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Dredged marine sediments stabilized/solidified with cement and GGBS: Factors affecting mechanical behaviour and leachability

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Abstract: Management strategies for the safe disposal of contaminated dredged marine sediment constitute a global-scale environmental issue. The stabilization/solidification method was investigated as a sustainable approach to the recycling of the sediment as a construction material. A systematic study of the factors affecting the mechanical performance and contaminant release was performed. The physico-chemical variables selected to assess the potential re-use of the sediment treated with Ordinary Portland cement (OPC) and Ground Granulated Blast Furnace Slag (GGBS) in an aquatic environment were: curing duration (7, 28, 56 and 98 days), curing temperature (5, 20 and 40 °C) and ambient (leachate) pH (1, 4, 7 and 10). Unconfined compressive strength (UCS) tests were conducted and extended-duration tank leaching tests were used to characterize the long-term leaching of Al, Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Ba, Pb. The results showed that S/S methods provide excellent immobilization of metals in marine sediment at a pH range of 4 to 10. Immobilization efficiencies > 99.9% for Mn, Fe, Zn, As, Ba, Pb and > 97.8% for Al, Cu and Zn were reported over 100 days. GGBS replacement is an effective way to further improve sediment properties by enhancing strength, mitigating sediment alkalization and offering a better immobilisation capacity for Fe, Ni and Zn. The release of metals (Al, Mn, Cu, As, Ba and Pb) was strongly associated with a coupling effect of the physico-chemical factors, with metal-specific responses to curing temperature, curing duration and pH. Mn mobility showed a dramatic sensitivity to ambient pH while Ba was less pH-dependent. Al release is related to strength and leached out by dissolution in all situations considered. Considering that dredged marine sediments may contain multiple metal contaminants which exhibit individual responses to remediation, treatment with GGBS may be considered a potentially suitable management option.

Keywords: Marine dredged sediment; Unconfined Compressive Strength; Tank leaching test; Leaching ratio.
1 Introduction

Dredging is carried out periodically in channels and ports worldwide in order to maintain sufficient depth for navigation, resulting in the transfer of several hundred million cubic metres of sediments from the aquatic to the terrestrial environment annually (Lirer et al., 2017; Snellings et al., 2016). These sediments typically have a high moisture content and poor geotechnical properties (low strength and stiffness); some have accumulated high concentrations of contaminants (such as metals, organotin compounds, polycyclic aromatic hydrocarbons (PAH) and polychlorinated biphenyls (PCBs)) as a result of natural and anthropogenic activities (Akcil et al., 2015). Europe alone produces about 300 million cubic metres of such contaminated sediment from dredging each year (Snellings et al., 2018). Traditional management strategies for these sediments include landfilling and confined or unconfined disposal at sea. However, these strategies have many disadvantages including cost, capacity limitations and potential for environmental contamination (Agius and Porebski, 2008; Akcil et al., 2015). As a result, there is an emerging need for cost-effective methods of treating this dredged material.

Stabilisation/Solidification (S/S) using cementitious materials is widely established as an effective method of improving the engineering properties of sediments and encapsulating contaminants (Barjoveanu et al., 2018; Lei et al., 2018; Palansooriya et al., 2020). In particular, the ability of OPC to improve geotechnical characteristics (Wang et al., 2013; Zentar et al., 2012) and immobilize contaminants (Jiang-Shan et al., 2017; Wang et al., 2019b) has been documented widely. However, OPC is responsible for 5–8% of global anthropogenic CO₂ emissions and accounts for 12–15% of total industry energy use (Ali et al., 2011; Scrivener and Kirkpatrick, 2008). This has prompted increased research to investigate more environmentally friendly and sustainable materials for S/S applications.

Pozzolanic materials such as Ground Granulated Blast Furnace Slag (GGBS), a by-product of the iron/steel industry, have been widely recognized as potentially suitable partial substitutes for cement (Cheah et al., 2016; Ge et al., 2018; Özbay et al., 2016). GGBS can consume cement hydrates (portlandite)
generating secondary hydrates through a pozzolanic reaction (GGBS hydration) (Goodarzi and Movahedrad, 2017; Özbay et al., 2016; Wang et al., 2019a; Xie et al., 2019) aiding the densification of soil (Nedunuri et al., 2020; Wang et al., 2018a), which in return accelerates cement hydration reactions (Wan et al., 2004). Additionally, the greater specific surface area of the GGBS particles leads to the formation of more hydration product nuclei in the cement-GGBS matrix (De Belie et al., 2010; Sanjuán et al., 2018). An appropriate ratio of OPC to GGBS not only optimises the engineering properties of treated soil (Cokca et al., 2008; Ge et al., 2018; Preetham and Nayak, 2019; Sheen et al., 2013; Wang et al., 2018b), it also enables further potential immobilization of contaminants (Wang et al., 2019a, 2018a; Yi et al., 2016).

Stabilized/Solidified sediment is recycled as a construction material where possible in preference to depleting other natural resources. In the natural environment, ambient temperatures have a significant influence on S/S mixtures, especially for cement-based materials (Pedrosa et al., 2020; Wang et al., 2017). Exposure to elevated ambient temperatures likely causes the ‘crossover’ effect in cement-based GGBS mixtures, whereby the early stage formation of hydrates occurs too quickly, leading to the non-uniform dispersion of the hydration products. This results in the formation of a hard paste in the pores and the hydrated shells and consequently, a low permeability at the surface of the particle (Aziz et al., 2012; Castellano et al., 2016; Tanyildizi, 2009). This restricts further hydration and reduces the final strength (Castellano et al., 2016). Elevated temperatures (20°C, 40°C and 60°C) were found to lead to a higher strength at early stage formation of a mortar incorporating 40% GGBS (Castellano et al., 2016), while crossover effects occurred after 28 days curing and an optimal temperature of 40 °C. Other studies recommend temperatures of 20°C (Ezziane et al., 2007) or 30 °C (Escalante-García and Sharp, 2001) as optimal for strength in slag/cement materials after 28 days of curing. Due to the complexity of the concurrence of cement hydration and GGBS hydration in cement/GGBS combinations, there is no consistent crossover effect for cement/GGBS mixes, especially when combined with natural soils. Additionally, when S/S sediments are re-used in an aqueous environment (such as for tunnels in river/lake/sea environments and underwater in-situ stabilization projects), the pH of the host water is
critical, with neutral and weak alkaline conditions (i.e. pH 7-10) deemed most suitable for the immobilization of contaminants (Du et al., 2014; Wang et al., 2016).

Due to the complexity of marine sediment properties and physico-chemical reactions between binders and metal contaminants, different binders tend to show different efficiencies for metal immobilization. For example, Ni and Zn are more mobile in marine sediments treated with lime and cement (Saussaye et al., 2016) and the immobilization efficiency of Cd is lowest when the sediment is mixed with a chelator and humic acid (Xu and Ying, 2017). Taneez et al. (2016) reported that when marine sediment was neutralized with bauxaline (for stabilization), the immobilization efficiency of Cu was lowest among the metals considered (Cd, Zn, As, Mo, and Cr).

Overall, most studies to date have focused on individual parameters and there remains a paucity of systematic research considering the combination of factors affecting the properties of S/S sediment (especially when recycled in an aquatic environment) in a unified way. Additionally, the immobilization efficiency of marine sediment stabilized/solidified with cement and GGBS requires further investigation, given the increased number of commercial projects considering this option. To this end, a comprehensive suite of mechanical and leaching tests was performed on a contaminated dredged sediment stabilized with cement and cement/GGBS binders, and subjected to ranges of curing time, curing temperature and ambient leachate pH. The specific aims included:

1) a comparative evaluation of the effect of curing duration, curing temperature, leachate pH and water immersion on the unconfined compressive strength of cement-only and cement/GGBS mixtures;

2) an assessment of the effect of curing duration, curing temperature and pH on the leachability of selected metals, as investigated through tank leaching tests, in addition to a consideration of the prevailing leaching mechanisms for various contaminants.
2 Materials and sample preparation

Approximately 50 kg of recently-dredged sediment was collected from a commercial harbour in Ireland (December 2016). Once in the laboratory, debris was removed, and the sediment was carefully homogenized to maintain uniformity and stored at -20 °C until time of use to prevent potential biogeochemical reactions. The sediment comprised approximately 83% sand, 16% silt and 1% clay (n=3), classifying as a silty sand. The particle size distributions are shown in Figure 1, with $D_{10}$ (the particle size for which 10% of the material is smaller) and $D_{50}$ (mean particle size) values of 0.015 mm and 0.17 mm respectively. The water content of the sediment ranged from 52.3-54.4% (n=5).

The OPC (Lagan CEM I 42.5 R) and GGBS used in this study were supplied by Ecocem Ireland Limited (Dublin, Ireland). The particle size distributions for OPC and GGBS are also presented in Figure 1. The sediment particle size is typically an order of magnitude larger than that of GGBS and OPC; with the GGBS particle size marginally smaller than that of the OPC. Blaine fineness values for OPC and GGBS were distinctly different at 3520 cm$^2$/g and 4490 cm$^2$/g respectively. The chemical constituents of both binders are listed in Table 1.

Two types of binder (12% OPC and 6% OPC + 6% GGBS; subsequently referred to as 12C and 6C6G respectively) were added as a proportion of the dry weight of the sediment as described in Zhang et al. (2018). Perspex moulds (100 mm height × 50 mm diameter) were used to form the UCS and tank leaching test samples after thorough mixing. The samples were removed from the moulds after 1 day, and subsequently cured in a sealed plastic bag. The samples cured at 5 °C and 20 °C were located in separate curing rooms with similar relative humidities of 60%; another batch of samples was immersed in a water tank (constant volume) with a constant temperature of 40 °C.

The microstructure of the fresh sediment, as characterised by Scanning Electron Microscopy (SEM) (Figure 2), consists of an abundance of platy structures. The element concentrations in the fresh sediment
are presented in Table 2, along with Soil Guideline Values (SGVs) from the UK (Environment Agency, 2002), Dutch target and intervention values for soil remediation (Pronk, 2000) and Irish upper level guidelines for disposal at sea (Cronin et al., 2006). Arsenic is the only element exceeding the limits in the Residential/Allotments category for the SGVs, whereas the values of Zn, Ba and Cd exceed their Dutch target values (Table 2). None of the Irish values are exceeded.

3 Test programme

The test programme comprised the following sequence of experiments:

(i) A suite of standard UCS tests was conducted on stabilized sediments using a UTS012 Fully Automated Cyclic Triaxial (FACT) Testing Machine. Supplementary UCS tests were carried out on specimens that had been immersed in liquid (deionized water) for tank leaching tests in advance. The loading rate used was 1.0 mm/min and all tests were performed in duplicate.

(ii) Metal concentrations (Al, Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Ba, Pb) in the parent sediment were determined using inductively coupled plasma mass spectrometry (ICP-MS) (PerkinElmer, Elan DRCe, USA) (McEneff et al., 2017; Wan et al., 2017). Prior to metal concentration determination, sediment samples (250 mg oven dried at 105 °C for 24 h and sieved to eliminate particles > 2 mm in size) were mineralized and dissolved using aqua-regia (HCl:HNO3 in the ratio 3:1) [SpA grade, Romil™, Cambridge, UK] in a closed vessel microwave digestion system (MILESTONE, 1200-Mega, Bergamo, Italy). The digestates were filtered (Whatman No. 541) and diluted to 50 ml in trace metal-free centrifuge tubes [Labcon, Petaluma, CA, USA]. The method was validated by using Certified Reference Materials (CRMs) (NIST SRM 2711a and Sewage Sludge 2: CRM029-50G), method blanks and duplicate samples.

(iii) The leachability of the treated sediment was assessed by Dutch tank leaching tests (EA NEN-7375, 2004) (Figure 3); these were performed using a liquid/solid ratio of 3.8 (middle of the permissible
range of 2-5) and an extended total leaching time of 100 days (compared to standard 64 day duration) was observed. In this test, leachate with different pH levels (1, 4, 7 and 10) was prepared using deionized water, hydrochloric acid (HCl) and sodium hydroxide (NaOH). The specimen was placed on a pedestal (inner diameter of 40mm and outer diameter of 50mm) with holes to allow contact between the liquid and the specimen. The liquids were added carefully to the tank without disturbing the sample. After each leaching interval specified in EA NEN-7375 (2004), the liquid was withdrawn and replaced immediately with fresh liquid. The pH of the leachate was measured for each leaching interval, while the sample mass and UCS were determined at the end of the test.

Microstructural analyses of the treated sediment were conducted on gold coated samples (Emitech K550, Quorum Technologies Ltd, West Sussex, United Kingdom) using scanning electron microscopy (SEM) in secondary electron mode using a Hitachi model S-4700 (Hitachinaka, Japan). The analyses were performed using an acceleration voltage of 20 kV, an emission current (Ic) of 10 mA and a working distance 12mm (Morrison et al., 2009; Jonker et al., 2015).

4 Results and Discussion

4.1 Unconfined Compressive Strength (UCS)

4.1.1 Effect of curing duration

In order to investigate the effect of curing duration on the strength of the OPC-only sediment and OPC/GGBS sediments, the UCS values for the 12C and 6C6G cases are presented in Figure 4 (with standard deviations shown) as a function of curing time (up to 98 days). The UCS of both mixtures increased over time as expected. The 6C6G mixes illustrated a reduced rate of strength gain from 28-56 days, sandwiched by higher rates in the 7-28 day and 56-98 day brackets (the latter high rate was the
justification for the extended period of 100 days for subsequent leaching tests). These trends can be explained by the reactions occurring in the mixtures, such as the OPC hydration reaction products for short-term strength gain and the pozzolanic reaction products for long-term strength gain.

The UCS values for the 6C6G mixtures were much greater than those for the 12C mixtures for all stages of curing, including early stages (approximately twice as high after 7 days). This reflects an acceleration of cement hydration by the mutual activation of OPC and GGBS, allowing earlier development of pozzolanic reactions. The improvement in the strength for the 12C mixtures became less pronounced beyond 56 days (4.5 times lower than the 6C6G mixtures after 98 days), indicating that the partial replacement of OPC by GGBS was highly beneficial for the improvement of sediment strength.

The effect of curing duration is also evident from the scanning electron micrographs in Figure 5. Platy structures from the parent sediment and a limited amount of needle-shaped ettringite were observed for the 12C samples at day 7 (Figure 5a-1). The slightly denser structure in Figure 5b-1 results from partial replacement with GGBS. Obvious changes occurred in both samples as curing time increased, with an abundance of ettringite evident in cementitious materials at 28 days (Figure 5a-2 and Figure 5b-2), resulting in a denser, less-porous structure. This increased the interlocking between aggregates, reflected in the increased UCS values. Evidence of further densification is apparent after 56 days for both mixtures (Figure 5a-3 and Figure 5b-3).

4.1.2 Effect of curing temperature

The effect of temperature (5°C to 40°C) on the 28-day UCS for both 12C and 6C6G mixtures is presented in Figure 6. The hollow datapoints refer to samples on which UCS tests were conducted after 100 days of immersion in the tank leaching apparatus. The corresponding SEM images are presented in Figure 7. In general, the UCS for the 12C mixture increased moderately with increasing temperature. This moderate change is difficult to observe from the results of the SEM analysis (Figure 7), as the morphologies of the microstructures at 5°C, 20°C and 40°C are similar; the hydrates cover the sediment
particles, filling the pores and a large quantity of needle-shaped ettringites are evident which strengthens bonding between inter-aggregates.

The greatest UCS values for the 6C6G mixture after 28 days occurred at a temperature of 20°C (Figure 6). This is also apparent from Figure 7b; there are more pores, fewer cementitious materials and weaker inter-aggregate connectivity, with more irregular ettringite crystals occurring at temperatures of 5°C and 40°C than 20°C. The lowest UCS values were observed at 5°C as the lower temperature had an adverse effect on cement-hydration and GGBS-hydration processes (Demirboğa et al., 2014). The explanation for the lower UCS values at 40°C is the aforementioned crossover effect. At 40°C, the rapid accumulation of hydrates during early-stage hydration of the cement and GGBS made dispersion in the sediment matrix more difficult, which restricted further chemical reactions, resulting in the lower UCS values at 40°C than at 20°C. This crossover effect does not arise in 12C samples probably due to the significantly lower amounts of hydrates produced in lower UCS mixes. This reduced sensitivity to temperature for GGBS mixtures is inconsistent with previous observations (Cheah et al., 2016; Ezziiane et al., 2007; Ogirigbo and Black, 2017; Wang et al., 2017).

Samples of 12C and 6C6G with prior immersion/leaching all exhibited the same UCS dependence on temperature as those without prior immersion/leaching, but with higher values. It is postulated that the hydrates, that normally cover cement particles and restrict activity, were dissolved in water after 100 days of water immersion (Baquerizo et al., 2014) and therefore accelerated further hydration. For 6C6G mixtures with prior immersion/leaching, a temperature of 20°C remained optimal for strength. The temperature had greater influence on the UCS of immersed samples as reflected by the much greater UCS values at 20°C than at 5°C and 40°C. This implies that the optimal temperature of 20°C is not only positive for strength development, it is also beneficial for resisting water erosion.

4.1.3 Effect of leachate pH

The effect of leachate pH on the UCS is shown in Figure 8 for both binder types (12C and 6C6G) with
different curing periods (7, 28 and 56 days). The dotted lines relate to UCS values without prior immersion which provide a frame of reference for those conducted after immersion (solid lines). For both binder types, lowest UCS values occur at pH=1 and greatest values at pH=10. This implies that acidity has a significant inhibiting influence on chemical reactions in the samples, with alkaline conditions most beneficial for sediment improvement. This is expected since strongly acidic conditions cause the alkaline materials to dissolve during the leaching process, restricting the hydration reactions and deactivating the GGBS. However, the relatively low pozzolanic reaction between portlandite and silica in cement-based materials can be accelerated by alkali activation such as the NaOH in the leachate solution, further strengthening the sediment structure (Khan et al., 2017). With the exception of the pH=1 case, all UCS values were above the corresponding UCS reference line, indicating that the effect of the additional (100 days) curing while immersed outweighed any negative impacts of water erosion.

The advantage of alkaline conditions can be verified from microstructural observations corresponding to 28 days of curing (Figure 9); a more compact structure occurs at pH=10 (Figure 9a-3) with fewer pores, more cementitious materials and denser soil aggregate compared with pH=1 (Figure 9a-1) and pH=7 (Figure 9a-2) for the 12C-sediment. A comparison of the three SEM images for the 6C6G-sediment at 28 days (Figure 9b), illustrates that the matrix and the cementitious materials on the surface of the sediment aggregates were dissolved at pH=1, resulting in a smooth surface on interior particles, weakening the connection between particles and decreasing interlocking capacity.

In the case of the relatively weak structure of the 12C-sediment after 7 days of curing, the UCS value after 100 days of immersion in a strong acid (pH=1) was substantially lower than the corresponding 7-day UCS reference value. However, the inhibiting effect of the strong acid reduced over time. Therefore, an extended curing time can be considered an efficient means of resisting acid erosion for OPC-sediment. For the 6C6G mixtures, all UCS values (after immersion in strong acid pH=1) were lower than the corresponding reference strength values. In a highly acidic environment, the UCS value is apparently insensitive to curing duration for the GGBS mixtures.
4.2 Leachability

4.2.1 Eluate pH (liquid after leaching)

The term leachate is used to refer to the initial liquid used in the leaching test (i.e. deionized water), whereas eluate refers to the liquid in existence after each leaching period. The eluate pH is used in soil stabilization scenarios as a gauge of the extent of contamination of the surrounding environment. The variation of eluate pH with leaching time is shown in Figure 10. The eluate pH values for the 12C and 6C6G mixtures are presented as a function of curing duration (7, 28, 56 and 98 days) (Figure 10a), where the leachate pH was 7. The large initial increase in pH can be attributed to OH\(^-\) dissolution from the sample matrix into the leaching liquid (Svensson and Allard, 2008). All eluate pH values began to decrease gradually after this peak and the final eluates were alkaline and stable. At all stages of leaching, the relationship between eluate pH and curing time is somewhat variable up to 56 days, but eluate pH values after a period of 98 days were consistently lower than the others. This is not surprising as the sample structure at this time was most stable and the physico-chemical reactions were effectively inactive. Therefore, extended curing periods help reduce the alkalization of the surrounding environment by cement-based materials.

The development of eluate pH over the leaching period as a function of leachate initial pH values (1, 4, 7 and 10) is shown in Figure 10b. With the exception of the pH=1 leachate, eluate pH levels exhibit the same pattern with time as observed in Figure 10(a). It should be noted that for the case of 6C6G in leachate with pH=10, the stable eluate pH is below 10. This observation is attributed to consumption of the OH\(^-\) in the leaching liquid by the chemical reactions in the binder, also resulting in a significant increase in strength. The eluate pH values for the pH=1 leachate increased with leaching time and stabilized at approximately pH=4. The effect of curing temperature on eluate pH was relatively minor (Figure 10b).

In general, the eluate pH values of the OPC-sediment (12C) were greater than those of the OPC/GGBS-sediment (6C6G), although the difference only became apparent after 10 days of leaching.
Beyond 10 days, the eluate pH remained approximately within the range: 9-12 for OPC-sediment and 8-10 for OPC/GGBS-sediment. In the case of the greatest leachate acidity (pH=1), the stabilized eluate pH values for OPC/GGBS-sediment were marginally greater than those of OPC-sediment. Overall, partial replacement of OPC by GGBS facilitated the mitigation of hazardous conditions represented by extreme pH values.

### 4.2.2 Leaching ratio

In order to eliminate the influence of potential sediment heterogeneity and to compare the immobilization levels across all contaminants, all leached element concentrations (after the 100-day period) have been normalized by their corresponding initial total concentrations in the sample. This normalized concentration is subsequently referred to as the leaching ratio in Figure 11-Figure 13. Chromium and Cd were omitted as their concentrations were frequently below the limit of detection of the analytical technique. The leaching ratios for the elements (Al, Mn, Fe, Ni, Cu, Zn, As, Ba and Pb) in the two mixtures (12C and 6C6G) subjected to different curing durations are presented in Figure 11.

The leaching ratio typically reduces with curing duration, attributed to the increasing chemical precipitation of low solubility metal compounds produced from the physico-chemical reactions in the cement-based materials over time, which have a beneficial effect on metal encapsulation and precipitation (Q. Y. Chen et al., 2009). However, the rate of decrease of the leaching ratio with time, herein referred to as the age sensitivity, varies. The greatest age sensitivity occurs for the release of Cu and Ni, whose concentrations decreased significantly after 7 days of curing. However, the age sensitivity of the other elements was moderate, probably due to their relatively low initial concentrations in the samples and rapid incorporation with the early stage hydrates. Interestingly, the strong differences between the 12C and 6C6G mixtures in terms of UCS values is not reflected in the metal release data. Water erosion in the OPC/GGBS mixture is evident from Figure 9b)-2, with the absence of ettringite and fewer cementitious materials when compared to the sample without water immersion (Figure 7b)-2). This dissolution of a
large amount of hydrates has an accelerating effect on metal mobility, resulting in the release of some metals including Al across all curing periods and Cu at 28, 56 and 98 days (even more pronounced for 12C).

The leaching ratios for the elements measured in the 12C and 6C6G mixtures are displayed in Figure 12 as a function of curing temperature. All samples were cured for 28 days at specific temperatures (5, 20 and 40°C) and then subjected to 100-days of leaching using deionized water leachate (pH=7). The encapsulation efficiency is clearly a function of the metal-binder combination and ambient conditions. Continued increase in temperature is beneficial for the immobilization of Al in the 12C-sediment as the cement hydration reactions are accelerated. On the other hand, the minimum Al leaching ratio for the 6C6G sediment occurs at the intermediate temperature of 20°C. This is expected as Al is a dominant participant in the hydration reaction for the formation of trisulfoaluminoferrite hydrates (AFt) and monosulfoaluminoferrite hydrates (AFm) (Liu et al., 2020; Marchon and Flatt, 2016). Elevated temperature also reduces Fe mobility in both samples, with only 0.005% of the original Fe concentration in the 6C6G sediment leached at 40°C. Elevated temperature inhibited the release of Ni, As and Pb in the 12C-sediment and Mn, Ni, Cu and Zn in the 6C6G-sediment. However, Cu, Zn and Ba in the 12C-sediment and Al, As, Ba and Pb in the 6C6G-sediment exhibited an optimal encapsulation temperature of 20°C. Nickel migration in the 12C sediment was evidently greater than that in the 6C6G sediment across all three temperatures. Copper in the 12C sediment at an optimal temperature of 20°C was more stable than that in the 6C6G sediment, while less stable at other temperatures.

Figure 13 shows leaching ratios for the two binder combinations at 7, 28 and 56 curing days, after leaching at pH values of 1, 7 and 10. The greater levels of acidity neutralizes and dissolves the sediment matrix, resulting in increasing solubility of metal-incorporated hydrate phases and therefore a significantly greater leaching ratio at pH=1 was observed. The observed extent of immobilization of the various elements can be summarised as follows:
i. The releases of Al in the 6C6G mixtures at pH=1 and pH=7 after curing times of 7, 28 and 56 days were all greater than corresponding releases for the 12C mixes. This implies that GGBS offers a weaker fixation ability for Al than OPC under acidic and neutral environments. The greater acid and water erosion were also detected in the microstructure of the 6C6G mixtures (Figure 9b)-1 and Figure 9b)-2)) compared with the 12C mixtures (Figure 9a)-1 and Figure 9a)-2), indicating that the greater amount of cementitious materials in the OPC/GGBS mixture dissolved releasing more aluminium hydroxide complexes. This lower buffering capacity of the GGBS mixtures has been previously reported (Q. Chen et al., 2009; Mahedi et al., 2020) and attributed to the higher hydroxyl content of OPC (as shown in section 4.2.1) which is capable of neutralising the acidic solution and therefore decreasing acid erosion. In contrast, the release of Al in the 6C6G-sediment under alkaline conditions (pH=10) was lower than that in the 12C-sediment. This is expected since the NaOH in the leachate can accelerate the activation of GGBS (Cao et al., 2018; Tänzer et al., 2015) and pozzolanic activity which has a significant influence on the leaching of Al and Fe (Goswami and Mahanta, 2007). This also explains the decreasing leaching ratio of Fe in both samples with increasing leachate pH which followed a cationic leaching pattern. Furthermore, the addition of GGBS offered a better fixation capacity for Fe indicated by the greater leaching ratio in the 6C6G mixtures (Figure 11-Figure 13) (Mahedi and Cetin, 2019).

ii. Among the metals studied, Mn showed the most dramatic response to ambient pH as evident from the pronounced difference in leaching ratio between pH=1 and the other pH values. For example, for the 6C6G mixtures after 7 days, the Mn leaching ratio was the highest of all elements (approximately 30%) at pH=1, while it was dramatically lower (0.002%) at pH=7 and 10 due to the dissolution and precipitation of manganese hydroxides. Extremely low release of Mn in cement-based materials has been reported elsewhere (Mahedi et al., 2020).

iii. GGBS is beneficial for immobilizing Ni, like Fe, signifying that pozzolanic reactions have a positive effect on the fixation of Ni. The release of Cu decreased within a certain range of leachate
pH and was minimal at pH=10 after 7 days of curing, probably due to active hydration at an early stage which accelerated the precipitation of copper oxide (CuO) and copper hydroxide (Cu(OH)$_2$) minerals. However, with the decline in physico-chemical reactions and the formation of a relatively stable sediment structure over time, the consumption of hydroxyl (OH$^-$) reduced under alkaline conditions. The excess of hydroxyl further accelerates the dissolution of Cu-bearing minerals, explaining the minimum release of Cu at neutral conditions when the curing duration increased to 28 and 56 days, in keeping with previous findings (Sobiecka et al., 2014; Zhang et al., 2016). The lowest sensitivity to ambient pH occurs for Ba. The maximum leaching ratio for Ba even at pH=1 was only 1.523% (6C6G), and the leaching ratios reduced with increasing pH of the leachate for both binder types.

Overall, although the leaching of the elements in the 12C and 6C6G specimens displays different responses to leachate pH and curing duration, S/S methods offer benefits for the immobilization of Mn, Fe, Zn, As, Ba, Pb in marine sediment with pure OPC and OPC/GGBS in the common pH range from 4 to 10, with >99.9% of metals immobilized in the sediment. The immobilization efficiencies of other elements (Al, Ni and Cu) were relatively greater, ranging from 97.8% to 99.8%. In general, GGBS treatment decreased the potential release of Fe, Ni, Cu, Zn and As at a leachate pH value of 1; of Fe, Ni, Zn, Ba and Pb at a leachate pH of 7 and of Al, Fe, Ni, Zn, Ba and Pb at a leachate pH of 10. The leaching of Cu under neutral and alkaline conditions was highly dependent on the curing duration. The leaching of Cu after 7 days of curing was lower in OPC/GGBS-sediment than that in OPC-sediment alone, whereas the OPC-only mixture was more beneficial for the fixation of Cu after 28 and 56 days of curing. It is clear that differences in effects of binders observed in the unconfined compressive strength tests did not correlate with leaching performance for Al, Mn, Cu, As, Ba and Pb, while a higher strength obtained for cement-based materials can decrease the release of Fe, Ni and Zn.

### 4.2.3 Leaching and leaching mechanism
In order to further understand the process of leaching, a detailed analysis of leaching as a function of time was conducted. In this context, Cr, Cd, Mn and Pb were excluded from the assessment as a result of their low leaching rates.

The cumulative releases of Al, Fe, Ni, Cu, Zn and As under neutral and alkaline conditions over the 100-day period of the tank leaching tests are presented in Error! Reference source not found., in which the cumulative releases are strongly dependent on the leaching step. They increase sharply with time initially, but upon reaching a critical point, the growth rate begins to decrease and becomes stable. This approximately linear increase in metal release with a lower growth rate after the critical time implies that the sediment structure is not stable over the whole leaching period. The spread of the slopes of the cumulative leaching curves under all conditions (i.e. curing time, curing temperature and ambient pH) is greater for some metals than for others. It can be postulated that the release behaviors of any one metal is dependent on the metal/sediment combination, in addition to the ambient influencing factors.

From calculations prescribed in EA NEN-7375 (2004), it can be concluded from the cumulative leaching results that the Al in both 12C and 6C6G specimens under all conditions was leached by dissolution (slope of logarithm of cumulative leaching amount versus logarithm of leaching time plot >0.65). The growth rate for Al was greater than other metals, exhibiting a considerable increase even after the critical point (Error! Reference source not found.). This implies that many Al-based compounds existing in the samples were unstable and soluble in water. However, the other five elements (Fe, Ni, Cu, Zn and As) leached via dissolution at pH=1, and the action of dissolution only occurred before the critical point in the pH=7 and pH=10 cases. This is expected, as soil aggregates are dissolved and soil structure is destroyed in a strongly acidic environment, while under neutral and alkaline conditions, freely soluble metal-based compounds and concentration differences between sample matrix and leachate tended to induce a rapid dissolution of metals into leaching liquids at an early stage of leaching. The dominant mechanisms of the metals over the whole leaching period under neutral and alkaline conditions are
diffusion \((0.35\lessdot\text{slope}\lessdot0.65)\) and depletion \((\text{slope}\lessdot0.35)\).

5 Conclusions

There is now a growing desire internationally to take a more responsible approach to the management of dredged contaminated sediments. The improved physical and chemical stability of mixes incorporating Ground Granulated Blast Furnace Slab (GGBS), relative to Ordinary Portland Cement-only (OPC) mixes, has been demonstrated through laboratory and field trials, primarily on construction projects in Japan and Scandinavia. There is a need to extend the knowledge base to consider different sediment types and importantly, the effect of a range of environmental conditions on strength and contaminant encapsulation potential, through systematic study. Based on unconfined compressive strength tests and tank leaching tests on a marine sediment stabilized with two different binder combinations (12% OPC and 6% OPC + 6% GGBS), the findings of this study include:

(i) GGBS (as a partial replacement for OPC) offers an effective means of improving the unconfined compressive strength of the sediment and provides excellent immobilization of Fe, Ni and Zn over the range of environmental conditions considered in this study. For other metals (Al, Mn, Cu, As, Ba and Pb), the release behaviours are a more complex function of the physico-chemical parameters examined (i.e. stabilizer, curing duration, curing temperature and ambient liquid pH).

(ii) Curing duration increases the UCS of S/S treated sediment and may offer benefits for both water and acid erosion. It also reduces metal mobility, with Cu and Ni exhibiting the greatest sensitivity to curing duration. GGBS is beneficial for immobilizing Ni irrespective of curing duration and temperature but is less efficient for the stabilization of Cu over longer curing periods (28, 56 and 98 days in this study) at 20 °C.

(iii) The strength of the OPC-only sediment increases with temperature, whereas an optimum
temperature of 20°C applies to the OPC/GGBS sediment due to the occurrence of a crossover effect. The leaching of Cu, Zn, Ba content for the OPC-sediment and Al, As, Ba, Pb content for the OPC/GGBS-sediment have an optimal temperature of 20°C for encapsulation.

(iv) Acidity has a significant inhibiting influence on sediment strength while alkaline conditions facilitate improvement of sediment strength. The leachability of the elements considered also illustrates significant pH dependence, except for Ba. The release of Al in GGBS mixtures is greater than in OPC mixtures at pH=1 and 7 while more stable under alkaline conditions. Manganese is most sensitive to pH with a high leaching ratio of approximately 30% at pH=1, while remarkably this value decreases to 0.002% at pH=7. The minimum release of Cu occurs under neutral conditions at longer curing periods of 28 and 56 days.

(v) The immobilization of metals was strongly dependent on the stage of leaching and cumulative leached concentrations initially increased sharply with time, but upon reaching a threshold, increased approximately linearly displaying a reduced growth rate. Cumulative leaching of Al increased most rapidly with leaching time and leached out by the way of dissolution. For heavy metals (Ni, Cu, Zn and As), dissolution only occurred in a strongly acidic environment, with diffusion and depletion acting as the predominant leaching mechanism under neutral and alkaline conditions.

While it is not possible to make overarching statements about the effectiveness of GGBS over OPC from a contaminant encapsulation point of view, due to the variety of contaminants that may be discovered in sediment and the variation in physio-chemical conditions possible; the research shows that GGBS can offer technical advantages over OPC as a management option in many practical scenarios. When coupled with its well-known sustainability credentials, it is clear that GGBS should be considered much more widely in stabilization/solidification problems than is currently the case.
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Figure 1 Particle size distribution curves for parent sediment, GGBS and OPC

Figure 2 SEM image of fresh sediment
Figure 3 Schematic of tank leaching test

Figure 4 Effect of curing duration on unconfined compressive strength (UCS)
Figure 5 SEM images of sediment with: a) 12C; b) 6C6G at curing times of 7, 28 and 56 days

Figure 6 Effect of curing temperature on UCS for specimens with and without 100 days of immersion/leaching
a) - 1 5°C

b) - 1 5°C

a) - 2 20°C

b) - 2 20°C
Figure 7 SEM images of sediment with: a) 12C; b) 6C6G at curing temperatures of 5, 20 and 40 °C

Figure 8 Relationship between leachate pH and UCS of sediment with: a) 12C; b) 6C6G
a) $\text{pH} = 1$

b) $\text{pH} = 1$

a) $\text{pH} = 7$

b) $\text{pH} = 7$
Figure 9 SEM images of 28-day sediment with a) 12C and b) 6C6G after leaching with pH= 1, 7 and 10
(a) different curing times

(b) different curing temperatures and leachate pH values

Figure 10 Eluate pH values at each test period:

(a) different curing times, (b) different curing temperatures and leachate pH values
Figure 11 Leaching ratios for samples (12C and 6C6G) with different curing durations of 7, 28, 56 and 98 days (leachate pH=7)

Figure 12 Leaching ratios for samples (12C and 6C6G) at different curing temperatures of 5, 20 and 40 °C (leachate pH=7)
Figure 13 Leaching ratios for samples (12C and 6C6G) with 7, 28 and 56 days of curing at leachate pH=1, 7 and 10
Figure 14 Relationship between cumulative leaching of the elements and leaching time

(Note: unit of curing duration t is days; unit of temperature T is °C)
Table 1. Chemical constituents of the parent sediment, OPC and GGBS binders

|       | SiO₂ | Al₂O₃ | Fe₂O₃ | CaO | MgO | SO₃ | Mn₂O₃ | TiO₂ | K₂O | Na₂O | BaO |
|-------|------|-------|-------|-----|-----|-----|-------|------|-----|------|-----|
| (%)   | (%)  | (%)   | (%)   | (%) | (%) | (%) | (%)   | (%)  | (%) | (%)  | (%) |
| Sediment | 75.51         | 8.225          | 3.335          | 1.32 | 0.755 | 1.9 | 0.03  | 0.64 | 1.45 | 1.11 | 0.21 |
| OPC   | 19.9          | 5.1             | 2.7             | 64.5 | 1.2  | 2.3 | -     | -    | 0.52 | 0.22 | -   |
| GGBS  | 34.11         | 11.16           | 0.85            | 41.1 | 6.57 | 0.1 | 0.25  | 0.87 | 0.48 | 0.43 | -   |
Table 2 Element concentrations, limit of determination (LOD) and metal limits (ppm) from selected standards.

| Element | Concentration  | LOD | Irish | Soil Guideline Values (UK) | Dutch |
|---------|----------------|-----|-------|---------------------------|-------|
|         | 12C | 6C6G |       | Residential/ Allotments | Commercial | Target | Intervention |
| Al      | 14114 | 15852 | 0.326 | -                         |       |
| Cr      | 24.32 | 24.04 | 0.123 | 370 | 130 | 5000 | 100 | 380 |
| Mn      | 282.9 | 310.8 | 0.087 | -                         |       |
| Fe      | 18880 | 18591 | 1.521 | -                         |       |
| Ni      | 24.23 | 22.75 | 0.28  | 60 | 50 | 5000 | 35 | 210 |
| Cu      | 38.48 | 32.37 | 0.736 | 110 |       | 36 | 190 |
| Zn      | 156.5 | 151.6 | 0.222 | 410 |       | 140 | 720 |
| As      | 22.62 | 23.06 | 0.153 | 70 | 20 | 500 | 29 | 55 |
| Cd      | 0.76  | 0.66  | 0.016 | 4.2 | 1.8 | 1400 | 0.8 | 12 |
| Ba      | 830.5 | 978.5 | 0.014 | - |       | 160 | 625 |
| Pb      | 60.83 | 66.10 | 0.062 | 218 | 450 | 700 | 85 | 530 |