Article

Strength and Contaminant Toxicity Leaching Characteristics of MgO-Solidified Silt

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Abstract: In this study, MgO as an environmentally friendly silt-solidifying material was first mixed with silt and then carbonized by injection with CO2. The strength and contaminant leaching characteristics of the MgO-solidified silt were studied using unconfined compressive strength and toxicity leaching tests, and the results were compared with those of cement-solidified silt. The unconfined compressive strength of the silt reached 111 kPa with 9% MgO content and a 14 d curing time. The CO2 injection further increased the unconfined compressive strength of the MgO-solidified silt by approximately 25%; the values for MgO-solidified silts without and with a CO 2 injection were approximately 60% and 80%, respectively, of those of the cement-solidified silts with the same additive additions. The leaching concentrations of nutrient salts and heavy metal pollutants in the silt decreased with increased MgO content. Compared with the dredged silt, MgO solidification with carbonization reduced the leaching of total nitrogen and total phosphorus by more than 10% and 50%, respectively; these values were approximately 5% points higher than those of cement-solidified silt. Of the heavy metals, the leaching concentration of Ni was reduced the most. This study provides a theoretical basis and technical support for low-carbon treatment and green resource utilization of dredging silt.

Keywords: dredged silt; unconfined compressive strength; contaminant leaching; magnesium oxide; carbonization

1. Introduction

Ecological dredging has been developed on a large scale by the Chinese water environment governance project to reduce the load of endogenous pollution and improve the water quality of river channels, lakes, and reservoirs [1,2]. Over 500 million m3 of dredged silt is produced each year [3]. This silt has a high moisture content, low strength, poor stability, low loading capacity, and contains large amounts of pollutants [4–6]. Dredged silt cannot be directly landfilled and, if not properly handled, can easily cause secondary pollution [7–9]. Solidification, as one of the most common methods for silt treatment, can not only improve the strength of the silt, but can also stabilize contained pollutants [10–12].

Cement is widely used for silt solidification; however, significant CO2 emissions are associated with its production, estimated at 1 t CO2/t cement [13,14]. This is not conducive to the realization of China’s strategic goal of carbon neutralization [15]. In addition, cement is generally alkaline and has an adverse effect on the environment [11,12]. Therefore,
the exploration of low-carbon and environmentally friendly solidifying materials addresses an urgent problem.

It has been reported that MgO is effective at solidifying soft soil [16,17]. It was found that the abundant prismatic-like nesquehonite and rosette-like hydromagnesite or dypingite can be produced by adding MgO and injecting CO2 into soft soil, which leads to an increase in the maximum dry density of soft soil and reduces its optimal moisture content [16]. Furthermore, the unconfined compressive strength (UCS) of reactive MgO-stabilized soil significantly increased after CO2 carbonization for several hours: the UCS increased with an increasing MgO–soil ratio and carbonization time [16]. Liu et al. [17] studied the effect of the initial moisture content and carbonization time on the strength of silty clay that was stabilized by reactive MgO and found that the highest strength of the stabilized clay was achieved after 6 h of carbonization. Wang et al. [18] investigated the influence of CO2 infiltration on the mechanical properties of MgO–fly ash–solidified sludge and found that the production of magnesium carbonates is the intrinsic mechanism explaining why the macro- and microscopic characteristics of carbonated sludge are greatly enhanced by carbonation–solidification, which leads to the transformation of interaggregate pores to interparticle pores. It is, however, not clear whether the use of MgO alone as a solidifying material can improve the strength of dredged silt and stabilize contained pollutants.

In this study, dredged silt from Lake Taihu, China, was collected for study. The silt was solidified by adding MgO and carbonized by a CO2 injection. The effects of these treatments were evaluated by the UCS and toxicity characteristic leaching tests, and the results were compared with those of cement-solidified silt. Combined with the results of scanning electron microscopy (SEM) and X-ray diffraction (XRD) tests, how MgO influences the strength and toxicity leaching characteristics of solidified silt was revealed. The objective of this study was to evaluate the feasibility of using MgO as a solidifying material for dredged silt.

2. Materials and Methods

2.1. Dredged Silt

Dredged silt samples were taken from an ecological dredging project on the west bank of Lake Taihu, China, and stored at 20 °C. The basic properties of the dredged silt, determined according to the Chinese Standard Geotechnical Test Method [19], are shown in Table 1. The moisture content of the silt was 60.74% and the density was 1.72 g/cm³. The concentrations of total nitrogen (TN), total phosphorus (TP), and heavy metals in the silt samples were analyzed using spectrophotometry and inductively coupled plasma–mass spectrometry [20].

Table 1. Basic properties of dredged silt from Taihu Lake.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Moisture content (%)</td>
<td>60.74</td>
</tr>
<tr>
<td>Liquid limit (%)</td>
<td>61.50</td>
</tr>
<tr>
<td>Plastic limit (%)</td>
<td>25.80</td>
</tr>
<tr>
<td>Density (g/cm³)</td>
<td>1.72</td>
</tr>
<tr>
<td>Organic content (%)</td>
<td>3.30</td>
</tr>
<tr>
<td>Sand fraction content (%)</td>
<td>2</td>
</tr>
<tr>
<td>Silt fraction content (%)</td>
<td>78</td>
</tr>
<tr>
<td>Clay fraction content (%)</td>
<td>20</td>
</tr>
</tbody>
</table>

Leaching tests were conducted using the Solid Waste–Extraction Procedure for Leaching Toxicity using an acetic acid buffer solution to obtain the leaching concentrations of each pollution index [21], as shown in Table 2. The concentrations of TN and TP in the silt were significantly higher than those of the metal pollutants. The concentrations of TN in the as-dredged silt and leached silt were 13,720.5 mg/kg and 58.86 mg/kg, respectively,
and those of TP were 545.9 mg/kg and 2.80 mg/kg, respectively. Of the heavy metal pollutants, the total amounts of Cr, Ni, and Cu were higher than those of Zn and Pb, and the Cu leaching concentration was the highest. The light MgO used in this study was produced by industrial calcination (750–1000 °C) and obtained from Tianjin Zhiyuan chemical reagent Co. Ltd., Tianjin, China, and had a purity exceeding 98%. The CO2 was industrially produced by Nanjing Sanqiao special gas Co. Ltd., Nanjing, China, and had a purity exceeding 99%. Ordinary Portland cement (PC 42.5) was employed.

Table 2. Total and leaching concentrations of pollution indicators of dredged silt (unit: mg/kg).

<table>
<thead>
<tr>
<th>Indicators</th>
<th>Total Concentration</th>
<th>Leaching Concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>TN</td>
<td>1372.05</td>
<td>58.86</td>
</tr>
<tr>
<td>TP</td>
<td>545.90</td>
<td>2.80</td>
</tr>
<tr>
<td>Cr</td>
<td>117.20</td>
<td>1.43</td>
</tr>
<tr>
<td>Ni</td>
<td>91.13</td>
<td>0.52</td>
</tr>
<tr>
<td>Cu</td>
<td>21.58</td>
<td>2.19</td>
</tr>
<tr>
<td>Zn</td>
<td>1.95</td>
<td>Not detected</td>
</tr>
<tr>
<td>Pb</td>
<td>1.67</td>
<td>Not detected</td>
</tr>
</tbody>
</table>

2.2. Test Methods

MgO contents of 5%, 7%, and 9% by mass of dry silt mass were considered. After fully mixing and stirring the silt and MgO, the mixture was filled into the cutting rings with a diameter of 39.1 mm and a height of 80 m. The solidified silt samples were cured in a constant-temperature and -humidity curing box at 20 °C and humidity above 95% for 3, 7, 14, and 28 d. The samples were then demolded and placed in a homemade CO2-injection device for carbonization at a pressure of 0.8 MPa for 2 h (see Figure 1). The device is a polymethyl methacrylate barrel with a diameter of 15 cm and a height of 20 cm, which is provided with an inlet valve and an outlet valve at the top and bottom, respectively. To compare the solidification effect of MgO alone, two tests were performed without the CO2-injection treatment. Samples containing the same additions of cement were also evaluated.

The UCS of the samples prepared under different conditions was measured according to the Chinese Standard for Geotechnical Testing [19]. Contaminant leaching concentrations were obtained according to the Solid Waste–Extraction Procedure for Leaching Toxicity—acetic acid buffer solution method [21]. To further understand the microscopic details of the reaction, the raw silt and MgO-solidified silts with and without CO2 injection and cured for 14 d were ground into fine powder, dried, and subjected to SEM and an XRD analysis.
3. Test Results

3.1. Unconfined Compressive Strength

Figure 2 shows the effect of the MgO content on the UCS of the solidified silt. The UCS values increased with the MgO content. For curing times of 3, 7, 14, and 28 d, respectively, the UCS increased from 8.3, 10.2, 12.3, and 14.3 kPa for 5% MgO content to 42.2, 73.1, 111.7, and 164.2 kPa for 9% MgO content. These values represent respective increases by factors of 4.08, 6.17, 8.08, and 10.48. These data indicate that longer curing time resulted in a more significant effect of the MgO content on the UCS of the solidified silt. The UCS values increased further after carbonization by the CO2-injection treatment. For example, the values for MgO-solidified silt with 9% MgO addition were 21.09%, 18.06%, 17.91%, and 17.36% higher with the CO2 injection than without the CO2 injection for curing times of 3, 7, 14, and 28 d, respectively. Improvement in the UCS was most obvious at a curing time of 3 d.

The average UCS values of the MgO-solidified silts at different curing times reached 61.64%, 68.19%, and 74.85% of those of cement-solidified silts with solidifying material contents of 5%, 7%, and 9%, respectively. When the MgO-solidified silts were subjected to a CO2 injection, these values reached 83.82%, 86.92%, and 88.73%, respectively. Therefore, the UCS of the MgO-solidified silt approached that of cement-solidified silt with an increase in the MgO content.

Figure 3 shows the effect of curing time on the UCS of the solidified silt. For curing times from 3 to 28 d, the UCS of solidified silt with MgO contents of 5%, 7%, and 9% increased from 8.3, 26.6, and 42.2 kPa to 14.3, 92.9, and 164.2 kPa, respectively, representing increases of 72.29%, 249.3%, and 289.1%, respectively. This result indicates that higher MgO content resulted in a more significant effect of curing time on the UCS of the solidified silt. Moreover, the UCS increased more rapidly in the early stages of curing and then slowed down: for curing times from 3 to 14 d, the UCS of the solidified silt increased by 48.19%, 156.4%, and 164.4% for MgO contents of 5%, 7%, and 9%, respectively, compared with increases of 16.26%, 36.22%, and 47.00% for the curing period from 14 to 28 d, respectively.

The effect of curing time on the strength of MgO-solidified silts with a CO2 injection was similar to those without a CO2 injection. The average UCS values of the MgO-solidified silt without a CO2 injection reached 70.70%, 70.24%, 67.89%, and 64.08% of those of the cement-solidified silt at curing times of 3, 7, 14, and 28 d, respectively; the analogous values for samples with CO2 injection reached 87.44%, 87.92%, 87.10%, and 83.51. Therefore, the shorter the curing time, the closer the UCS of the MgO-solidified silt was to that of the cement-solidified silt.
Figure 2. Effect of MgO content on unconfined compressive strengths (UCS) of the dredged silt.

Figure 3. Effect of curing time on unconfined compressive strengths (UCS) of the dredged silt.
3.2. Contaminant Leaching

Figures 4–8 show the leaching concentrations of nutrient salts (TN and TP) and heavy metal pollutants (Cu, Cr, and Ni) in the dredged silt following the different treatment methods. Figure 4 shows that the TN concentrations in the leached MgO-solidified silt with MgO contents of 5%, 7%, and 9% were 58.23, 57.91, and 55.86 mg/kg, respectively. Higher MgO content resulted in a lower leaching concentration. Compared with the as-received silt sample, the TN leaching concentrations decreased by 1.07%, 1.61%, and 5.10%, respectively, but were still 33% higher than those of cement-solidified silt with the same additive content. Following the CO2 injection, the TN leaching concentrations of the MgO-solidified silts were further reduced by 0.21%, 10.65%, and 10.15%, respectively, which was approximately 28% higher than the equivalent values for the cement-solidified silts.

Figure 4. Variation in TN leaching concentration under different treatments.

Figure 5 shows that the effect of MgO on the TP leaching concentration was much more significant than that of TN. The TP concentrations of the leached solidified silts with MgO contents of 5%, 7%, and 9% were 1.50, 1.44, and 1.37 mg/kg, respectively. These values were, respectively, 46.43%, 48.57%, and 51.07% lower than those of the as-received silt sample but 11.8% higher than those of the cement-solidified silts. The TP concentrations of the leached MgO-solidified silts with the CO2 injection were, respectively, 6.00%, 6.94%, and 8.03% lower than those without the CO2 injection, which was approximately 5% points higher than those of the cement-solidified silt.

Figures 6–8 show that the variations in the leaching concentrations of Cu, Cr, and Ni were similar to those of TN and TP and decreased with increasing MgO content. Compared with the as-received silt sample, the concentrations of Cu, Cr, and Ni in the leached solidified silts with MgO contents of 5%, 7%, and 9% with the CO2 injection decreased by 1.09–47.54%, 7.69–65.38%, and 15.38–45.45%, which were 4.7%, 10.7%, and 33.3% higher than those of the respective cement-solidified silts. MgO solidification with the CO2 injection had the most significant impact on the leaching of Ni, with a reduction in the Ni leaching concentration of more than 65%, but the values for Cu leaching from the MgO-solidified silt were closest to those of the cement-solidified silt.
Figure 5. Variation in TP leaching concentration under different treatments.

Figure 6. Variation in Cu leaching concentration under different treatments.

Figure 7. Variation in Cr leaching concentration under different treatments.
4. Discussion

Mechanism of MgO Solidification and CO₂ Carbonization

The as-dredged silt and MgO-solidified silts with and without a CO₂ injection with different MgO contents were examined by SEM to reveal the mechanism of MgO solidification and CO₂ carbonization. Figure 9 shows the respective SEM images. Figure 9a shows that connections between the as-received silt particles were relatively loose and the particles had obvious pores. In the MgO-solidified silt samples without a CO₂ injection (Figure 9b–d), obvious flocculated material filled the surface pores. This was identified as brucite (Mg(OH)₂) that had been generated by the MgO addition. As the MgO content increased, the pore size significantly reduced and the pores were gradually filled. The strength of the silt after solidification with MgO was therefore enhanced. In the SEM image of the silt solidified with 5% MgO with a CO₂ injection (Figure 9b–d), obvious flocculated material filled the surface pores. This was identified as hydromagnesite (Mg₅(CO₃)₄(OH)₂·4H₂O) and dypingite (Mg₅(CO₃)₄(OH)₂·5H₂O) that were produced by carbonization. With an increase in the MgO content, the silt boundaries gradually blurred, the pores were progressively refined, and an increase in the amount of flake-like cementing material was observed on the silt surface. When the MgO content was 9%, the silt pores were almost completely filled and the surface was covered by flake-like and prismatic material (nesquehonite (MgCO₃·3H₂O)), which showed an overall dense block structure. Although the pores of the solidified silt without the CO₂ injection were almost filled, the filling was relatively loose: the structure of the MgO-solidified silt with the CO₂ injection was denser, and cementation between the silt particles was stronger.
Figure 9. SEM images of the dredged silt under different conditions.

Figure 10 shows the XRD patterns of the as-dredged silt and MgO-solidified silts with and without a CO₂ injection. It can be seen that the as-dredged silt was mainly composed of quartz (SiO₂). The MgO-solidified silt mainly contained compounds of silicon, aluminum, magnesium, and calcium. Except for the original quartz in the as-dredged silt, the main phase was Mg(OH)₂. The diffraction peaks of MgO are hardly seen in the figure, indicating that the hydration reaction of MgO was basically completed. With the CO₂ injection, the diffraction peaks of Mg(OH)₂ gradually disappeared and a large number of MgCO₃·3H₂O, Mg₅(CO₃)₄(OH)₂·4H₂O, and Mg₅(CO₃)₄(OH)₂·5H₂O appeared, which are the products of the full carbonization of Mg(OH)₂. This also indicates that MgCO₃·3H₂O, Mg₅(CO₃)₄(OH)₂·4H₂O, and Mg₅(CO₃)₄(OH)₂·5H₂O were the main carbonation products and facilitated the strength growth of MgO-solidified silt with the CO₂ injection.
5. Conclusions

To explore low-carbon and environmentally friendly solidifying materials, MgO was used to solidify dredged silt from Lake Taihu, China. The effectiveness of silt solidification by MgO was evaluated using the UCS and toxicity characteristic leaching tests, and the results were compared with solidification by cement. The following conclusions were obtained:

1. The UCS of MgO-solidified silt increased with the MgO content and curing time. The UCS of the solidified silt with a MgO content of 9% and curing time of 14 d reached 111 kPa. A longer curing time resulted in a more significant effect of the MgO content, and a higher MgO content resulted in a more notable effect of curing time on the UCS.

2. The CO₂-injection treatment could further increase the UCS of the MgO-solidified silt by approximately 25%. The UCS values of the MgO-solidified silt without and with the CO₂ injection were approximately 60% and 80%, respectively, of those of the cement-solidified silt.

3. The leaching concentrations of nutrient salts (TN and TP) and heavy metal pollutants (Cu, Cr, and Ni) in the dredged silt decreased with increasing MgO content. The concentrations of TN and TP in the leached MgO-solidified silt with the CO₂ injection were only approximately 5% higher than those of the cement-solidified silt. The effect of MgO solidification with carbonization was most significant for Ni, the leaching concentration of which was reduced by more than 65%. However, the concentrations of Cu in the leached MgO-solidified silts were closest to those of the cement-solidified silts.

4. SEM and XRD results showed that the addition of MgO to the silt formed flocculated magnesium hydroxide colloidal material that filled the silt pores. As the MgO content increased, the pore size significantly reduced and the silt pores were gradually filled. Flake-like material appeared following carbonization, which was identified as hydromagnesite and dypingite. Compared with the addition of MgO alone, its combination with carbonation enhanced the overall compactness of the silt, thus greatly improving its strength.

Author Contributions: All authors contributed to the study’s conception and design. Material preparation, data collection, and analysis were performed by S.S., X.Z. and Y.G. Resources, methodology, and validation were performed by H.W., Y.T. and J.C. The first draft of the manuscript was written
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by S.S. and X.Z., and all authors commented on previous versions of the manuscript. All authors have read and agreed to the published version of the manuscript.

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**Data Availability Statement:** The datasets used and analyzed in the current study are available from the corresponding author upon reasonable request.

**Conflicts of Interest:** Author Yan Tang was employed by the company Zhejiang Huadong Geotechnical Investigation & Design Institute Co., Ltd. Author Junhao Chen was employed by the company PowerChina Huadong Engineering Co., Ltd. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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