

EVALUATION OF DIFFERENT BINDER COMBINATIONS OF CEMENT, SLAG AND CKD FOR S/S TREATMENT OF TBT CONTAMINATED SEDIMENTS

Per LINDH^{***} , Polina LEMENKOVA^{***} 

^{*}Swedish Transport Administration, Gibraltargatan 7, Malmö, Sweden

^{**}Lund University, Division of Building Materials, Box 118, SE-221-00, Lund, Sweden

^{***}Université Libre de Bruxelles. École polytechnique de Bruxelles (Brussels Faculty of Engineering), Laboratory of Image Synthesis and Analysis, Bld. L, Campus de Solbosch, Avenue Franklin Roosevelt 50, Brussels 1000, Belgium

per.a.lindh@gmail.com, polina.lemenkova@ulb.be

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Abstract: The seabed in the ports needs to be regularly cleaned from the marine sediments for safe navigation. Sediments contaminated by tributyltin (TBT) are environmentally harmful and require treatment before recycling. Treatment methods include leaching, stabilisation and solidification to remove toxic chemicals from the sediments and improve their strength for reuse in the construction works. This study evaluated the effects of adding three different binder components (cement, cement kiln dust (CKD) and slag) to treat sediment samples collected in the port of Gothenburg. The goal of this study is to assess the leaching of TBT from the dredged marine sediments contaminated by TBT. The various methods employed for the treatment of sediments include the application of varied ratios of binders. The project has been performed by the Swedish Geotechnical Institute (SGI) on behalf of the Cementa (HeidelbergCement Group) and Cowi Consulting Group, within the framework of the Arendal project. An experiment has been designed to evaluate the effects of adding CKD while reducing cement and slag for sediment treatment. Methods that have been adopted include laboratory processing of samples for leaching using different binder combinations, followed by statistical data processing and graphical plotting. The results of the experiment on leaching of TBT for all samples are tested with a varied ratio of cement, slag, CKD and water. Specimens with added binders 'cement/CKD' have demonstrated higher leaching compared to the ratio 'cement/slag/CKD' and 'cement/slag'. The 'CKD/slag' ratio has presented the best results followed by the 'cement/slag/CKD', and can be used as an effective method of s/s treatment of the sediments. The results have shown that the replacement of cement and slag by CKD is effective at TBT leaching for the treatment of toxic marine sediments contaminated by TBT.

Keywords: cement kiln dust, CKD, marine sediments, tributyltin, TBT, leaching, s/s soils

1. INTRODUCTION

The Cement Kiln Dust (CKD) is a fine-grained, solid, highly alkaline material and a by-product of cement (Barnat-Hunek et al., 2018). As an industrial solid waste material, it can be used for replacing cement as a stabiliser and a binary binder (Mansour, 2021). Adding CKD is an effective solution to stabilise the ground for the construction of infrastructure, buildings and roads (Rimal et al., 2019). Besides, adding CKD increases compressive strength (Ribeiro and Morelli, 2009; Shoaie et al., 2017; Shen et al., 2021), resistivity and durability of soils (Abdel-Gawwad et al., 2019; Yaseri et al., 2019). The benefits of CKD as an additive binder in cement consist in increased tensile strength and improved characteristics of cement (Baghrichie et al., 2020; Silva et al., 2015). The CKD can be added in various ratios, to improve the properties of soils. Such experiments show a significant increase in compressive strength, for instance, obtained for a CKD–slag mixture with 70% of CKD and 30% of slag at a water/binder ratio of 0.40 (Chaunsali and Peethamparan, 2011). Further examples of the experimental CKD applications are presented in the existing papers (Sariosseiri & Muhunthan, 2008; Yoon et al., 2010; Ahmad et al., 2014; Adeyanju et al., 2020).

Massive amounts of cement produced annually lead to the increase of CKD as a by-product (Najim et al., 2014). Therefore, utilising CKD is quite economical and environmentally friendly contributing to sustainable development. For instance, rational economic reasons for the application of CKD include the reduced costs of the construction works (Al-Homidy et al., 2017), and optimised economic solutions regarding the workflow (Faisal et al., 2021). The environmental benefits of replacing cement by the CKD consist in reduced air pollution due to the decreased CO_2 emissions (Bagheri et al., 2020). Besides, CKD is efficient in wastewater treatment, acting as a coagulant in removing heavy metals (Hasaballah et al., 2021).

Marine sediments need to be regularly removed for safe navigation (Baltic Marine Environment Protection Commission, 2015; Akcil et al., 2015). However, dredged masses of the marine sediments should be treated before the reuse. The treatment includes stabilisation and removal of contaminants. Marine sediments are often contaminated by heavy metals, toxic chemicals and tributyltin (TBT), which presents an environmental problem in coastal regions (Sundqvist et al., 2009; Kim et al., 2011).

Large amounts of dredged sediments polluted by chemicals are harmful to marine ecosystems and human health (Evans, 1999; Antizar-Ladislao, 2008). The TBT belongs to the persistent

pollutants especially harming the marine environment. Originated as paints on ships protecting hulls against fouling during constructions (Wojtkiewicz et al., 2015; Alzieu, 2000), the TBT has been prohibited since 1989 (Blanck & Dahl, 1998). However, certain amounts of TBT remain in the waters of the Baltic Sea with negative consequences on the environment (Abraham et al., 2017; Eklund & Watermann, 2018). Treatment of the marine sediments contaminated by TBT required the development of special methods (Berto et al., 2007; Furdek Turk et al., 2020; Sheikh et al., 2020; Bandara et al., 2021).

The techniques of leaching were applied in this work for the treatment of marine sediments using guidance standards provided by the Swedish Institute for Standards (SIS), <https://www.sis.se/en/> Leaching of metals and chemical pollutants from soils and sediments is widely used as an effective method of environmental treatment of soils under EN 12457-4 and NEN 7275 standards (Lu et al., 2016; Kuterasińska-Warwas & Król, A. 2017) or EN 12457-2 for leaching (Pazikowska-Sapota et al., 2016; Mizerna & Król, 2018).

Besides geochemical treatment, marine sediments should be cleaned and stabilised before their reuse in the construction works, to improve their geotechnical and environmental properties (Herbich, 1990; Li et al., 2019; Houlihan et al., 2021). Stabilisation and binding of the marine sediments, as well as other weak soils, such as clay, loam or silt, can be done by adding CKD using mechanical methods of mixing and geochemical treatment (Wareham and Mackechnie, 2006; Ghavami & Rajabi, 2021).

Various approaches of soil treatment are being continuously reported in the previous works, which reflects the need for improved methods (Moh, 1962; Bandyopadhyay, 1981; Turner 1994; Shoab et al., 2000; Fabian et al., 2010; Schifano and Fabian, 2010; Fiertak and Stryzewska, 2013; Lindh et al., 2018; Li et al., 2019; Wang et al., 2020). The impact of external effects on soils can be assessed using techniques of data analysis (Dahlin et al., 1999; Nosjean et al., 2020; Lemenkov and Lemenkova, 2021b; Zahran, 2020). Geochemical methods include hydrogeological appraisals, drilling (Shah et al., 2021), image analysis (Källén et al., 2014, 2016). Methods of soil treatment and evaluation of its performance under varying external effects include stabilisation and solidification (s/s) aimed to improve the mechanical performance of soil and increase its strength before recycling (Wang et al., 2011; Tang et al., 2020; Zhang et al., 2018; 2020, 2021; Fan et al., 2021).

Treatment of the marine sediments contaminated by TBT using s/s method presents an environmentally effective method for recycling of soils. The s/s treatment can use various agents, e.g. kaolinite, quicklime, cement (Radenović et al., 2019). In this way, the s/s technique enables to get environmentally compatible soils with removed heavy metals or organic matter and improved properties (De Gisi et al., 2020). Various methods and techniques of soil treatment are reported in previous studies (Lindh, 2004; Li et al., 2017; Lemenkov and Lemenkova, 2021a).

The purpose of this research is to explore different combinations of binders – CKD, cement and slag – for s/s treatment of the marine sediments contaminated by TBT. The s/s treatment aims to remove the TBT contaminants. The objective of this study is to assess the properties of sediments after treatment by various ratios of binders and to evaluate the applicability of CKD. The specific research question is to assess if the replacement of cement or slag by CKD is optimal for s/s treatment of the marine sediments. The study experiment has been performed in south-western Sweden with sediment samples collected in the Port of Gothenburg (Fig. 1).

2. METHODOLOGY

The project has been performed in the Swedish Geotechnical Institute (SGI) on behalf of the Cementa (HeidelbergCement Group) and Cowi Consulting Group in the framework of the Arenal project. The samples have been dredged and collected from the seabed of the Port of Gothenburg, Sweden (Fig. 1). The

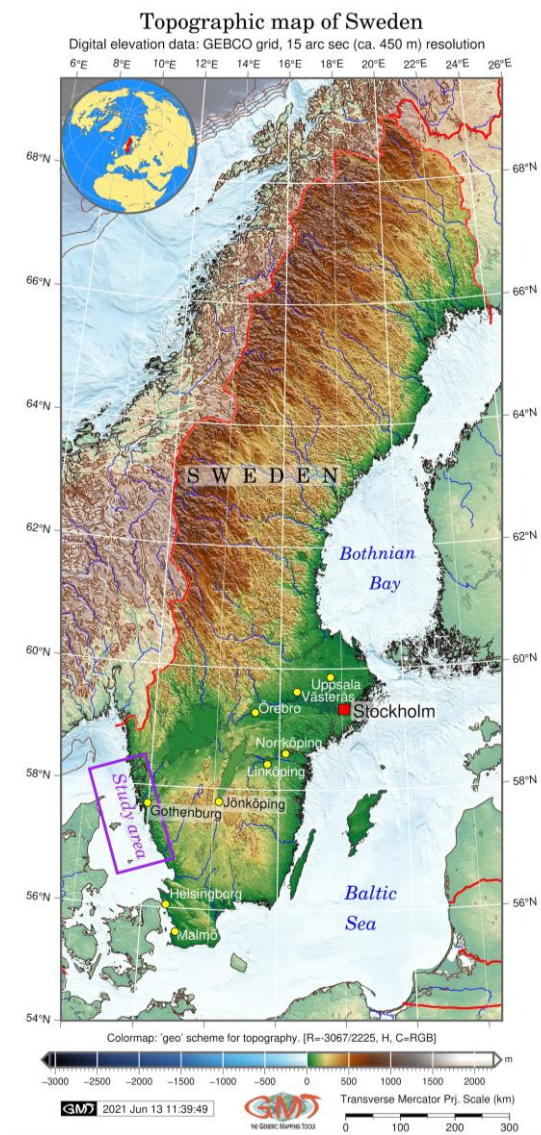


Fig. 1. Location of the study area. Data source: General Bathymetric Chart of the Oceans (GEBCO). RGB colour model. RGB, red, green blue. Mapping: Generic Mapping Tools (GMT). Map source: authors

Leaching as one of such methods has been used and presented in previous works (e.g. Norén et al., 2021; Alshemmari et al., 2020). Leaching is a process of extraction of solute from the carrier substance by a solvent (Richardson et al., 2002). Natural leaching often occurs in cement materials due to natural weathering. In experimental works, leaching can be applied to remove contaminants from sediments. Thus, chemical leaching is a common method for extracting solvents from soils (Shen et al., 2020).

methodology includes the s/s treatment of the sediment samples, statistical data processing and graphical plotting.

The laboratory tests on the stabilisation of the marine sediments were performed in SGI following the standard procedure. Treatment of sediments was based on the guidance of the SIS, ISO 18772:2008 (Swedish Institute for Standards, 2021a). During the s/s treatment of the marine sediments, the TBT constituents were leached from the sediments samples in changed testing conditions: varied ratio of binder and water. The methodology follows the existing procedure of leaching to examine the quality of the marine sediments to stabilise specimens for further recycling.

Leaching tests in sediment samples were performed according to the SIS guidance on leaching for chemical and ecotoxicological tests of soils with high content of contaminants, ISO 18772:2008. Leaching tests were aimed to clean the sediments from the high concentrations of TBT in the collected specimens. The test specimens were demolded before the start of the experiment, which took periods of 2.25 days, 9 days and 36 days. Leaching aimed to improve the physical and chemical properties of soil specimens (low strength, stability and stiffness) and decrease high moisture to ensure the recycling of soils in geotechnical works.

The compressive strength has been measured following the standards of SIS, Geotechnical investigation and testing – Laboratory testing of soil – Part 7: Unconfined compression test, ISO 17892-7:2017 (Swedish Institute for Standards, 2017a). It has been tested as a maximum load that could be applied to evaluate the performance of the s/s sediments under varied tested conditions indicating the durability and resistance of sediment samples. The tests on the compressive strength were performed to evaluate what binder ratio presents the most effective performance for the contaminated marine sediments. The compressive tests aimed to measure the capacity of soils and sediments to withstand loads.

2.1. Hypothesis

The overall hypothesis consists of three assumptions.

1. First, it is possible to replace cement or slag by CKD with the maintained performance of TBT leaching for tested specimens of the marine sediments collected from the Port of Gothenborg.
2. Second, the CKD as a replacement of cement for stabilising marine sediments is an effective means for improving their compressive strength.
3. Third, the diffusion gradient becomes stronger when the amount of water increases concerning surface, which drives the leaching of easily soluble substances.

2.2. Sampling of Dredged Sediments

The marine sediments have been dredged from the seabed of the Port of Gothenborg, Kattegat Strait, southern Sweden.

Sampling took place within the framework of the pilot project Arendal 2 in the SGI. Preparation of specimen sediments has been performed following a standardised workflow. The specimens were collected using sampling techniques (Lindh, 2001, 2003) to obtain representative data which adequately reflects the

characteristics of sediments. Specimens of dredged sediments were collected and stored in plastic barrels (Fig. 2). Afterwards the specimens were intensively mixed with binders for 5 min. Blending samples with mortar was done using a mixing device (Fig. 3).



Fig. 2. The barrel used in a project. The size of the barrel was determined to achieve the optimal stirring. The homogeneous sediment masses were obtained for further testing. Photo by Per Lindh



Fig. 3. Mixing tool for the homogenisation of the dredged material. Photo by Per Lindh

2.3. Preparation of Specimens

After mixing and homogenisation, the specimen samples were filled into the cylinder sleeves where they reached a consistency that enabled to pour them. The standard piston sampling cylinder had an inner diameter of 50 mm and a height of 170 mm. The specimens were stored submerged with storage $T = 20\text{ }^{\circ}\text{C}$. Afterwards, the specimens were treated according to the existing standards of SIS:

1. Water ratio has been determined according to the standard of the Geotechnical investigation and testing – Laboratory testing of soil – Part 1: Determination of water content (ISO 17892-1:2014) SS-EN ISO 17892-1 2 (Swedish Institute for Standards, 2014a).
2. Dry matter and density have been determined following the standard of the Geotechnical investigation and testing – Laboratory testing of soil – Part 2: Determination of bulk density (ISO 17892-2:2014) SS-EN ISO 17892-2 3 (Swedish Institute for Standards, 2014b).
3. Grain distribution of sediments was tested using the standard test method for Particle-Size Distribution (Gradation) of Fine-

Grained Soils Using the Sedimentation (Hydrometer) Analysis, STD-80029734 (Swedish Institute for Standards, 2021b).

- The determination of TBT from the leachate followed the standard organotin compounds: ISO 17353:2004 (Swedish Institute for Standards, 2005 (see Subsection 2.7)).

The specimens were mixed with different water content proportions to obtain two variants of mixtures prepared from the marine sediments (Cell 1 and Cell 2). A certain amount of water has been removed and used partially for changing water ratio in sediments and partially for using in leaching tests. Fig. 4 shows the water ratio in the two different mixtures: 168% and 219%, respectively.

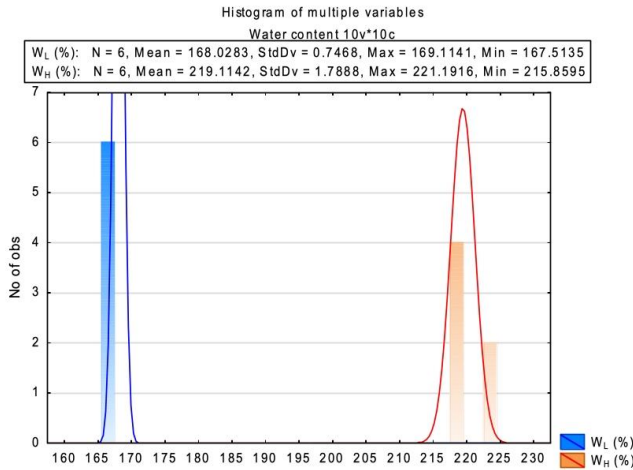


Fig. 4. Histogram showing the two variants of water ratio determined according to the SS-EN ISO 17892-1

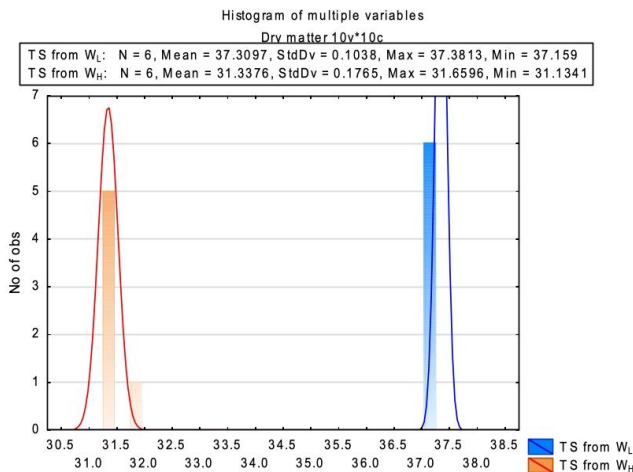


Fig. 5. Histogram showing the proportion of dry matter in the two mixtures of sediment samples

The soil permeability was tested to evaluate the capacity of porous sediments to allow water to pass through, as measured during experiments. The concept of permeability is crucial in hydrogeology as a fracture permeability of soils (Guerriero et al., 2013). The bulk density and dry density of the two mixtures with dredged material are shown as follows. The W_L (“water low” mixture) has a density of 1,316 tons/m³, and a dry density of 0,491 t/m³, while the W_H (“water high” mixture) had a density of 1,254 tons/m³ and a dry density of 0.393 t/m³. The impact of various ratios of two binders and two water-to-binder ratios (w/b) are

investigated. Such proportions were chosen in this study, but the results can also be extrapolated to other ratios in different test conditions. The standard deviation for a higher water ratio is higher (Fig. 4), as it is more difficult to maintain the same homogeneity for the higher water ratio. The actual dry substances were 31.3% and 37.3%, respectively (SS-EN 12880), Fig. 5.

The grain distribution in the dredged marine sediments was determined using the SIS standard for Geotechnical investigation and testing – Identification, description and classification of the rock, ISO 14689:2017 (Swedish Institute for Standards, 2017b). The grain distribution analysis has been done during the initial laboratory test and during the pilot experiment, Fig. 6.

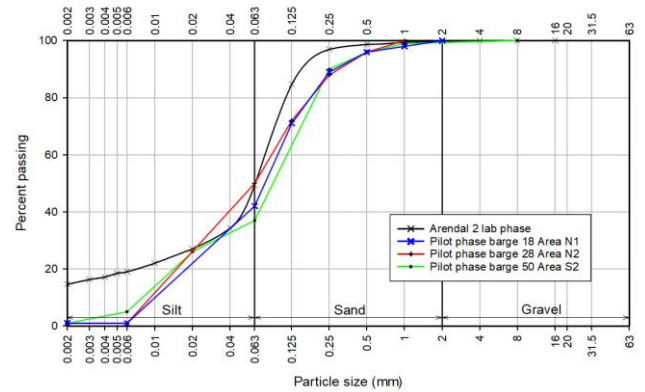


Fig. 6. Graph showing grain distribution in the soil material, classified as silty muddy sand, silty sand and sandy silt

The grain distribution was based on the classification of sediments according to the particle sizes. The sediments have been identified during the pilot laboratory phase of the experiment as the three-grain types: (1) silty muddy sand; (2) silty sand; (3) sandy silt. The material comes from the areas N1, N2 and S1. The clay content in samples used in the pilot phase was almost absent.

2.4. Experimental Setup

The experimental design was based on statistical experimental planning in order to systematically evaluate the performance of specimens under the effects of various binder combinations and water ratios. The mixture optimisation was performed as simplex ternary graphs and process optimisation according to the 2*2-factor experiment. Two independent parameters tested in the study included the following ones: (1) binder combination and (2) water ratio. The amount of binder in a water/binder ratio plays a significant role in final results on the stabilisation of the marine sediments. Thus, the results were checked at the two sets of binder and water ratios (Fig. 7).

2.5. Mixture Optimisation

To determine the optimal mixture between the three different binder components, a simplex experimental test was performed (Fig. 7). A simplex test is summarised as the three different binders represented by the parameters X, Y and Z (as $X + Y + Z = 1$). The equation spans a triangle where the corners are represented by each binder component, here: (1) cement; (2) slag; (3) CKD.

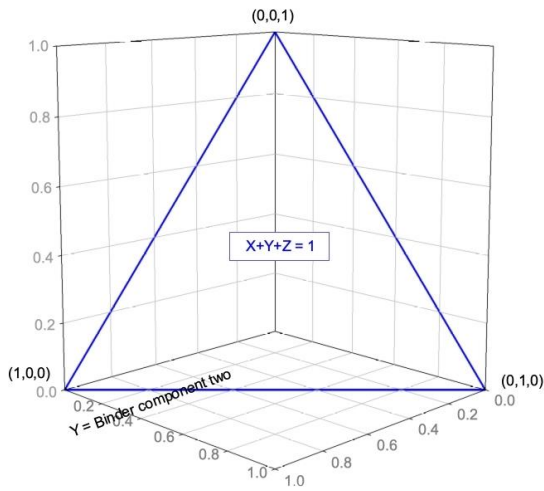


Fig. 7. Graph showing all possible combinations of the three components as different binders presented by a plane with three straight lines bordering the plane. The vertices represent a binder, 1.0 = 100%

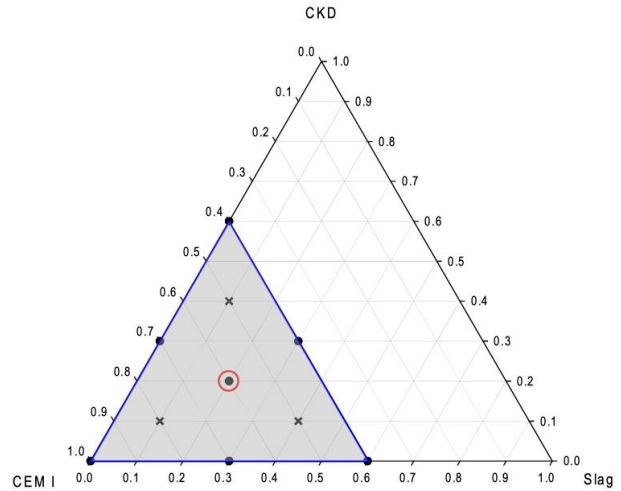


Fig. 9. Augmented simplex experiment for estimation of higher-order interaction effects (cement, slag and CKD). The test points represented by x constitute extra experiments. CKD, cement kiln dust

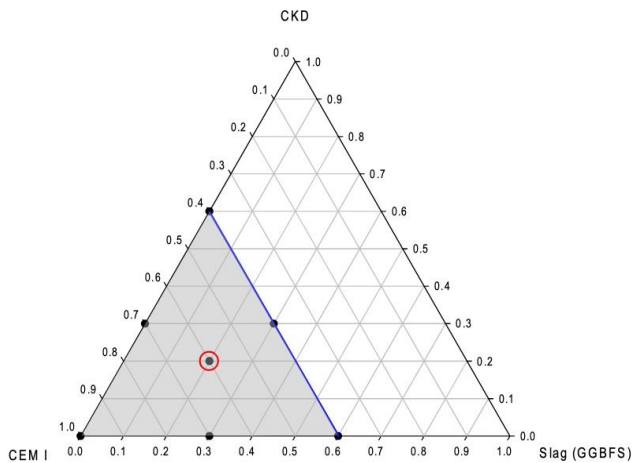


Fig. 8. The grey area shows a constrained simplex centroid experiment (cement, slag and CKD). CKD, cement kiln dust

A mixture of the two binder components can be represented in the inner intersections of the triangle, while a combination of all the three components is shown in the interior of the triangle. The total amount of binder is constant regardless of where the mixture is set up on the triangle, yet the ratio of all three binders vary.

Since only slag or CKD does not act as a binder, a limited simplex centroid experiment has been designed and shown in Fig. 8. Here the limitation is set up to 60% of slag and 60% of CKD. The seven black dots correspond to the selected test samples. A red circle in the middle of the graph (Fig. 8) indicates a double test. Since all samples were double-checked as a double experiment, a sum of 32 (16 × 16) sample experiments has been performed in total.

To increase the resolution, one of the simplex experiments was performed as an augmented simplex experiment (Fig. 9). Some types of binder could be used as a single component as no curing starts, e.g. slag must be activated with a high pH. This problem was solved using a restriction for this binder, e.g., possible combinations were obtained by maximising the slag content or fly ash up to 60%.

2.6. Process Control

The process control included technical parameters controlled according to the standards (Swedish Institute for Standards, 2014a, 2014b, 2021a, 2021b) and changed during the experiment aimed to increase the stability. The process parameters included the following variables: (1) amount of binder (2) water ratio in the sediments (3) time of mixing in a workflow process. The binder mixture in this case is not considered as a process parameter, since it has already been defined and determined during the simplex test.

2.6.1. Processes with Two Factors

The process parameters were used as a 2*2-factor experiment in a simple case where only water ratio and binder content were varied. Here, two factors have been tested: (1) the amount of binder; (2) water ratio. The test was performed on the two levels: low and high water ratio, and low and high amount of binder, respectively see Fig. 10.

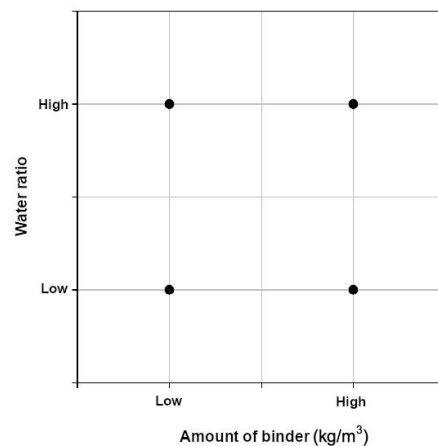


Fig. 10. An example of the 2*2-factor experiment with the amount of binder (cement, slag and CKD) and water ratio as constituent factors. CKD, cement kiln dust

2.6.2. Combination of Approaches

Combinations of various ratios of water/binder in the tests can significantly affect the results of strength, stabilisation and TBT leaching. Therefore, to combine mixing experiments with factor experiments, these were combined as a multi-stage expanded experiment, Fig. 11. This provided an opportunity to study the interplay between the water ratio and the binder quantity on the performance of tested sediments.

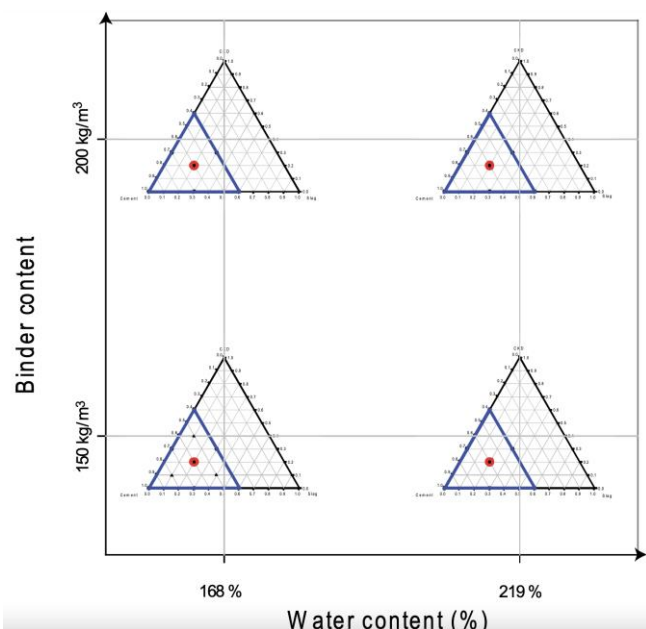


Fig. 11. Simplex experiments arranged following the 2*2-factor experiment. The combined methodology provides both an optimal binder mix (cement, slag and CKD) and an optimal amount of binder concerning the process parameters. CKD, cement kiln dust

Combining simplex experiments with factor experiment aimed to optimise the binder mixture and to determine how the change in mixing parameters affect the result.

2.6.3. Extra Test Setups

The experimental extra test setups (Fig. 11) are based on the methodology of the traditional factor experiment and as a factor experiment in a simplex setup (Fig. 12). The two additional rounds of testing have been performed to determine the limits that could be used to test strength: ED 1 and ED 2 (Fig. 13). These two additional experimental setups were designed as constrained simplex experiments. The experimental setup one (ED1) was designed to optimise the use of CKD and the experimental setup two (ED2) was designed to optimise the use of slag.

The test points marked as black dots were additions to test the limitation of the slag mixture. These experiments were designed to determine the limits of where various binder components can contribute to the increase of the total strength of the dredged sediment masses. Fig. 14 shows different experimental setups with various binder combinations and relative compressive strength. Partial replacement of cement by CKD contributes to the higher strength of the stabilised sediments and increases the environ-

mental quality of mixtures by reducing the amount of cement and replacing it with CKD.

2.7. Determination of the TBT Concentrations from the Leachate

The analysis of the TBT concentrations has been performed by ALS Global. ALS refers to the SIS standard in the accreditation certificate for organotin compounds: ISO 17353:2004 (Swedish Institute for Standards, 2005).

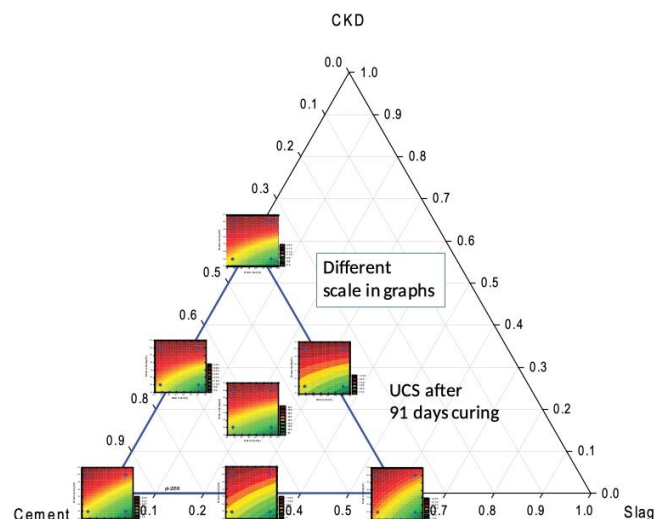


Fig. 12. Ternary diagram showing 2*2-factor experiments reported in a simplex setup of binders (cement, slag and CKD). CKD, cement kiln dust

In this specific test package, the TBT substances have been extracted in a liquid-liquid extraction from water to hexane. The instrument that is then used in the analysis is one Gas Chromatography – Inductively Coupled Plasma – Sector Field Mass Spectrometry (GC-ICP-SFMS), <https://www.alsglobal.se/en/isotope-analysis/laboratory>. In this approach different TBT's were separated from each other in the GC system.

Separation in GC takes place due to the different sizes of the TBT substances, fat solubility, charge, etcetera. The substances were then passed on to the plasma which excites and ionises the tin atoms which were then passed through the mass spectrometer which can further separate the tin atoms depending on their m/z value (mass-to-charge ratio, in this way one can also distinguish isotopes of the same substance).

Finally, the atoms/molecular fragments reached a detector that generated a signal. This means that the tin atoms reach the detector at different times because the different TBT substances are separated over time in the GC system. In this way, it is possible to correlate a specific TBT to a specific time when the signal (peak) is registered. The area on the peak is in proportion to the amount of substance at that time.

3. RESULTS AND DISCUSSION

The study has evaluated the effect of adding three different binders (cement, CKD and slag) in various proportions and changing water ratio to assess the TBT leaching and strength of the

contaminated marine sediments after stabilisation. The results included tests of the following parameters in sediments:

1. compressive strength;
2. permeability;
3. TBT leaching.

The results of the test experiments performed with a factor combination of low water ratio (LW) and low amount of binder (LB), cement, slag and CKD in various proportions showed that the largest increase in the compressive strength occurred with a mixture of 60% slag and 40% cement. Thus, water holding capacity increases with the increased binder content, slag proportion in a binder and curing time suggesting that high-slag binders are the most suitable for removing contaminants.

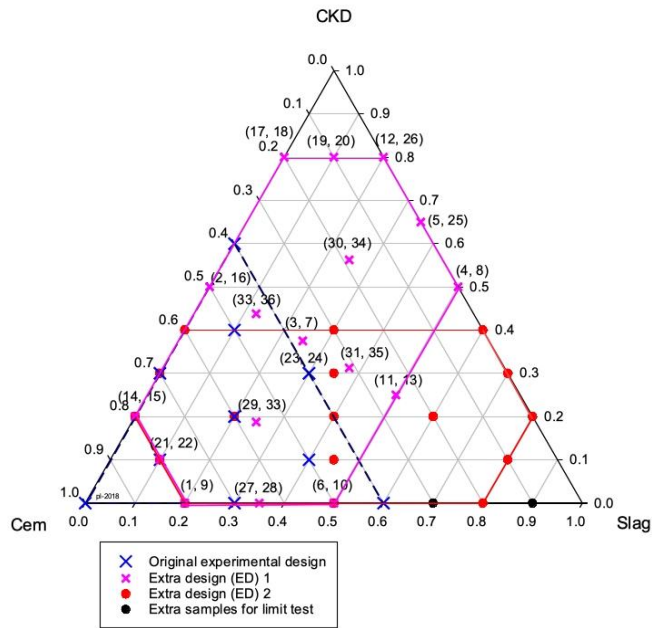


Fig. 13. Ternary diagram of different experiments performed with the factor combination LW and LB, cement, slag and CKD. CKD, cement kiln dust; LB, low amount of binder; LW, low water ratio

3.1. Leaching of TBT with Addition of CKD

The samples of specimens with sediments were evaluated on TBT leaching after 2.25 days, 9 days and 36 days of stabilisation. The results of the leaching test were compared for different content of binder and water ratios. The bracketed pairs of values superimposed on Figs. 14 and 15 indicate the compressive strengths measured for the double experiments for higher and lower water ratio and binder content, which explains the large disparity in some cases, e.g. (5,25), (1,9). Here the combinations that did not show an increase in the compressive strength is caused by a too low pH to activate slag (Fig. 14).

The results on leaching for day 2.25 are shown in Fig. 15. Here the variations in the TBT leaching are shown after 2.25 days of stabilisation for the binder ratio (maximal values): 0.6 slag, 0.4 CKD and high water – low binder ratio at 19, 23, 34 and 37.

The colour values in Fig. 15 show values of the TBT leaching (ng/L) as follows: <480, 580, 680, 780, 880, and >900 for each colour gradation on the ternary diagram from dark green to dark red, accordingly. The highest values (red colours) are notable for higher concentrations of cement and CKD while the lowest (green

colours), corresponding to the <480 ng/L correlate with higher proportions of slag.

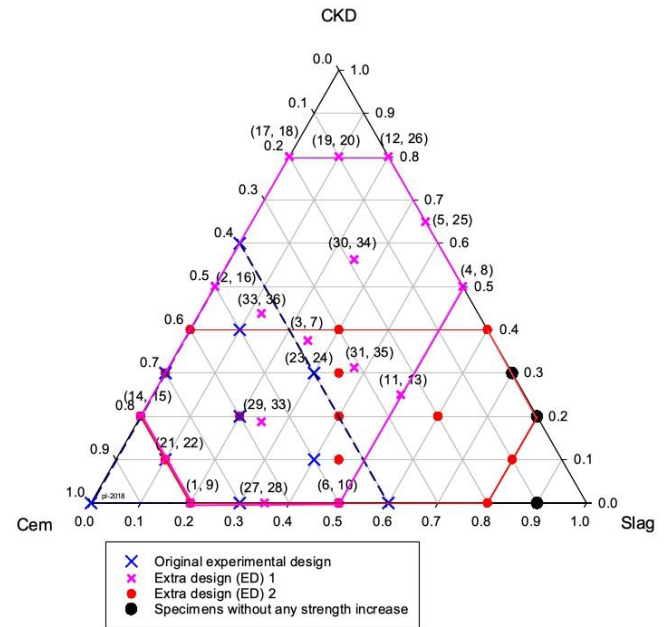


Fig. 14. Ternary diagram showing test and experimental points (approaches) where no growth strength was detected. CKD, cement kiln dust

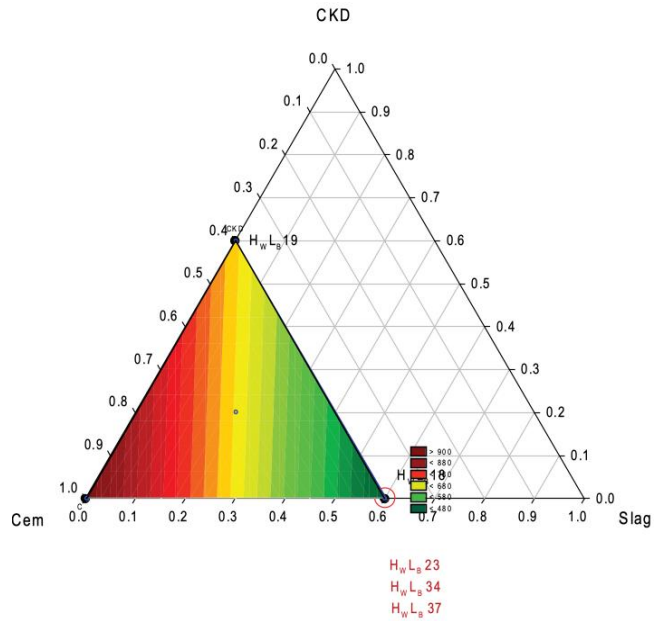


Fig. 15. Ternary diagram showing response area for leaching of TBT after 2.25 days. TBT, tributyltin; CKD, cement kiln dust

Fig. 16 shows a correlation between the increasing pH values of the tested specimens with added CKD and the leaching of TBT. For comparison, mixtures with pure cement and “cement/slag” ratio were used in the pilot project. For statistically independent comparison, the standardised samples were used in this project with pure cement and cement/slag combinations. In general, there is a linear correlation between the increasing pH values and higher leaching despite the different samples.

The R2 value of 0.62 shows that the pH explains a fairly large part of the variation in leaching of the sediment samples. However, parts of the variation are due to the differences in leaching of TBT over time. Since the points for the recipe cement/slag/CKD are slightly below the trend line, it indicates that samples give a slightly lower leaching effect than expected based on the pH of the leachate. The cement/slag/CKD generated a higher pH value than cement/slag, which should have given higher leaching because the natural pH of soil affects the stability or functionality of agents and different mineralogy. Leaching obtained is judged to be close to equivalent to cement/slag, Fig. 16.

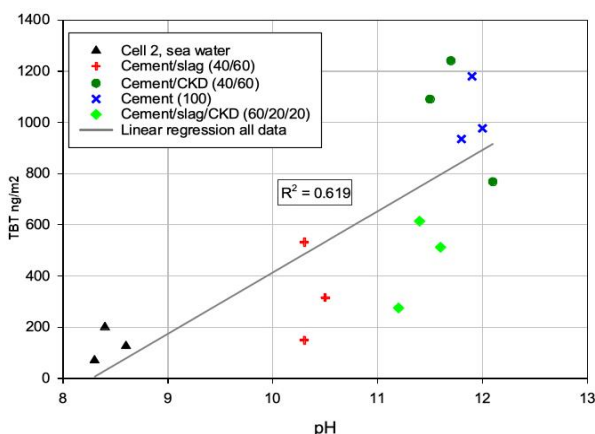


Fig. 16. Correlation between the pH and leached amount of TBT by various binders in defined ratios. TBT, tributyltin; CKD, cement kiln dust

The cement/slag/CKD mixture thus can contribute to the reduced leaching through a mechanism that is not dependent on the pH. Chemical composition and the pH of soils are reflected in their industrial impact (Hiller et al., 2021) with the leaching of heavy metals demonstrating the pH-dependent behavior (Szarek-Gwiazda, 2014; Ai et al., 2019; Suzuki et al., 2020; Sun & Yi, 2021; Nyembwe et al., 2021). In this study, leaching has shown a direct correlation with the higher pH corresponding to the increased leaching (Fig. 16).

3.2. Leaching of TBT Over Time

The sample specimens of sediments were received from the two different batches (Cell 1 and Cell 2). Washing specimens was performed during the first step of the leaching tests. The performance of the laboratory-made specimens in Cell 1 and the *in-situ* specimens stored in Cell 2 demonstrated differences in leaching experiments. Cell 2 provided a more stable leaching pattern with lower mass flows compared to the laboratory-produced specimens that were not subject to leaching.

It can be explained by the technical design of the test: when specimens were removed from Cell 2 at the end of the leaching tests, the soil was already washed out. The hypothesis of washing also explains why shaking experiments on crushed material from Cell 2 showed 10–100 times lower leaching than crushed materials from the laboratory-produced samples during the data evaluation from the pilot test control programme.

In all experiments, the processed samples in the laboratory demonstrated leaching to a greater extent than the field sample from Cell 2 during the first time steps of the experiments (Fig. 17).

However, for the most of samples, the differences between the lab and the field sample decreased after 36 days. Such a trend is noted both for the TBT and for potassium. Both substances are assumed to have leaching controlled in whole or in part by leaching in the initial phase.

The results of the experimental tests on leaching TBT over time for all binders (cement, slag and CKD) and changed amounts of water are shown in Fig. 17. The cement/slag/CKD mixture of binder is represented by a double experiment when the sediment specimens were tested on two different occasions.

The results of the experiments on leaching of TBT over time for all samples with varied ratio of binders (cement, slag and CKD) and changed amounts of water demonstrated the following outcomes (Fig. 17): the ratio of cement/slag/CKD 60/20/20: 500 ng/m² at day 2.25, 600 ng/m² at day 9, 280 ng/m² at day 36; cement/CKD 40/60: 780 ng/m² at day 2.25, 1220 ng/m² at day 9, 1100 ng/m² at day 36; cement/slag 40/60 (no CKD): cement/slag 40/60 (no CKD): 580 ng/m² at day 2.25, 310 ng/m² at day 9, 180 ng/m² at day 36. The increase of leaching was observed for the following analysed parameters: a higher amount of water in the experiments, the addition of CKD to cement (ratio of cement/CKD 40/60) contributes to higher leaching compared to ratio cement/slag.

The tests demonstrated notable leaching of TBT from the sediment samples on day 9 and significantly decreased amount of TBT on day 36 compared to the results on day 2.25 of the experiment (Fig. 17). On day 2.25, a relatively high variability is seen between the experiments. On days 9 and 36, the variability is notably lower. The experimental results show that the speed of leaching of TBT from the sediments changes at different ratios of added binders and water for TBT leaching in a seawater environment.

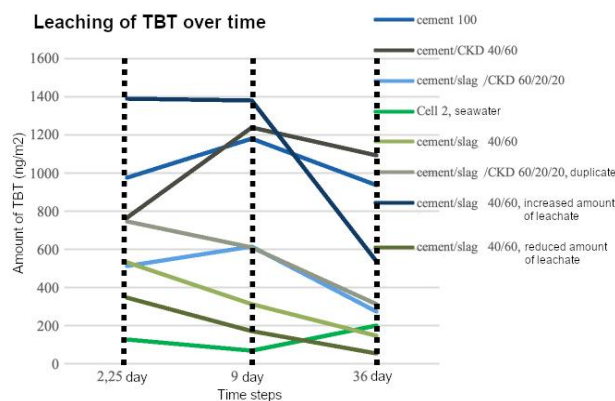


Fig. 17. Leaching of TBT over time for all binders (cement, slag and CKD) and changed amounts of water in leaching experiment. CKD, cement kiln dust; TBT, tributyltin.

The results show that variation of the ratio of the independent binders affects TBT leaching in the sediment specimens. These include binder ratios (cement/slag/CKD), water ratio in sediment samples) and time of the experiment showing time-dependent leaching. The binder and water ratio are the primary factors affecting the TBT leaching of the tested sediment samples.

The pH of mixtures containing cement/slag stabilised around 10–10.5 in the performed lab experiments. Leaching varies considerably between different samples in simplex experimental tests. However, leaching stabilised over time, with a reduced difference between the ratio. Sediment samples that contain ce-

ment/slag/CKD in ratio 60%/20%/20% give similar leaching results as those with ratio cement/slag (60%/40%), which is identical to the results received in a pilot experiment.

Leaching of TBT is affected not only by the pH value but also by leaching conditions (i.e. ratio between leached surface and leachate amount). For example, the leaching of TBT increased at a higher amount of water in the surface leaching tests, because of the buffering effect of the seawater. The results of performed tests on the treatment of the contaminated sediments showed the applicability of CKD as a cement replacement in soil stabilisation and TBT leaching of soil specimens. As the experiment length was limited by 36 days, the results are related to the short-term leaching conditions in the field based on the processing sediments obtain *in situ* from the Port of Gothenborg.

3.3. Unconfined Compressive Strength (UCS)

The results for the UCS are presented for the two experimental setups (one (ED1), designed to optimise the use of CKD and two (ED2), designed to optimise the use of slag), as described in subsection 2.6.3. Figs 18 and 19 show variations in the unconfined compressive strength (UCS) by various combinations of the binder in the specimen. Within tested combinations, the analysis of changes in UCS (gradual changes from dark to light green in Fig. 18 and from green to red colours in Fig. 19) revealed changes in UCS.

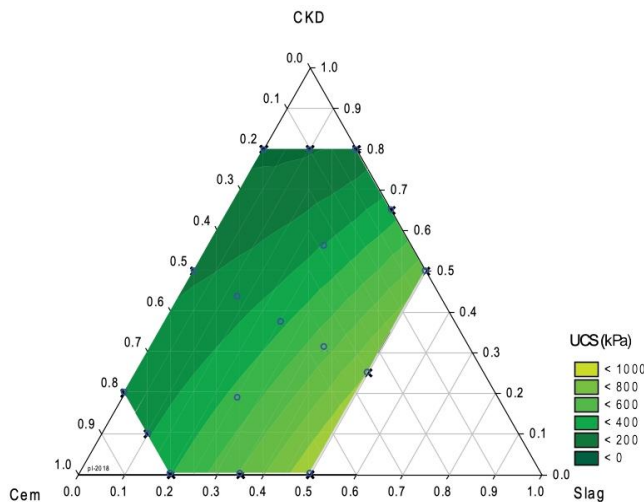


Fig. 18. ED1 compressive strength for binders (cement, slag and CKD). CKD, cement kiln dust; UCS, unconfined compressive strength

Thus, Fig. 18 shows the increase of UCS with the increase of cement in a binder from 0.2 to 0.5 (changes from dark green to light green colours in the horizontal axis) and CKD from 0.2 to 0.8 (left axis areas in Fig. 18) and inversely proportional for slag (an increase of UCS along with the decrease of slag from 0.5 to 0.8, the right axis in Fig. 18). Accordingly, Fig. 19 shows the increase of UCS with the increase of cement in a binder from 0.2 to 0.8 and CKD from 0.6 to 0.8 (bright red areas in Fig. 19).

The significant increase of UCS is noticeable along with the increase of cement proportion from values 0.4 to 0.8 (corresponds to the red areas in Fig. 19). These ratios of binders shown in the ternary diagrams presented an increase of values of UCS with

adding of cement (0.30–0.38 increased values from 700 up to 900, orange colours).

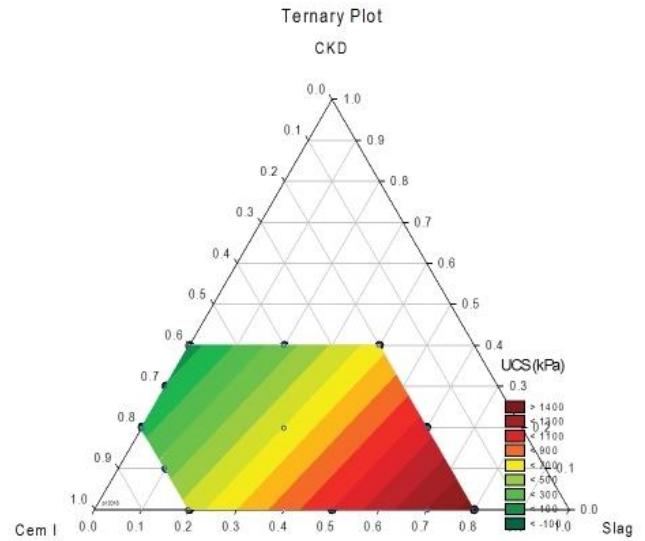


Fig. 19. ED2 compressive strength for binders (cement, slag and CKD). CKD, cement kiln dust; UCS, unconfined compressive strength

Further gradual increase in compressive strength is noticeable and reaches its maximum at values of cement from 0.7 to 0.8 and CKD from 0.8 to 1.0 (crimson red colours, Fig. 19). Adding of slag presents the controversial effects of decreasing the UCS with an increase of slag in the ratio (from 0.0 to 0.4, the right axis in Fig. 19).

4. CONCLUSION

This study presented the results of detailed experiments of the s/s treatment of TBT contaminated sediments using CKD, cement and slag in different proportions. The application of three-component binders for the treatment of marine sediments results in the development of stabilised and solidified compact soils (produced in various combinations of cement, CKD and slag with a varied ratio of water) with removed TBT contaminants.

Treatment of the marine sediments contaminated by TBT is important since cleaned and stabilised sediments can be recycled and reused in the construction works. The performed tests showed variations in leaching of TBT in the specimens at different binder ratios (cement, slag, CKD). The laboratory tests were performed to assess the applicability of CKD as a cement replacement for contaminated sediments and to assess the leaching and strength of samples.

The study demonstrated that CKD has promising applications for s/s treatment of the marine sediments contaminated by TBT. The addition of CKD contributes to a higher pH compared to the binders cement/slag. At the same time, CKD affected leaching, despite the alkaline effect of the material on pH. The samples with only added cement/CKD demonstrated higher leaching compared to the ratio “cement/slag/CKD” and “cement/slag”. The “CKD/slag” ratio presented the best results and can be used for s/s treatment of the sediments.

The results also proved that using CKD as a cement replacement in the treatment of TBT contaminated marine sediments is beneficial not only and environmentally (effective in TBT leach-

ing), but also technically (increased UCS and stability of specimens). Thus, the gradual increase in UCS has been noted and reached its maximum along with the increased values of cement and CKD.

The actuality of this study consists in theoretical and practical aspects. The first includes the increased knowledge on the performance of sediments in tests on leaching and stabilisation. The second includes practical tests of the technical methods on s/s treatment of the contaminated sediments.

Environmental monitoring in Sweden includes coastal regions of the Baltic Sea (Sveriges geologiska undersökning, 2021). Therefore, the development of methods for the s/s treatment of the contaminated marine sediments is an essential task (Cato, 1977; Sánchez-García et al., 2010). Presented laboratory experiments demonstrated changes in leaching in various combinations of binder (cement, CKD, slag) which contributes to the marine environmental monitoring in Sweden.

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Per Lindh:  <https://orcid.org/0000-0002-0577-9936>

Polina Lemenkova:  <https://orcid.org/0000-0002-5759-1089>