leaching tests consist of submerging a monolith sample with a known external surface (cylinder, parallelepiped) for 64 days. the ration L/S (cm^3/cm^2) is about 10/1. The deionized water is renewed after 0.25, 1, 2.25, 4, 9, 16, 36 and 64 days. This test is primarily designed to

determine the leaching of inorganic components from monolithic material. It is used to quantify diffusion and effective diffusion coefficient. This test cannot be used for soluble materials [76]. Toxicity characteristics leaching procedure (TCLP) was designed to determine leaching of organic and inorganic components from industrial wastes. During TCLP tests, around 20 g of crushed sample is allowed to react for 18h with acetic acid. The method 1313 allows representation of liquid solid partitioning in function of pH for organic, inorganic and semi-volatile constituents in a solid material (e.g. stabilized sediments). This method is realized in a liquid/solid ration of 10 ml/1 g of dry sample [77]. It consists of nine parallel extractions of crushed sample in dilute acid or base and water (more details can be found in [77]).additionally, the method 1315 can performed on monolithic sample, like TLT, to evaluate release rates of chemical species in saturated conditions. The sample is allowed to react with reagent water in a liquid/surface ratio of about 9±1 mL/cm² at nine predetermined intervals (for more details about experiments, see leaching environmental assessment framework). The method 1316, is relatively like 1313 method, is designed to provide intrinsic parameters controlling the leaching of inorganic chemical species under equilibrium conditions. It provides the liquid/soldi partitioning at neutral pH [78]. Operationally, the sample is allowed to react with reagent water for a solid/liquid ration varying between 0.5 and 10 depending on the sample's particle size [78]. The tests described above are all performed on small sample mass for relatively a short time duration which makes prediction of the long-term leachability of tested material difficult. Nowadays, column tests which are used in the field of environmental geochemistry are used successfully for the leachability of stabilized materials. Column tests consist of submitting stabilized materials to natural weathering by deionized water [79, 80]. There are no limitations related to disposal configuration, sample mass, liquid/solid ration, and time duration. Several configurations can be performed depending on the objective of the study. Column tests can be run in saturated or unsaturated conditions.

In general, laboratory tests can provide several information related to stabilized material reactivity and chemical species release. So, they can be considered as complementary tests except those that have the same operational procedure (e.g. TLT and 1315 method). The leachates chemical composition is compared to environmental regulation which allow to evaluate the effectiveness of S/S technology [46]. However, laboratory tests can not represent field conditions. There is no unique mathematical model than can extrapolate laboratory results to field conditions [81]. This can be explained by several factors: i) in field conditions hydrogeological factors may favor surface runoff and consequently contact time between stabilized matrix and reagent is reduced, ii) leachable surface is greater within field conditions than that with laboratory conditions, iii) sample dimensions are generally larger for field conditions, vi) liquid/solid ratio, v) flushing frequency which is dictated by climatic conditions within field conditions and predefined for laboratory conditions, and vi) one of the most influencing factor is that the presence of heavy metals results in retard of cementitious materials' hydration to set and harden [49]. Consequently, the best practice to evaluate contaminants immobilization within a S/S technology is to implement field cells despite their elevated cost [47, 82].

5. Integration of contaminated sediments in circular economy

Worldwide dredging operations are behind the production of high volumes of sediments frequently contaminated. The prevention of this kind of wastes is most difficult as it is the result of many natural processes added to anthropogenic sources. The most suitable solution in the

hierarchy of the integrated waste management diagram is to reuse/recycle these sediments as alternative materials. This solution could help to avoid the consumption of new finite primary resources (gravel, sand, clays, etc.) and to reduce the degradation of natural ecosystems. Various applications could use contaminated sediments due to their significant volume, and their chemical, mineralogical and geotechnical characteristics. Mineralogically, dredged sediments are mainly composed of silicates, carbonates, clays, sulfides, (oxi)hydroxides and organic matter. From a particle size point of view, classification of sediments can fall into different categories: gravel, sand, silt and clays.

Making use of wastes is always welcome when it is economically viable, technically feasible, environmentally sustainable and socially acceptable. The direct reuse of sediments in their initial state is often difficult. Therefore, many parameters should be controlled and mastered: the initial water content and salinity, organic matter, heavy metals content and speciation, etc. Water content is particularly a common impediment to valorization, usually handled by water removal readily after dredging. In case of noteworthy contamination level, a pretreatment should apply to the sediment before suitable use for removal or immobilization of the contaminants. In most cases, the nature of the sediment and the final application will prescribe the useful pretreatment(s) [43]:

- ✓ Physical methods (flotation, hydro-cycling, vacuum extraction, etc.),
- ✓ Physicochemical methods (phosphating, acidification, dichlorination, leaching, etc.),
- ✓ Chemical treatments (solidification-stabilization using a hydraulic binder such as cement, lime, pozzolans, clay, or other binders, such as polymer resins),
- ✓ Biological treatments (bioremediation, phytoremediation, etc.),
- ✓ Heat treatment (thermal desorption, incineration, vitrification, pyrolysis).

Civil engineering and public infrastructures sectors efficiently use alternative materials as stabilized sediments. For instance, massive infrastructures worldwide used a substantial amount of sediments such as the Palm island in Dubai, the Hong Kong airport, or the National theatre of London [83]. They were also used in large-scale road projects in Dunkirk Harbor in France while respecting economic and environmental constraints [84-86]. However, these sediments have been stabilized before their use by hydraulic binders (cement, lime, fly ashes and slag) and/or mixed with other mineral granular correctors (sand, slags, etc.) to satisfy the mechanical performances and environmental requirements.

More generally, civil engineering frequently valorizes dredged sediments because construction field is an extensive consumer of natural resources to produce aggregates (with sand) or hydraulic binder as Portland cement (with clay and limestone). Partial or total substitution of natural sand as aggregate in mortars, concretes or in unfired bricks is a common valorization of dredged sediments [87-91], with actual production of effective harbors' accropodes [84], pedestrian and vehicles pavers [92], synthetic cobblestones [93] or eco-friendly foamed concrete [94]. In some cases, a synthetic polymer replaces Portland cement to produce polymer mortars with epoxy resin [95, 96], or floor-covering concrete tiles with polyester resin [97]. Silt content and soluble organic matter, two intrinsically linked parameters, may impair mechanical and durability properties of produced mortars and concretes (hydration, setting time, compressive strength, shrinkage strain). Dredged sediments are thermally or chemically

pretreated [98], or sieved for coarser fractions [20] to overcome these issues. A high-temperature thermal treatment (>1100°C) produces synthetic lightweight aggregates to make structural lightweight concretes [99-102]. Overall, sand substitution is usually between 10 and 20 wt.% and can rise to 100 wt.% (up to 95 wt.% of the total of the mix) for structures with low loading.

Recently, a second approach emerged with the valorization of sediments without thermal treatment through substitution of cement for production of mortars and concretes (until 40 wt.%) [91]. However, most studies focus on thermally pretreated sediment able to remove organic matter and enhance pozzolanic activity [103], to produce blended or composite sediment-based cement (usually 8 to 15 wt.% in the raw mix, until 40 wt.%) [104]. Efficient and durable results are obtained with a replacement rate of 10 wt.%. Dredged sediments may also enter the composition of self-consolidating concrete [105]. For more information on the use of dredged sediments as supplementary cementitious materials (SCM), the curious reader can refer to a recently published review (Amar et al., 2020). Finally, Portland cement production may use dredged sediments to replace a part of raw materials in the clinker mix, predominantly with freshwater sediments (dam, fluvial) for their low chloride contents, but not only [106]. Nonetheless, heterogeneity of sediments deposits might be an obstacle for the upscaling to industrial level, requiring proper characterization before each considered application.

Sediments were also good alternative materials to substitute the use of clays and sand in building bricks and tiles manufacturing. Many applications in the laboratory scale were investigated in the literature [107, 108]. Full-scale production examples were reported in France [109], Germany [110] and China [111]. For all these applications, sediments undergo a common Novosol® pretreatment aiming to immobilize heavy metals and metalloids and remove the organic matter by a chemical treatment (phosphatation) followed by calcination (> 650°C) [112]. Even though the use of the sediments in brick making presents several advantages from a technical, economic and environmental point of views, many challenges are faced when coming to consumption at a large scale and introduction to the market. Sensitization and communication with final customers were recommended [113]. Another valorization way exists, such as coastal nourishment [114], as a growing medium for lettuce production [115], or production of ceramic glaze [116].

6. Durability of stabilized sediments

As discussed above, S/S technology allows stabilization of contaminated sediments by physical and chemical processes. The main objective of a S/S technology is to produce a safe stabilized matrix at the long-term scale. However, exposure of stabilized sediments to climatic conditions may alter their physical properties and consequently the immobilization of contaminants. Sun et al (2020) [117] studied the effect of rainfall on the effectiveness of S/S of heavy metals-polluted sediments. The authors concluded that deterioration of mechanical resistance of stabilized sediments depends on curing time. The deterioration of mechanical resistance is negatively correlated to curing time; higher is the curing lowest is the deterioration of mechanical resistance. After 28-days of curing, the deterioration of mechanical resistance was about 8%, while after 7-days of curing time the deterioration was around 34%. Consequently, at filed conditions the effect of rainfall can be reduced if the stabilized matrix had enough time to set. ~~The~~ Moreover, the deterioration of mechanical resistance didn't affect the stabilization of heavy metals. However, at the long term-scale the stabilization of heavy metals can be affected by increasing the available surface area of reactive grains and contact time between oxidizing agents and reactive grains (Insert Figure 3~~Figure 3~~). Cracks formation and development may also increase the contact time and surface contact between oxidizing agent and reactive grains may result in remobilization of contaminants. Cracks can be formed due to

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freezing-unfreezing cycles and precipitation of expansive minerals because of sulfate attack [118]. However, the stabilized matrix can be protected by adding protection layer that can limit water vertical infiltration and limit evaporation [119].

Insert Figure 3.JPG here: Effect of climatic conditions on the effectiveness of S/S technology

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