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Stabilisation of a dredged marine sediment: geotechnical and contaminant leaching properties

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ABSTRACT: In this paper, contaminated sediment dredged from a commercial harbour in Ireland was stabilized using Ordinary Portland cement (OPC) alone and in combination with Ground Granulated Blast Furnace Slag (GGBS). The engineering characteristics of the stabilized sediments were established from Unconfined Compression Strength tests. Tank leaching tests were conducted to investigate the long term release of 11 elements (Al, Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, Ba, Pb) relative to their original concentrations in the stabilized sediment. Leached concentrations after 100 days were below 1% of the original concentration for most elements. Cement replacement with suitable amounts of GGBS increased stabilized strength and also decreased the extent of leaching of Al, Cr, Mn, Fe, Ni, Cu and Pb. The performance of OPC alone was superior for restricting leaching of Zn, Cd and As. Both Al and Fe are key elements in the stabilization process; the results suggest that Al stabilizes more slowly than Fe. Given that GGBS is both a more cost-effective and more environmentally-friendly material than OPC and that it is commonly used in concrete, its potential for stabilization of sediments warrants further study.

KEY WORDS: Dredged sediment; stabilization; unconfined compression strength; contamination; metals; tank leaching test.

1 INTRODUCTION

Dredging is carried out periodically in channels and ports to maintain sufficient water depth for navigation. As a result, large amounts of sediment are moved from an aquatic to a terrestrial environment annually. Traditional management strategies for these sediments include landfilling, confined aquatic disposal or disposal directly at sea. However, these strategies have disadvantages such as high cost, capacity limitations and potential environmental pollution [1,2]. As a result, there is an emerging need for cost-effective methods of treating and/or re-using dredged material.

One management option for dredged sediments is to reuse them as a construction material. The parent sediments typically have a high moisture content and therefore 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 (PCB)) as a result of natural and anthropogenic activities [1]. Solidification/stabilization using a cementitious additive is widely acknowledged as an effective method of improving engineering properties of sediments and encapsulating contaminants [2–4]. The ability of Ordinary Portland Cement (OPC) to improve geotechnical characteristics [5–8] and immobilise contaminants [9–11] has been documented widely. Appropriate OPC replacement with Ground Granulated Blast Furnace Slag (GGBS), a by-product of the steel industry, optimizes the engineering properties of treated soil (strength, swelling, resistance to sulphate attack, etc.) [12,13] and further inhibits the leachability of contaminants [9,14,15].

Leaching tests are important in the environmental assessment of solidified and stabilized (S/S) contaminated sediments. Two broad categories of leaching tests exist: extraction and dynamic tests, differentiated by whether or not equilibrium is established during the test period [15]. Dynamic diffusion has been shown to be more representative of the long-term leaching behaviour of S/S contaminated sediments [16]. In this paper, a dynamic tank leaching test was chosen to measure the leaching behaviour of a marine sediment stabilised with various compositions of OPC and GGBS. The total concentration of priority metal pollutants in the sediment was determined using aqua regia extraction tests. The effect of different binder combinations/dosages on the Unconfined Compression Strength was also investigated.

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 and stored in a freezer (-20°C) until time of use. The sediment comprised 82.9% sand, 16.4% silt and 0.7% clay (average from n=3 samples), classifying as a silty SAND. The particle size distribution is shown in Figure 1, with D10 and D90 values of 0.015 mm and 0.17 mm respectively. The natural (gravimetric) water content of the sediment fell in the range 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 shown in Figure 1. The sediment particle size distribution is typically an order of magnitude larger than that of the GGBS and OPC; with the GGBS particle size distribution marginally smaller than that of the OPC. Blaine fineness values for OPC and GGBS were distinctly different at 3520 cm²/g and 4490 cm²/g respectively. The chemical constituents of both binders are listed in Table 1.
Figure 1. Particle size distributions for sediment, GGBS and OPC

Table 1. Chemical constituents of OPC and GGBS

<table>
<thead>
<tr>
<th>Chemical</th>
<th>SiO₂ (%)</th>
<th>Al₂O₃ (%)</th>
<th>Fe₂O₃ (%)</th>
<th>CaO (%)</th>
<th>MgO (%)</th>
<th>SO₃ (%)</th>
<th>Mn₃O₄ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>OPC</td>
<td>19.9</td>
<td>5.1</td>
<td>2.7</td>
<td>64.5</td>
<td>1.2</td>
<td>2.3</td>
<td>-</td>
</tr>
<tr>
<td>GGBS</td>
<td>34.11</td>
<td>11.16</td>
<td>0.85</td>
<td>41.1</td>
<td>6.57</td>
<td>0.1</td>
<td>0.25</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Chemical</th>
<th>TiO₂ (%)</th>
<th>K₂O (%)</th>
<th>Na₂O (%)</th>
<th>Cl (%)</th>
<th>Ca₃A (%)</th>
<th>FCaO (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>OPC</td>
<td>-</td>
<td>0.52</td>
<td>0.22</td>
<td>0.08</td>
<td>9.1</td>
<td>1.8</td>
</tr>
<tr>
<td>GGBS</td>
<td>0.87</td>
<td>0.48</td>
<td>0.43</td>
<td>0.01</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

The binders were added as a proportion of the dry mass of the sediment and a total of 8 different compositions (T1-T8) were developed (Table 2) with the following test goals:
- T1, T2, T6, T7 and T8 were prepared to investigate the effect of OPC content on the Unconfined Compression Strength (UCS).
- T2, T3, T4 and T5 (all 6% binder mixes) were prepared to evaluate the effect of the replacement of OPC with GGBS on the UCS. The term κ is hereafter used to refer to the ratio of GGBS to OPC in a particular mix (ranging from 0 to 2 in this study).
- Specimens of T2, T4 and T5 were prepared for tank leaching tests.

Table 2. Composition of samples

<table>
<thead>
<tr>
<th>Sample I.D.</th>
<th>T1</th>
<th>T2</th>
<th>T3</th>
<th>T4</th>
<th>T5</th>
<th>T6</th>
<th>T7</th>
<th>T8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Composition (C = % OPC, G = % GGBS)</td>
<td>4C</td>
<td>6C</td>
<td>3C</td>
<td>3G</td>
<td>2C</td>
<td>4G</td>
<td>2G</td>
<td>8C</td>
</tr>
<tr>
<td>OPC (% of dry weight)</td>
<td>4</td>
<td>6</td>
<td>3</td>
<td>2</td>
<td>4</td>
<td>8</td>
<td>12</td>
<td>16</td>
</tr>
<tr>
<td>GGBS (% of dry weight)</td>
<td>0</td>
<td>0</td>
<td>3</td>
<td>4</td>
<td>2</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>κ = GGBS/OPC</td>
<td>0</td>
<td>0</td>
<td>1</td>
<td>2</td>
<td>0.5</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

Perspex moulds (100 mm high × 50 mm diameter) were used to sample for the UCS and tank leaching tests after thorough mixing. The specimens were removed from the moulds when the structure was formed, and subsequently cured in a sealed plastic bag at 20 ± 2 °C.

3 TEST PROGRAMME

UCS tests were conducted on the stabilized sediments after curing times of 7, 28 and 56 days using a UTSO12 Fully-Automated Cyclic Triaxial (FACT) Testing Machine. The loading rate used was 1.0 mm/min. Three replicate UCS tests were carried out for each of T1 to T8.

In addition, 250 mg sediment samples (dried in an oven at 105 °C over a 24 h period and sieved to eliminate particles greater than 2 mm in size) were mineralized and dissolved using aqua-regia (HCl:HNO₃ in the ratio 3:1) in a closed vessel microwave digestion system (Figure 2) (MILESTONE, 1200-Mega, Bergamo, Italy). The suspensions obtained were filtered (Whatman No. 541) and diluted to 50 ml prior to analysis by Inductively-Coupled Plasma Mass Spectrometry (ICP-MS) (PerkinElmer, Elan DRCe, USA). The method was validated by using Certified Reference Materials (CRMs) (NIST SRM 2711a and Sewage Sludge 2: CRM029-50G).

Figure 2. Milestone 1200-mega microwave digestion system

Tank leaching tests (Figure 3) were performed following the NEN-7375 (2004) standard [18] using a liquid/solid ratio of 3.5 (permissible range of 2-5) and observing an extended leaching time of 100 days (tests are typically 64 days long).

Figure 3. Tank leaching test
The elemental concentrations in the sediments (in parts per million, ppm) are presented in Table 3, along with Soil Guideline Values (SGVs) from the UK [19] and Dutch target and intervention values for soil remediation [20]. It can be observed from Table 4 that As (arsenic) is the only element exceeding the Dutch intervention values, whereas the concentrations of Zn, Ba and Cd exceed their Dutch target values. The Ba concentration also exceeds the Dutch intervention values.

Table 3. Element concentrations and metal limits (ppm)

<table>
<thead>
<tr>
<th>Chemical element</th>
<th>Concentration (ppm)</th>
<th>UK SGVs [19]</th>
<th>Dutch [20]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Residential/Allotments</td>
<td>Commercial</td>
<td>Target</td>
</tr>
<tr>
<td>Al</td>
<td>14471.07</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Cr VI</td>
<td>22.34</td>
<td>130</td>
<td>5000</td>
</tr>
<tr>
<td>Mn</td>
<td>287.23</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Fe</td>
<td>19715.96</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Ni</td>
<td>24.26</td>
<td>50</td>
<td>5000</td>
</tr>
<tr>
<td>Cu</td>
<td>34.13</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Zn</td>
<td>142.09</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>As</td>
<td>22.52</td>
<td>20</td>
<td>500</td>
</tr>
<tr>
<td>Ba</td>
<td>877.8</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Cd</td>
<td>0.93</td>
<td>1-8</td>
<td>1400</td>
</tr>
<tr>
<td>Pb</td>
<td>65.11</td>
<td>450</td>
<td>700</td>
</tr>
</tbody>
</table>

At lower levels of cement replacement with GGBS (i.e. \( \kappa \leq 1 \)), the GBS/OPC combinations lead to improved UCS values (Figure 4). This is expected because the latent cementitious reactivity of GGBS is activated by hydration materials, such as alkaline activators Ca(OH)\(_2\), NaOH and KOH, which can break the Si-O and Al-O bonds [25]. While it is not well understood how the activator works on the GGBS, previous research [26] has suggested that cement-hydration reactions, slag-hydration reactions and some interaction between them can be expected. In addition to these three reactions, it is conceivable that there are some activators within this marine sediment itself giving rise to the higher strengths in comparison with those for OPC-only mixes. However, when the proportion of GGBS is excessive (i.e. the \( \kappa = 2 \) case in Figure 4), these reactions are not sufficiently activated, leading to a decrease in strength.

5.2 Leaching properties
5.2.1 Immobilization efficiency of different stabilizers

The concentrations of leached elements (over the 100 day period) were normalized by their initial concentrations in the stabilized sediment, in order to eliminate the influence of the sediment heterogeneity. The impacts of different stabilization strategies on this leaching ratio are shown in Figure 5. The highest immobilization levels were achieved for Mn, Fe and Pb (lower than 0.0131%). For other target elements, the stabilization treatment was effective in decreasing the mobility (the leaching ratio varied from 0.0234% to 1.36%).

![Figure 4. Effect of OPC and GGBS on UCS](image)

![Figure 5. Leaching of metals as a proportion of initial concentrations](image)

The effectiveness of the various binders over 100 days can be summarized as follows (> signifies more effective):
- Al, Cr, Mn, Ni, Cu and Pb:
  - \( \kappa = 2 \) (4C2G) > \( \kappa = 0.5 \) (2C4G) > \( \kappa = 0 \) (6C)
- **Fe**: $\kappa=0.5 \approx \kappa=2 > \kappa=0$
- **Zn, Cd**: $\kappa=0 > \kappa=2 > \kappa=0.5$
- **As**: $\kappa=0 > \kappa=0.5 > \kappa=2$
- **Ba**: $\kappa=0.5 > \kappa=0 > \kappa=2$

GGBS achieves a better efficiency for the immobilization of Al, Cr, Mn, Ni, Cu and Pb, and the benefit of replacing cement with GGBS for the stabilisation of Cr and Pb found here is consistent with other publications [27, 28]. In the cases of Zn, Cd and As [29], the performance of OPC alone is superior to that of blended OPC/GGBS mixes. Similar Fe concentrations at $\kappa=0.5$ and $\kappa=2$ may suggest that a limiting value is reached at the former ratio. The highest immobility level for Ba was observed at $\kappa=0.5$ which is consistent with the trend between UCS and $\kappa$.

### 5.2.2 Leaching characteristics

The cumulative releases of Al, Fe, Ni, Cu, Zn and Ba over the course of the tank leaching test period are presented in Figure 6 (Mn, Cd and Pb are not shown given their low normalized values in Figure 5). The cumulative leaching increased sharply with time initially, but upon reaching a critical point (defined as the intersection of the linear portions as shown in Figure 6), the rate decreased dramatically. It should be noted that curve shapes and critical point times are similar for all three binder compositions for the metals tested, suggesting that the leaching mechanism is independent of binder types considered here.

It is well known that aluminium and iron oxide minerals play a major role in stabilizing soil structure, leading to a decrease in soil dispersion and an increase in microaggregation [30]. The critical point times of Al (which varied from 17.6 to 20.6 days) are greater than those of Fe (5.2 to 6.6 days), implying that the stability of Al occurs more slowly than that of Fe. Furthermore, the aggregates and structure produced by Fe ions may be more stable than Al.

In case of the Ni and Cu, a rapid increase in leaching occurred after 64 days and was particularly obvious for sediment stabilized with pure OPC, indicating that OPC replacement results in lower releases and slower leaching rates. The trend displayed for Zn is less clear. The results also showed that there was little difference between GGBS and OPC in relation to the leaching of Ba.
6 CONCLUSIONS

Binders incorporating GGBS are well known to be more cost effective and environmentally friendly than cement-only binders. In this study, the efficiency of various combinations of OPC and GGBS in stabilizing a marine silty sand sediment was investigated in terms of geotechnical and metal leaching properties. The main conclusions are as follows:

- Greater UCS values can be obtained by partial replacement of OPC by GGBS. For the marine sediment studied here, a GGBS-OPC ratio of 1:1 provided the highest strengths.

- The maximum leaching ratio (leached concentration to initial concentration) recorded after leaching for 100 days was 1.36% (for Cu in 6C sediment). The minimum leaching ratio was 0.0023% (for Mn in 2C4G sediment).

- Increased GGBS content resulted in more efficient immobilisation of Al, Cr, Mn, Ni, Cu and Pb. GGBS has benefits for leaching of Fe but it may be insensitive to the exact quantity. In the cases of Zn, Cd and As, the performance of OPC alone was superior to that of OPC and GGBS combined.

- The variation in cumulative leaching concentration with time suggests that the leaching mechanism is independent of the binder combinations studied.

- The time to the ‘critical point’ and flatter curves thereafter suggest that the stability of Al occurs more slowly than that of Fe. It is suggested that the aggregates and structure produced by Fe ions are more stable than that by Al.

This research confirms that for the silty sand sediment considered, OPC replacement with GGBS mixes can be used effectively to optimise the engineering properties and the leaching potential for most of the elements studied. Further research is required to investigate appropriate encapsulation strategies in other sediment types.

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REFERENCES:


