# Supplementary Materials

# Low-carbon remediation of contaminated marine mud sediment for efficient in-situ recycling and application

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## 1. Mix design and formulation

Table S1 Oxides composition of different raw materials

| Compound Wt (%) | MM | OPC | FA | SL |
|---|---|---|---|---|
| $SiO_2$ | 58.49 | 19.89 | 46.19 | 34.38 |
| $Al_2O_3$ | 24.29 | 5.790 | 43.43 | 20.60 |
| $Fe_2O_3$ | 6.760 | 4.867 | 2.670 | 0.691 |
| $K_2O$ | 3.020 | 0.542 | 0.706 | 0.478 |
| MgO | 2.170 | 1.453 | 1.210 | 9.980 |
| CaO | 1.640 | 62.12 | 2.870 | 30.75 |
| $Na_2O$ | 1.130 | 0.440 | 0.494 | 0.612 |
| $TiO_2$ | 0.943 | 0.413 | 0.910 | 0.520 |
| Cl | 0.582 | 0.057 | - | 0.073 |
| S | 0.491 | - | - | 1.540 |
| $P_2O_5$ | 0.182 | 0.096 | 0.339 | 0.061 |
| MnO | 0.085 | 0.098 | 0.031 | 0.152 |
| $SO_3$ | - | 2.540 | 0.906 | - |

Table S2 PMD-1: Mix proportions

| Series | Mixtures label | Detail of the proportion | S/L |
|---|---|---|---|
| D1 | CE5-15-80 | 5% OPC + 15% Granular filler + 80% Marine mud | 0.15 |
| | FA5-15-80 | 5% Fly ash + 15% Granular filler + 80% Marine mud | 0.15 |
| | SL5-15-80 | 5% Slag + 15% Granular filler + 80% Marine mud | 0.15 |
| D2 | FA5-CE0.5-94.5 | 5% Fly ash + 0.5 OPC + 94.5 % Marine mud | 0.18 |
| | SL5-CE0.5-94.5 | 5% Slag + 0.5 OPC + 94.5 % Marine mud | 0.18 |
| D3 | FA5-95-NaOH | 5% Fly ash + 95% Marine mud + NaOH solution (8M) | 0.20 |
| | SL5-95-NaOH | 5% Slag + 95% Marine mud + NaOH solution (8M) | 0.20 |
| D4 | MM-20-80-W | 20% Granular filler + 80% Marine mud + Water | 0.15 |
| | MM-20-80-NaOH | 20% Granular filler + 80% Marine mud + NaOH solution (8M) | 0.15 |

Table S3 Degree of compressibility of PMD-1 mixture

| Series | Mixture label | Curing duration (Days) | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | 1 | 2 | 3 | 4 | 5 | 6 | 7 |
| D1 | CE5-15-80 | M | M | M | M | M | H | H |
| | FA5-15-80 | L | L | L | L | L | L | M |
| | SL5-15-80 | L | L | L | L | L | L | M |
| D2 | FA5-CE0.5-94.5 | L | L | L | L | L | L | M |
| | SL5-CE0.5-94.5 | L | M | M | M | H | H | H |
| D3 | FA5-95-NaOH | L | L | L | M | M | M | M |
| | SL5-95-NaOH | M | M | H | H | H | H | H |
| D4 | MM-20-80-W | L | L | L | L | L | L | M |
| | MM-20-80-NaOH | L | L | M | M | M | M | M |

**L: Low, M: Medium, H: High*

Table S4 Contaminant regulatory level according to Chinese standard GB and US EPA

| Contaminant | Regulatory level Chinese standard GB36600-2018 (mg/kg) | Regulatory level U.S. EPA 540/2-86/001 (mg/kg) |
|---|---|---|
| Arsenic ($^{75}$As) | 120 | 5 |
| Barium ($^{137}$Ba) | DF. | 100 |
| Cadmium ($^{111}$Cd) | 47 | 1 |
| Chromium ($^{52}$Cr) | 30 | 5 |
| Lead ($^{208}$Pb) | 800 | 5 |

**DF. refers to deficiency*

Table S6 PMD-2: Mix proportions

| Series | Mixture label | Detail of the proportion | (S/L) |
|---|---|---|---|
| D1 | CE15-10-75 | 15% OPC + 10% Granular filler + 75% Marine mud | 0.15 |
| | FA15-10-75 | 15% Fly ash + 10% Granular filler + 75% Marine mud | 0.15 |
| | SL15-10-75 | 15% Slag + 10% Granular filler + 75% Marine mud | 0.15 |
| D2 | FA10-90-NaOH-8 | 10% Fly ash + 90% Marine mud + NaOH solution (8M) | 0.20 |
| | SL10-90-NaOH-8 | 10% Slag + 90% Marine mud + NaOH solution (8M) | 0.20 |
| | FA10-90-NaOH-6 | 10% Fly ash + 90% Marine mud + NaOH solution (6M) | 0.20 |
| | SL10-90-NaOH-6 | 10% Slag + 90% Marine mud + NaOH solution (6M) | 0.20 |
| D3 | MM15-85-NaOH-6 | 15% Granular filler + 85% Marine mud + NaOH solution (6M) | 0.15 |

Table S7 Degree of compressibility of PMD-2 mixture

| Series | Mixture label | Curing duration (Days) | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | 1 | 2 | 3 | 4 | 5 | 6 | 7 |
| D1 | CE15-10-75 | H | H | H | H | H | H | H |
| | FA15-10-75 | L | L | L | L | L | L | M |
| | SL15-10-75 | L | L | L | L | L | L | M |
| D2 | FA10-90-NaOH-8 | M | M | M | M | M | M | H |
| | SL10-90-NaOH-8 | H | H | H | H | H | H | H |
| | FA10-90-NaOH-6 | M | M | M | M | M | M | H |
| | SL10-90-NaOH-6 | H | H | H | H | H | H | H |
| D3 | MM15-85-NaOH-6 | L | L | M | M | M | M | M |

**L: Low, M: Medium, H: High*

Table S8 Heavy metals contamination assessment of PMD-2

| Series | Mixture label | Heavy metals (mg/kg) | | | | |
|---|---|---|---|---|---|---|
| | | $^{75}$As | $^{137}$Ba | $^{111}$Cd | $^{52}$Cr | $^{208}$Pb |
| D0 | MM | 25.678 | 150.872 | 0.441 | 85.005 | 50.419 |
| D1 | CE15-10-75 | 0.053 | 0.044 | ND. | 1.556 | ND. |
| | FA15-10-75 | 0.301 | 0.248 | 0.003 | 0.077 | ND. |
| | SL15-10-75 | 0.051 | 0.070 | 0.002 | 0.007 | ND. |
| D2 | FA10-90-NaOH-8 | 17.017 | 0.050 | 0.002 | 0.303 | ND. |
| | SL10-90-NaOH-8 | 11.224 | 0.008 | ND. | 1.541 | ND. |
| | FA10-90-NaOH-6 | 8.123 | 0.031 | 0.001 | 0.227 | ND. |
| | SL10-90-NaOH-6 | 5.817 | 0.009 | ND. | 0.356 | ND. |
| D3 | MM15-85-NaOH-6 | 8.739 | 0.262 | 0.004 | 0.573 | 0.003 |

**ND. refers to non-detectable*

## 2. Raw marine mud physical properties

The physical properties of the collected marine mud as provided by the engineering firm were performed by ASTM testing standards and presented respectively in the Tables below:

Table S9 Specific gravity of soil solids

| Testing standard | ASTM D854-10 | |
|---|---|---|
| Sample Code | Sample ID | Soil gravity (G20°C) |
| MM50431 | ABH06 13.0~14.0 | 2.72 |
| MM50437 | ABH07 13.0~14.0 | 2.71 |
| MM50443 | ABH08 16.5~17.5 | 2.74 |
| MM50444 | ABH08 23.0~23.5 | 2.68 |
| MM50450 | ABH10 14.5~15.5 | 2.76 |

Table S10 Determination of water content

| Testing standard | ASTMD2216-19 | |
|---|---|---|
| Sample Code | Sample ID | Water content (%) |
| MM50413 | ABH02 11.0~12.0 | 54.0 |
| MM50415 | ABH02 19.0~20.0 | 21.2 |
| MM50419 | ABH04 9.0~10.0 | 58.6 |
| MM50426 | ABH05 9.0~10.0 | 66.5 |
| MM50428 | ABH05 17.0~18.0 | 20.5 |
| MM50432 | ABH06 16.5~17.5 | 50.2 |
| MM50440 | ABH07 27.0~27.5 | 34.4 |
| MM50443 | ABH08 16.5~17.5 | 51.0 |
| MM50449 | ABH10 12.5~13.5 | 57.9 |
| MM50451 | ABH10 20.5~21.0 | 17.1 |
| MM50456 | ABH11 13.0~14.0 | 45.9 |

Table S11 Determination of Atterberg limits

| Testing standard | ASTM D4318-17e1 | | | |
|---|---|---|---|---|
| Sample Code | Sample ID | Liquidity limit (%) | Plasticity limit (%) | Plasticity Index (%) |
| MM50131 | ABH01 18.5~19.5 | 42 | 20 | 22 |
| MM50137 | ABH03 11.0~12.0 | 77 | 40 | 37 |
| MM50139 | ABH03 19.0~20.0 | 47 | 25 | 22 |
| MM50146 | ABH09 16.5~17.5 | 73 | 40 | 33 |
| MM50154 | ABH12 15.0~16.0 | 78 | 43 | 35 |
| MM50155 | ABH12 19.0~20.0 | 44 | 25 | 19 |

Table S12 Particle size analysis

| Testing standard | | ASTM D422-63 | | |
|---|---|---|---|---|
| Sieve No. | Retained sieve mass (g) | Percentage of sieve retention (%) | Percentage of cumulative sieve retention (%) | Total sieve ratio (%) |
| 50.0mm | 0.0 | 0.0 | 0.0 | 100.0 |
| 37.5mm | 0.0 | 0.0 | 0.0 | 100.0 |
| 25.0mm | 0.0 | 0.0 | 0.0 | 100.0 |
| 12.5mm | 0.0 | 0.0 | 0.0 | 100.0 |
| 9.50mm | 0.0 | 0.0 | 0.0 | 100.0 |
| 4.75mm | 1.06 | 1.0 | 1.0 | 99.0 |
| 2.00mm | 11.38 | 10.4 | 11.4 | 88.6 |
| #20 | 14.99 | 15.5 | 26.9 | 74.9 |
| #40 | 8.48 | 8.8 | 35.7 | 67.1 |
| #60 | 4.61 | 4.8 | 40.4 | 62.9 |
| #140 | 8.56 | 8.9 | 49.3 | 55.0 |
| #200 | 4.94 | 5.1 | 54.4 | 50.5 |
| <#200 | 55.10 | | | |
| Total mass | | 109.12g | | |
| Test Methods | | Gravimetric method | | |
| D (mm) | | P (%) | | Total sieve ratio (%) |
| 0.0396 | | 47.7 | | 42.2 |
| 0.0287 | | 44.7 | | 39.6 |
| 0.0189 | | 40.0 | | 35.5 |
| 0.0114 | | 34.1 | | 30.2 |
| 0.0085 | | 27.7 | | 24.5 |
| 0.0062 | | 22.7 | | 20. |
| 0.0032 | | 16.6 | | 14.7 |
| 0.0013 | | 10.8 | | 9.6 |
| 0.0009 | | 9.5 | | 8.4 |

3. **Solidified marine mud morphological analysis**

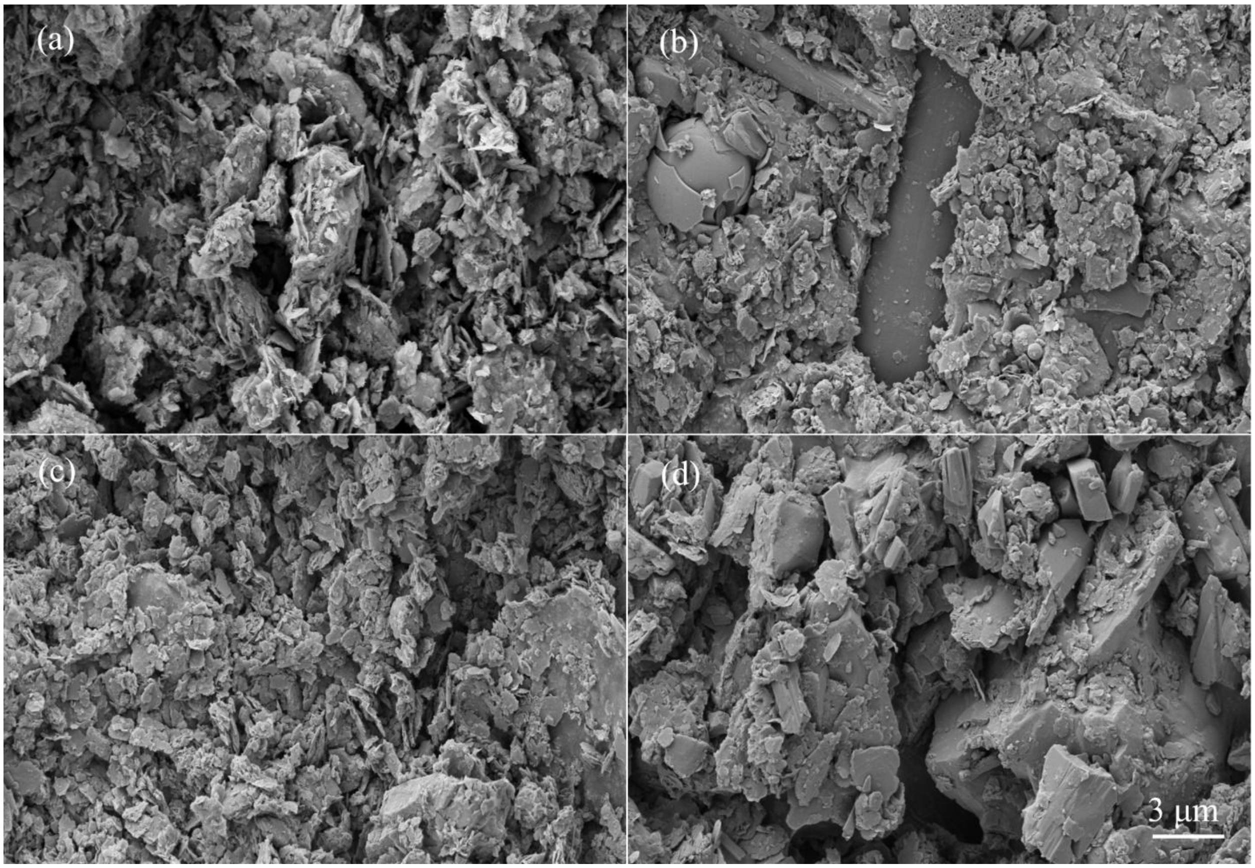


Fig. S1 Microstructure characterization of the solidified-marine mud: (a) SEM of marine mud treated with OPC, (b) SEM of marine mud treated with Fly ash, (c) SEM of marine mud treated with slag, (d) SEM of marine mud solidified with 5% sand and 95% marine mud

**TEM oxides distribution analysis:** In CE25-75, the absence of Na confirms no alkaline activation, while the matrix contains 6.04% $Al_2O_3$, 14.00% $SiO_2$, and 77.86% CaO, consistent with OPC incorporation; 2.10% $Fe_2O_3$ likely comes from original MM. Conversely, FA25-75 shows 44.92% Na, possibly from sodium-bearing minerals; and 19.67% $Al_2O_3$, 2.45% CaO, and 3.48% $Fe_2O_3$, reflecting the FA silicate-rich and low-calcium profile. In SL25-75-NaOH-6, 23.40% Na confirms activation, while 19.99% $Al_2O_3$ and 38.59% $SiO_2$ support geopolymer formation. The high CaO (30.75%) from slag boosts strength, supported by UCS tests, and 3.69% $Fe_2O_3$ aids stability. Lastly, MM5-95-NaOH-6 contains 24.76% Na, 28.60% $Al_2O_3$, 34.75% $SiO_2$, 6.32% CaO, and 5.57%

$Fe_2O_3$. These compositions indicate effective activation and matrix formation, with Al and Si driving geopolymerization, Ca contributing to strength, and Fe improving structural integrity.

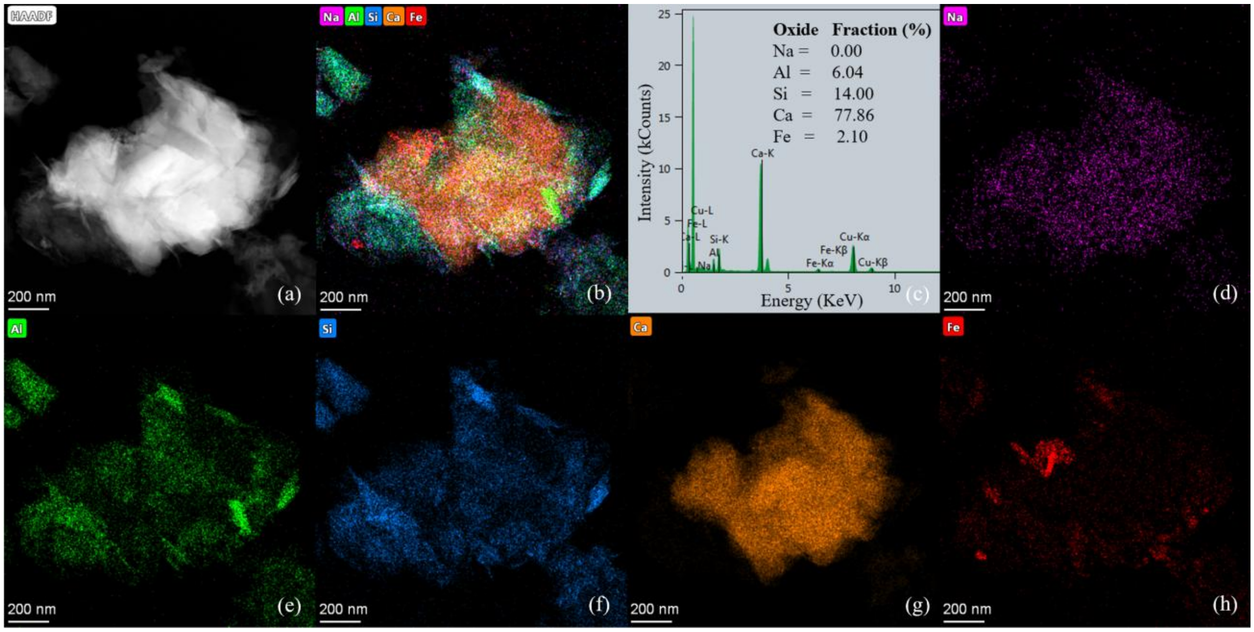


Fig. S2 TEM elemental distribution mapping of the marine mud treated with OPC, structure containing: (a) High-angle annular dark-field (HAADF) imaging of the sample, (b) Overall distribution of the predominant oxides, (c) Fraction of the distributed chemical oxides, (d) 0.00% Na oxide fraction, (e) 6.04% Al oxide fraction, (f) 14.00% Si oxide fraction, (g) 77.86% Ca oxide fraction, (h) 2.10% Fe oxide fraction

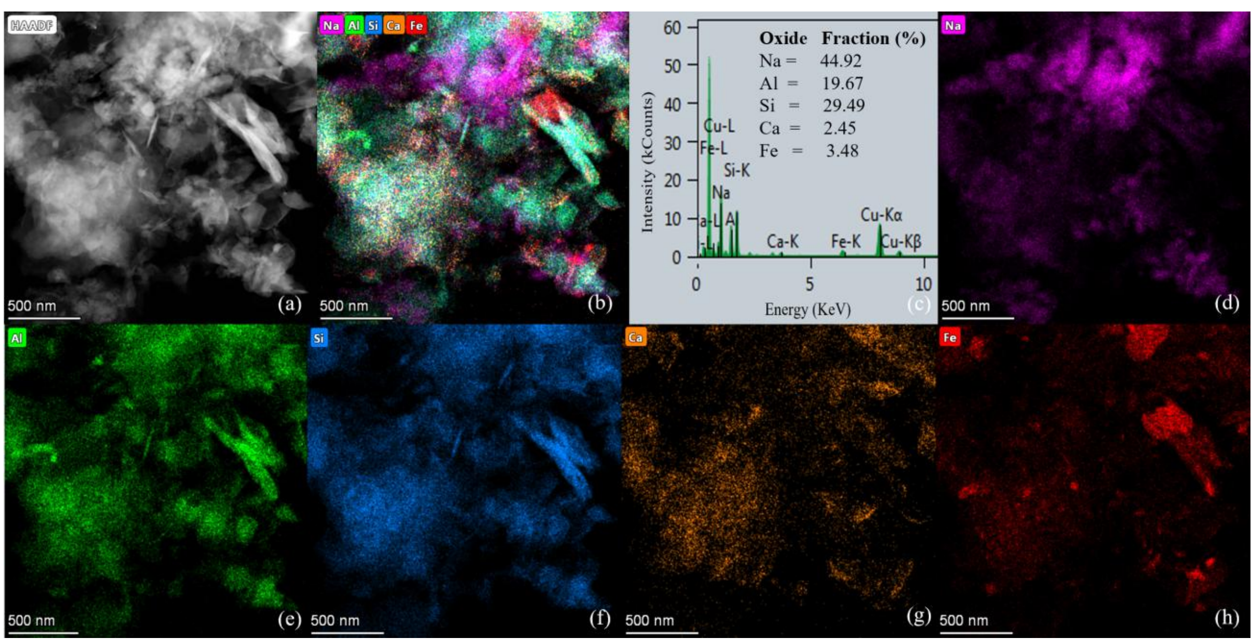


Fig. S3 TEM elemental distribution mapping of the marine mud treated with fly ash (FA), structure containing: (a) High-angle annular dark-field (HAADF) imaging of the sample, (b) Overall distribution of the predominant oxides,

(c) Fraction of the distributed chemical oxides, (d) 44.92% Na oxide fraction, (e) 19.67% Al oxide fraction, (f) 29.49% Si oxide fraction, (g) 2.45% Ca oxide fraction, (h) 3.48% Fe oxide fraction.

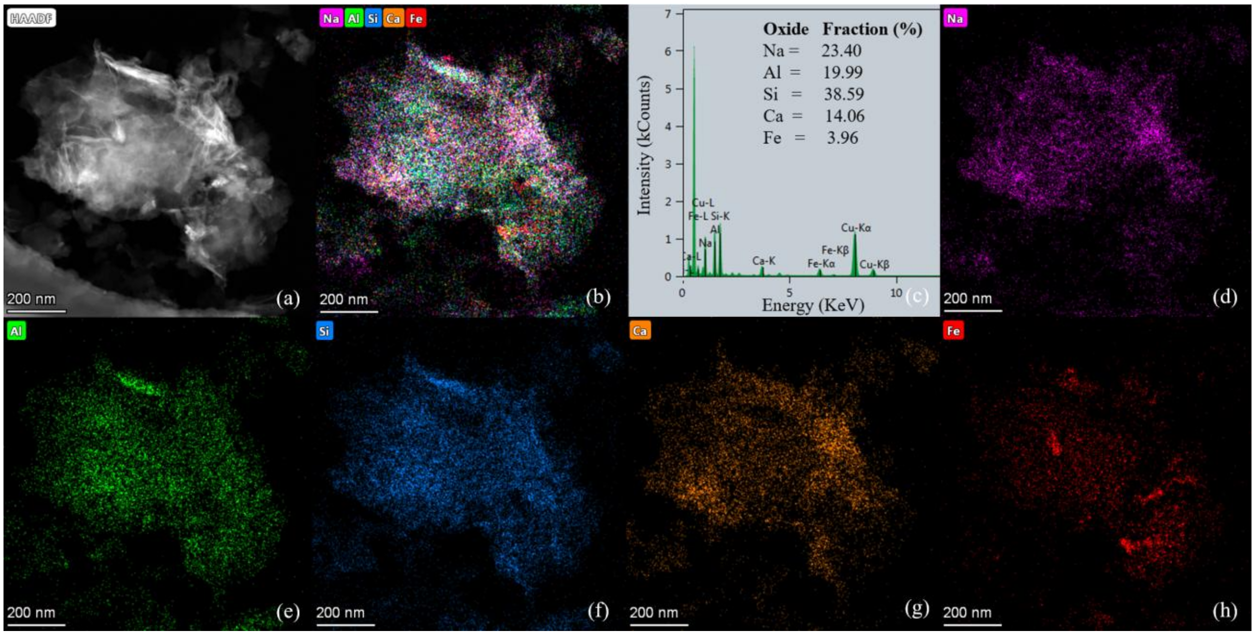


Fig. S4 TEM elemental distribution mapping of the marine mud solidified with 25% slag and 75% marine mud activated with 6M NaOH solution, structure containing: (a) High-angle annular dark-field (HAADF) imaging of the sample, (b) Overall distribution of the predominant oxides, (c) Fraction of the distributed chemical oxides, (d) 23.40% Na oxide fraction, (e) 19.99% Al oxide fraction, (f) 38.59% Si oxide fraction, (g) 14.06% Ca oxide fraction, (h) 3.96% Fe oxide fraction.

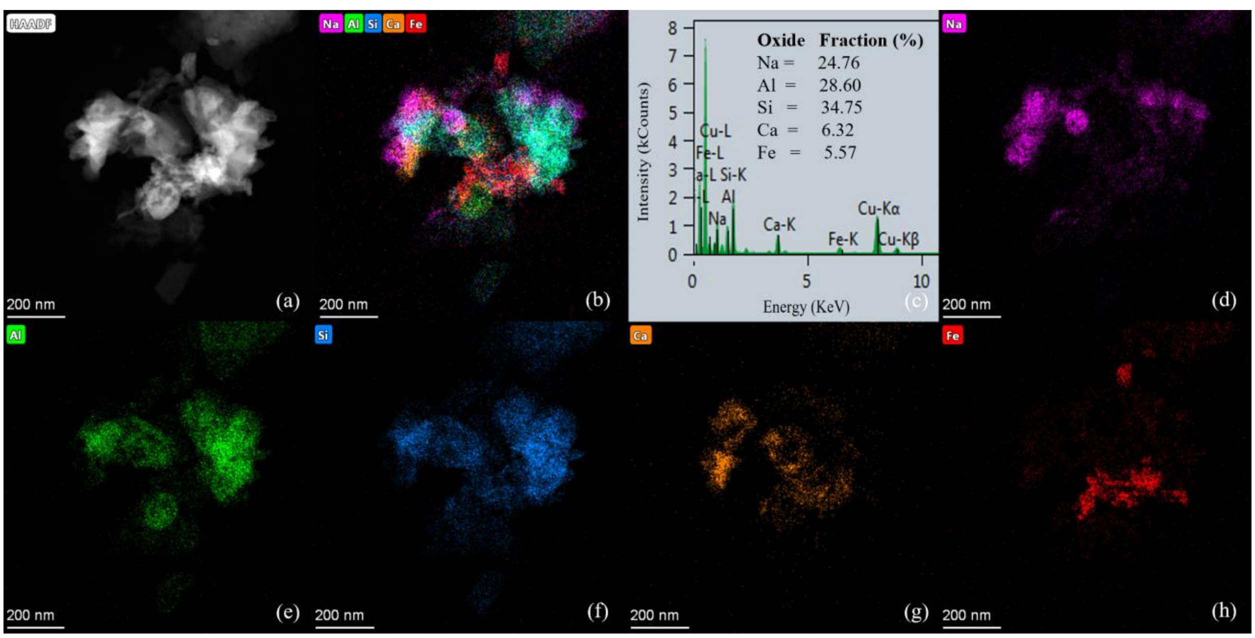


Fig. S5 TEM elemental distribution mapping of the marine mud solidified with 5% river sand and 95% marine mud activated with 6M NaOH solution, structure containing: (a) High-angle annular dark-field (HAADF) imaging of the sample, (b) Overall distribution of the predominant oxides, (c) Fraction of the distributed chemical oxides, (d) 24.76%

Na oxide fraction, (e) 28.60% Al oxide fraction, (f) 34.75% Si oxide fraction, (g) 6.32% Ca oxide fraction, (h) 5.57% Fe oxide fraction.

## 4. Pilot-scale experiments design

### *4.1 Test objectives*

1. Verification under natural conditions to evaluate whether the solidification formulas and construction techniques developed in the laboratory could achieve the required strength, permeability, and environmental safety standards;
2. Process parameter scaling to determine adjustment factors for binder dosage, mixing energy consumption, and curing time under scale-up conditions, thereby providing a reliable basis for subsequent engineering design;
3. Environmental impact assessment by monitoring turbidity, heavy metal migration, noise, and other key indicators, enabling a comparative evaluation of ecological disturbances caused by different treatment pathways.

### *4.2 Targeted performance indicators*

(1) 28-day Unconfined Compressive Strength (UCS) of not less than 800 kPa.

(2) Equivalent foundation bearing capacity of not less than 150 kPa.

(3) Annual differential settlement within 30 mm.

(4) External chloride ion and $NH_4^+$ concentrations meeting marine sediment disposal standards.

### *4.3 Process flow and implementation*

The implementation followed a structured sequence. It began with site investigation and drilling to determine the thickness of the soft mud layer and delineate treatment sub-areas. This was followed by the installation of open ditches and drainage wells, the placement of a 0.8 m sand cushion, and the application of 30 kPa short-term preloading with continuous drainage for 7 days. In the solidification stage, the blank control zone was treated using a deep-mixing machine with double-pass bottom-up mixing at a rotation speed of not less than 35 rpm, using slurry with a water-to-cement ratio of 0.8. In the formulation test zones, a long auger drill with eccentric grouting was employed, followed by two-pass cross mixing to incorporate the designed binder formulations into the slurry. After 3 days of mixing, preloading was removed, and samples could undergo 28 days of natural curing under real-time monitoring of temperature and humidity. At the end of curing, plate load tests, shear wave velocity measurements, and permeability coefficient tests were conducted, followed by recompression tests at 56 days. Finally, a 0.3 m gravel layer was placed on the surface and compacted to 95% density, creating conditions for subsequent load-bearing applications.

### *4.4 Key technical parameters and control points*

To ensure experimental reliability, several parameters were strictly controlled. The binder dosage deviation was maintained within ±3%, and slurry density was stabilized at 1.50 ± 0.05 t/m³. Pile verticality deviation was kept below 1/100, while real-time monitoring ensured that mixing torque remained above 35 kN·m. Furthermore, short-term settlement rates were limited to ≤10 mm/day, with continuous monitoring every 6 h using automatic leveling equipment.

### *4.5 Monitoring and testing program*

A comprehensive monitoring program accompanied the process. Water quality monitoring included measurements of turbidity, suspended solids, pH, dissolved oxygen, chloride ions, and $NH_4^+$ concentrations every 10 minutes during construction, with follow-up checks within 24 h of completion. The backfilling foundation deformation was recorded automatically using settlement plates and horizontal displacement gauges. The mechanical performance was assessed through UCS tests at 7, 28, and 56 days, as well as plate load and CBR tests. Finally, permeability and leaching performance were evaluated using laboratory permeability coefficient tests combined with seawater immersion analysis for heavy metal release.

### *4.6 Recommended values*

| **Designation** | **Recommended characteristics** |
|---|---|
| Test area | Within the planned area, a 30 m × 20 m section of marine mud was selected, with representative conditions and convenient for construction |
| Treatment volume | 100 m³ of wet sediment (with moisture content meeting the required standard) |
| Partition layout | 4 independent test pits/steel sheet pile cofferdams (5 m × 5 m × 1.5 m deep) used respectively for three formulations plus one blank control. |