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# An innovative stabilization/solidification treatment For contaminated soil remediation: demonstration project results

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**Abstract**

*Background, aim, and scope* An innovative stabilization/solidification (S/S) process using high-performance additivated concrete technology was developed for remediating soil contaminated by metals from abandoned industrial sites. In order to verify the effectiveness of this new ex situ S/S procedure, an area highly contaminated by metallic pollutants (As, Cd, Hg, and Pb), due to the uncontrolled discharge of waste generated from artistic glass production on the island of Murano (Venice, Italy), was selected as a case study. The technique transforms the contaminated soil into an aggregate material suitable for reuse as on-site backfill. This paper reports the main results of the demonstration project performed in collaboration with the local environmental protection agency (ARPAV).

*Materials and methods* An ex situ treatment for brownfield remediation, based on the transformation of contaminated soil into very dense, low porous, and mechanically resistant granular material, was set up and tested. Specific additives (water reducers and superplasticizers) to improve the stabilized material properties were developed and patented. A demonstration plant assembled on the study area to treat 6 $m^3$ $h^{-1}$was then tested. After excavation, the contaminated soil was screened to remove coarse material. The fraction Ø >4 mm (coarse fraction), mainly composed of glass, brick, concrete, and stone debris, was directly reused on site after passing through a washing treatment section. The highly polluted fraction Ø≤4 mm (fine fraction) was treated in the S/S treatment division of the plant (European patent WO/2006/097272). The fine fraction was mixed with Portland cement and additives defined on the basis of the high performance concrete technique. The mixture was then granulated in a rolling-plate system. After 28 days curing in an onsite storage area to allow for cement hydration, the stabilized material was monitored before its in situ relocation. The chemical, mechanical, and ecotoxicological reliability and performance of the treatment was checked. Metal leachability was verified according to four leaching test methods: Italian Environmental Ministry Decree (1998), EN 12457 (2002) *tout court*, amended only with $MgSO_4$ and, lastly, with artificial sea water. The mechanical properties were measured according to BS (1990) and AASHTO (1999) to obtain the Aggregate Crushing Value

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and California Bearing Ratio, in that order. Moreover, leachate samples prepared with artificial seawater were assessed via the *Crassostrea gigas* embryotoxicity test and *Vibrio fischeri* bioluminescence inhibition test to discriminate the presence of potential ecotoxicological effects for the brackish and saltwater biota.

*Results* Outcomes from all leachate samples highlighted the effectiveness of the remediation treatment, fully complying with the Italian legislation for non-hazardous material reuse under a physicochemical viewpoint. The stabilized granular material demonstrated high mechanical strength, low porosity, and leachability. Moreover, ecotoxicological surveys indicated the presence of low toxicity levels in leachate samples according to both toxicity tests.

*Discussion* Remediated soil samples revealed a significant decrease in leachability of heavy metals as a consequence of the application of additivated cement that enhanced granular material properties, resulting in improved compactness due to the reduction in water content. The toxicity data confirmed this state-of-the-art technique, indicating that leachates could be deemed as minor acutely toxic.

*Conclusions* The proposed S/S treatment proved to be able to remediate soil contaminated by heavy metals through trapping pollutants in pellet materials presenting adequate physicochemical, mechanical, and ecotoxicological properties in order to prevent leachability phenomena, their reclamation, and reuse being made easier by its granular form.

*Recommendation and perspectives* This project foresees long-term monitoring activity over several years (until 2014) to consider treatment durability.

**Keywords** Heavy metals contamination · High-performance concrete (HPC) · Leaching tests · Soil remediation · Stabilization/Solidification · Toxicity monitoring

## 1 Background, aim, and scope

Thousands of industrial soils around the world are contaminated by metallic pollutants, mainly due to waste deposits, chemical leakages, or fall-out from atmospheric emissions. Various in situ and ex situ techniques have been used to reduce the impact of metals in the soil, including excavation, solidification, stabilization, soil washing, electroremediation and phytoremediation. Excavation and disposal, in particular, is quite an expensive procedure because landfill costs have recently increased as have those of new soil importation to replace that which has been removed (Tandy et al. 2009).

Solidification/stabilization (S/S) technologies, especially as in situ applications, are very common in the USA (Conner 1990; USEPA 2000) and are increasing in Europe (EPA UK 2004; Van-Camp et al. 2004; Harbottle et al. 2007; SedNet 2008). Solidification consists of the physical encapsulation of contaminants in a solid matrix, while stabilization involves chemical reactions to reduce contaminant mobility (Mulligan et al. 2001). Basically, the S/S process is the result of mixing contaminated soil with binders (mostly cement or mixtures of cement and lime) and perhaps specific additives in order to support the precipitation of heavy metals in the form of hydroxyls and the reduction of their mobility by cement hydration (USEPA 2002).

Nevertheless, traditional S/S in situ treatments do not appear to be very reliable in guaranteeing the immobilization of metal contamination not only in the surface soil but also at depth (Al-Tabbaa and Evans 1996) and the stabilized material tends to maintain the starting soil-specific surface and permeability to water, which could negatively influence the durability of the leaching performance. These disadvantages are further worsened when the stabilized material is exposed to seawater that contains aggressive agents such as chloride and sulphate ions, which might further reduce the remediated soil resilience. Besides, there are many applicability constraints for in situ S/S use related to excess moisture, soil inhomogeneity, presence of debris, and deep contamination (Mulligan et al. 2001). It, thus, appears that the in situ application constraints might be easily controlled via an ex situ approach.

A new ex situ S/S treatment procedure to remediate soil contaminated by metallic pollutants was developed and specifically addressed to granular material production, in order to facilitate treated soil reuse for excavation landfill or other nonstructural purposes.

The aim is to produce a reusable mechanically resistant, cement-based granular material with low leachability rates, and no relevant ecotoxicological effects, targeting, in particular, brackish and marine biota. Specifically, this approach is oriented to reduce contaminated brownfield remediation costs, limiting waste soil dumping, and supporting soil reclamation and reuse.

The most relevant innovations of the developed S/S approach are:

1. the application of a preliminary mechanical size selection used to separate large and relatively clean particles, from small and polluted ones;
2. the addition of innovative additives to the soil finer fraction that allow high-performance soil stabilization levels;
3. providing the stabilized soil with a granular form that can be easily managed, also in the case of maintenance work for underground pipelines.

This paper reports the results of the validation program of physicochemical, mechanical, and ecotoxicological properties of by-products generated through this innovative stabilization/solidification technique performed at a large-scale demonstration level.

## 2 Materials and methods

### 2.1 The study area

After many laboratory and pilot tests, a demonstration project for the evaluation of this new technology was planned for a selected study area on the island of Murano (Venice, Italy), where an artistic glass factory was operating until 1993. The soil was heavily contaminated by metal oxides as well as silicates. In 1995, Venice Municipality acquired the area for housing purposes, but soil remediation was required. A survey showed that the contamination was mainly due to metallic pollutants such as arsenic (As), cadmium (Cd), mercury (Hg), and lead (Pb), the average concentrations of which are shown in Table 1, over an area of about 14,000 m$^2$ for a mean depth of 2 m. The large volume of contaminated soil (30,000 m$^3$) and its location discouraged the option, under both an economic and practical viewpoint, of completely removing the contaminated soil and replacing it with another with suitable characteristics. It was, therefore, decided to experiment with a new S/S technique in collaboration with the local authorities.

### 2.2 S/S demonstration plant and procedure

The demonstration plant placed on-site was designed to treat about 6 m$^3$ h$^{-1}$ and composed of a series of operating units as explained in Fig. 1. First of all, the contaminated soil was excavated and screened. The coarse fraction (Ø> 4 mm) was mainly composed of glass, brick, concrete, and stone debris and could be immediately reused after a washing procedure with tap water, whereas the resulting wastewater was treated in a physicochemical wastewater treatment plant that allows its immediate reuse during the washing process. Conversely, the highly polluted fine fraction (FF; Ø≤4 mm), mainly composed of silt and clay, was considered for S/S treatment.

The proposed S/S technique, covered by a European patent (WIPO 2006), was designed to produce stabilized material in granular form by mixing contaminated soil (FF) with Portland cement, water, and a specific additive (SA) called Mapeplast ECO1 with water reducers and waterproofing agents. A typical formulation is: 65–70% of FF, 23–25% of Portland Cement (CEM I 52.5 R), 2% of the SA, and 3–5% of water. The specific additive is adjoined to the mixture to improve S/S performance on the basis of the High-Performance Concrete (HPC) approach (Russell 1999), allowing the water/cement ratio (W/C) to be reduced to 0.36 and enhancing the ability to support stabilized long-term mechanical soil properties, increasing density, lowering leachability, and prolonging its life in severe conditions. Indeed, HPC is a relatively new term that is used to describe concrete obtained from specific cement and additives that conforms to a set of standards above those of the most common applications. This approach allows a W/C<0.4 to be achieved (Nawy 2000), and the considered SA allows better results to be obtained than other conventionally marketed additives, as stated in the relative patent (WO/2006/097272; WIPO 2006).

Table 1 Mean average background values of soil contamination by metallic pollutants from the study area

| Heavy metals | Soil concentrations | Residential use limits[a] |
|---|---|---|
| As (mg kg$^{-1}$ dm[b]) | 204 | 20 |
| Cd (mg kg$^{-1}$ dm[b]) | 12 | 2 |
| Hg (mg kg$^{-1}$ dm[b]) | 1.8 | 1 |
| Pb (mg kg$^{-1}$ dm[b]) | 2.321 | 100 |

[a] EMD (1999)

[b] Soil dry matter

The mixture is granulated via a rolling-plate system. The rotation of the plate and presence of the SA promote the formation of the "micrograins." The grain-specific surface area and its corresponding diameter, maintained in the range 20–100 mm, are controlled by changing the process parameters (speed and inclination of the plate). After 28 days curing in a storage area dedicated to allow cement hydration, the stabilized material is deemed as ready to be replaced in situ. The washed coarse fraction and stabilized granular soil material are both reused as excavation filler.

### 2.3 Soil sampling and sample preparation

A total of 5,000 m$^3$ of polluted soil has been treated, resulting in 2,500 m$^3$ of granular material and 2,500 m$^3$ of clean coarse fraction. During the S/S treatment, subsamples were collected of the granular material by-product, resulting in six final composite samples (P1, P2, P3, P4, P5, and P6). Each subsample is representative of a 250 m$^3$ lot of stabilized grains.

The assessment of sample leachability was evaluated by four leaching protocols:

1. Samples were individually and repeatedly (time-by-time leaching solution renewal) leached using increasing contact times (2, 8, 24, 48, 72, 102, 168, 384 h) in order to compare the relative cumulative concentration of heavy metals with the regulatory requirements (Solid/Liquid ratio (S/L)=1:10; EMD 1998)
2. Samples were leached with deionized water (24 h, S/L= 1:10) under mechanical agitation according to the EN 12457-1-4:2002 procedure
3. Samples were leached with deionized water amended with $MgSO_4$ (10 g l$^{-1}$) (24 h, S/L=1:10) to simulate the

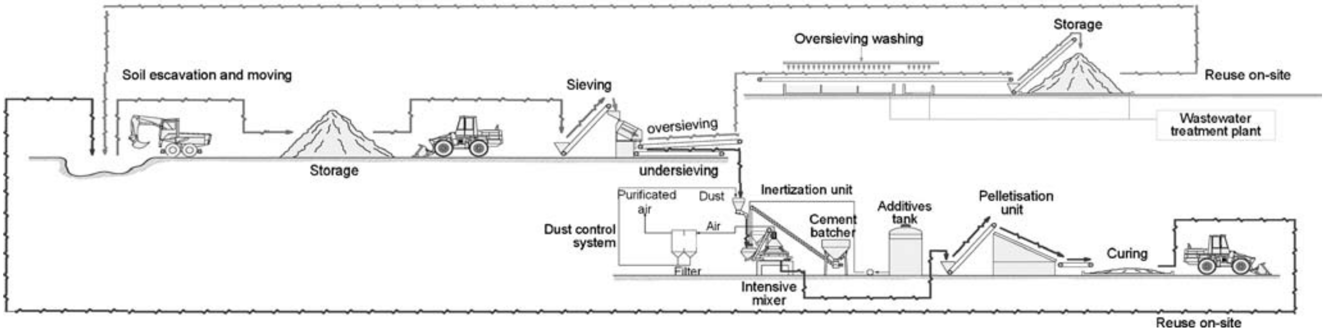

**Fig. 1** Contaminated soil remediation treatment flow chart

presence of salt water under mechanical agitation as in the EN 12457-1-4:2002 procedure

4. Samples were individually leached using artificial seawater (ASTM 2004; 24 h, S/L=1:10) under mechanical agitation to simulate potential metallic pollutants being released to salt water and causing ecotoxicological effects.

The EMD (1998) (1) and artificial seawater as leaching medium (4) procedures considered all samples, whereas the method EN 12457-1-4:2002 (2) and EN 12457-1-4:2002 amended with $MgSO_4$ (3) only P1, P2 and P3, and P4, P5, and P6, in that order. Leachate samples for physicochemical analyses were stored at room temperature (20±1°C) after acidification and filtering (0.45 μm sterile acetate nitrate

**Table 2** Metallic concentrations in leachates (EMD (1998) (1)) generated from pellet samples

| | Heavy metals | Cumulative leaching time (h) | | | | | | | | $\Sigma$ | Regulatory limits[a] |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | 2 | 8 | 24 | 48 | 72 | 102 | 168 | 384 | | |
| P1 | As ($\mu$g l$^{-1}$) | 9.9 | 6.2 | 3.7 | 1 | 0.6 | 0.5 | <0.5 | <0.5 | 22 | 50 |
| | Cd ($\mu$g l$^{-1}$) | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <4 | 5 |
| | Hg ($\mu$g l$^{-1}$) | 0.1 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | 0.1 | 1 |
| | Pb ($\mu$g l$^{-1}$) | <5 | <5 | <5 | <5 | <5 | <5 | <5 | <5 | <40 | 50 |
| P2 | As ($\mu$g l$^{-1}$) | 1.1 | 0.8 | 0.7 | 2 | 1 | 1 | <1 | 2 | 9 | 50 |
| | Cd ($\mu$g l$^{-1}$) | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <4 | 5 |
| | Hg ($\mu$g l$^{-1}$) | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.4 | 1 |
| | Pb ($\mu$g l$^{-1}$) | <5 | <5 | <5 | <5 | <5 | <5 | <5 | <5 | <40 | 50 |
| P3 | As ($\mu$g l$^{-1}$) | 1.4 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | 1 | 50 |
| | Cd ($\mu$g l$^{-1}$) | <0.2 | <0.2 | <0.2 | <0.2 | <0.2 | <0.2 | <0.2 | <0.2 | <4 | 5 |
| | Hg ($\mu$g l$^{-1}$) | 0.06 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | 0.06 | 1 |
| | Pb ($\mu$g l$^{-1}$) | <5 | <5 | <5 | <5 | <5 | <5 | <5 | <5 | <40 | 50 |
| P4 | As ($\mu$g l$^{-1}$) | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <4 | 50 |
| | Cd ($\mu$g l$^{-1}$) | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <1.6 | 5 |
| | Hg ($\mu$g l$^{-1}$) | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.4 | 1 |
| | Pb ($\mu$g l$^{-1}$) | <1.3 | <1.3 | <1.3 | <1.3 | <1.3 | <1.3 | <1.3 | <1.3 | <10.4 | 50 |
| P5 | As ($\mu$g l$^{-1}$) | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <4 | 50 |
| | Cd ($\mu$g l$^{-1}$) | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <1.6 | 5 |
| | Hg ($\mu$g l$^{-1}$) | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.4 | 1 |
| | Pb ($\mu$g l$^{-1}$) | <1.3 | <1.3 | <1.3 | <1.3 | <1.3 | <1.3 | <1.3 | <1.3 | <10.4 | 50 |
| P6 | As ($\mu$g l$^{-1}$) | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <4 | 50 |
| | Cd ($\mu$g l$^{-1}$) | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | <1.6 | 5 |
| | Hg ($\mu$g l$^{-1}$) | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.05 | <0.4 | 1 |
| | Pb ($\mu$g l$^{-1}$) | <1.3 | <1.3 | <1.3 | <1.3 | <1.3 | <1.3 | <1.3 | <1.3 | <10.4 | 50 |

[a] EMD (1998)

**Table 3** Metallic concentrations in leachates (EN (2002) (2)) generated from pellet samples

| Heavy metals | P1 | | P2 | | P3 | | Regulatory limits[a] |
|---|---|---|---|---|---|---|---|
| | Before S/S | After S/S | Before S/S | After S/S | Before S/S | After S/S | |
| As ($\mu g\ l^{-1}$) | 86 | 3 | 103 | <1 | 96 | <1 | 10 |
| Cd ($\mu g\ l^{-1}$) | 5 | <1 | 3.0 | <1 | 3 | <1 | 5 |
| Hg ($\mu g\ l^{-1}$) | 1 | <0.5 | <0.5 | <0.5 | <0.5 | <0.5 | 1 |
| Pb ($\mu g\ l^{-1}$) | 11 | <3 | 7 | <3 | 5 | <3 | 10 |

[a] EMD (1999)

filter), while those for ecotoxicological evaluations were kept at 4°C±1°C for no more than 3 days.

### 2.4 Physicochemical and mechanical analyses

The pH was measured using a pH meter HI 9025 Microcomputer from HANNA Instrument®, salinity was checked with a refractometer, and Dissolved Oxygen (DO) by a WTW multi-parametric device.

As (Limit of Detection (LOD)=1 $\mu g\ l^{-1}$, $p=0.01$), Cd (LOD=1 $\mu g\ l^{-1}$, $p=0.01$), Hg (LOD=0.5 $\mu g\ l^{-1}$, $p=0.05$), and Pb (LOD=3 $\mu g\ l^{-1}$, $p=0.01$) were detected via Inductively Coupled Plasma Atomic Optical Emission Spectroscopy (ICP-OES; Spectro Flame Compact E, Analytical Instruments; USEPA 1992; APHA 1998; APAT and IRSA-CNR 2003).

The mechanical characteristics were verified after curing to verify the granular material suitability as filler (BS 1990). The protocol required the granular material crushed fraction to be measured after 100 kN load application, providing the aggregated crushing value (ACV), that is the weight percentage ratio between the crushed material fraction after the load application passing a 2-mm mesh sieve ($M_2$) and its initial amount before the test start ($M_1$) (ACV=100 ($M_2\ M_1^{-1}$)). The higher the ACV value the lower the mechanical characteristics of aggregated material. In addition, the California Bearing Ratio (CBR) test was performed according to the AASHTO (1999) method to compare the bearing capacity of stabilized granular material with that of well-graded crushed stone.

### 2.5 Toxicity test

Toxicity bioassays were performed according to standardized protocols with bioindicator species that are widely applied and recognized to assess potential ecotoxicological effects to transitional and marine biota. The embryotoxicity test with the oyster *Crassostrea gigas* (Thunberg) was conducted on the basis of the ASTM (2004) method modified for the gametes pool (Libralato et al. 2007), while the bioluminescent inhibition test with the bacterium *Vibrio fischeri* was executed according to the Azur Environmental (1998) 100% protocol. All tests were performed in triplicate using a geometrical scale (1.5%, 3%, 6%, 12%, 25%, 50%, 100% of leachate).

Toxicity data were expressed as Toxic Unit at 50% of the population exhibiting a response (TU50) which was derived from Effect Concentration where 50% of the population exhibited a response (EC50) value (TU50=100 ($EC50^{-1}$)). When EC50 value was not quantifiable, toxicity was expressed as percentage of effect (PE). TU50 data were displayed with 95% confidence limit values (Libralato et al. 2007).

Toxicity data were integrated according to the Wilke et al. (2008) scoring system for ecotoxicological waste characterization. The system established that a leachate Sample (S) presents a score of 1 if S≤1 TU50 (not toxic), a score of 2 if 1 TU50<S≤10 TU50 (minor acutely toxic), a score of 3 if 10 TU50<S≤100 TU50 (major acutely toxic) and a score of 4 if S≥100 TU50 (highly toxic). The final class weight score is then calculated dividing the sum of all test scores for the same sample by the number of tests performed.

## 3 Results

### 3.1 Leaching and mechanical data

The average values of chemicals (As, Cd, Hg, and Pb) originating from the survey area and reported in Table 1

**Table 4** Metallic concentrations in leachates (EN (2002)) amended with $MgSO_4$ (3)) generated from pellet samples

| Heavy metals | P4 After S/S | P5 After S/S | P6 After S/S | Regulatory limits[a] |
|---|---|---|---|---|
| As ($\mu g\ l^{-1}$) | <1 | <1 | <1 | 10 |
| Cd ($\mu g\ l^{-1}$) | <1 | <1 | <1 | 5 |
| Hg ($\mu g\ l^{-1}$) | <0.5 | <0.5 | <0.5 | 1 |
| Pb ($\mu g\ l^{-1}$) | <3 | <3 | <3 | 10 |

[a] EMD (1999)

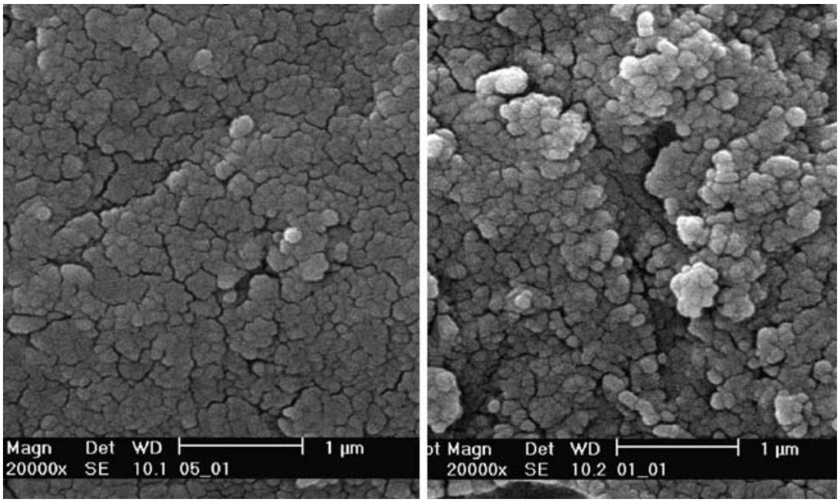

**Fig. 2** Scanning electron microscope pictures of S/S-treated soil with (*left*) and without (*right*) the patented additive

showed that all considered metalloid and metal concentrations exceeded the relative Italian standard for soil residential use (EMD 1999; Surico at al. 2003).

Chemical analyses on leachates generated according to methods (1), (2), and (3) are displayed in Tables 2, 3, and 4, respectively. Data from Table 2 highlighted that the cumulative concentration of As, Cd, Hg, and Pb was lower than the relative regulatory limits (EMD 1998) for all samples.

The results in Table 3 show that leachates from samples P1, P2, and P3 did not exceed regulatory limits for groundwater (EMD 1999), nor did leachates from samples P4, P5, and P6, as indicated in Table 4. These results confirmed the curing effects of the devised S/S process and the substantial immobilization of metalloids and metals.

Concerning mechanical properties, the samples presented an average ACV=24±2% ($n$=6), which is between that of mixed gravel and clayey limestone (BS 1990). In addition, the CBR ranged between 37 and 49, stating that pellet specimens presented characteristics quite similar to materials currently used for medium traffic intensity road foundations (EN 2007).

The microstructure of samples with and without the SA addition was observed under a scanning electron microscope. As shown in Fig. 2, the granular material porosity is greatly lowered when the additive is taken into account (left picture), reducing grain-specific exchange surface area and increasing its compactness.

## 3.2 Toxicity data

Toxicity results are shown for both bioassays, as well as integrated toxicity judgements according to Wilke et al. (2008), in Table 5, where also pH values were displayed. In particular, all samples required pH adjustment via HCl 1 M aliquots addition to obtain the suitable pH range in order to carry out toxicity tests (7.45–8.30). Salinity and DO values remained on 33–34‰ and 6.0–8.0 mg $l^{-1}$ ranges, in that order. Negative and positive controls were in line with Azur Environmental (1998) and Libralato et al. (2007).

**Table 5** Ecotoxicological effects resulting from leachate samples prepared in artificial marine sea water (4)

| Samples | pH[a] | *C. gigas* | *V. bischeri*[b] | Toxicity data integration (Wilke et al. 2008) | | |
|---|---|---|---|---|---|---|
| | | TU50 | PE | *C. gigas* | *V. fischeri* | Σ/2 |
| P1 | 7.49 (9.56) | 1.23 (1.19–1.28) | 9.00 | 2 | 1 | 1.5 |
| P2 | 7.62 (10.84) | 1.18 (1.16–1.20) | 32.44 | 2 | 1 | 1.5 |
| P3 | 8.35 (10.17) | 1.57 (1.51–1.65) | 37.77 | 2 | 1 | 1.5 |
| P4 | 8.09 (10.22) | 1.16 (1.15–1.17) | 9.91 | 2 | 1 | 1.5 |
| P5 | 7.88 (10.46) | 1.16 (1.15–1.16) | 33.00 | 2 | 1 | 1.5 |
| P6 | 7.74 (10.63) | 1.16 (1.15–1.18) | 30.00 | 2 | 1 | 1.5 |

[a] Original pH is in brackets

[b] After 30 min contact time

*TU50* Toxic Units, *PE* percentage of effect

All samples showed similar toxicity levels within the same bioassay: between 1.16 and 1.57 TU50 for *C. gigas* and 9.00 PE and 37.77 PE for *V. fischeri*. Wilke et al. (2008) ranking indicated that all leachate samples could be deemed as minor acutely toxic.

## 4 Discussion

The results demonstrated that the proposed S/S soil remediation treatment provides several physicochemical, mechanical, and ecotoxicological advantages. The combination of sieved contaminated soil (Ø≤4 mm mesh), cement, and additives, properly mixed and converted to granular material, showed not only low leachability rates but also the ability to reduce the bioavailability of metallic pollutants from remediated soil samples. This was confirmed by the fact that leachates never exceeded regulatory limits for As, Cd, Hg, or Pb (EMD 1998, 1999). Moreover, toxicity bioassays, which can provide an integrated judgment of leachate samples, showed minor acutely toxic levels. Indeed, the leachate residual toxicity will be studied further, in order to ascertain if it could be due to the mixture effect of specific contaminants that have not been detected during this study or could be attributed to the cement or all other additives.

In particular, it has been evidenced that the additive (SA), specifically developed for this S/S process, which introduces the fundamental concepts of HPC to the S/S process, plays an essential role in reducing aggregates exchange surface area, boosting the performance of the S/S remediation technique.

Furthermore, the enhancement of soil mechanical properties compared to international standard values (BS 1990; EN 2002) suggested that soil granular material may be reused not only in situ but also for other applications, although a case-to-case basis chemical assessment would be required.

Although the cost of this application (around 80–120 € $m^{-3}$) is higher than the S/S in situ one (around 40–60 € $m^{-3}$), which presents several feasibility limitations as mentioned before, it is similar to other S/S ex situ applications (about 97–200 € $m^{-3}$), but with the advantage of increasing the contaminated soil remediation performance (FRTR 2008).

## 5 Conclusions

The application of the principles of high-performance concrete allowed a new stabilization/solidification technology to be developed for the remediation of heavy metals contaminated sites. The strong reduction of the mixing water (W/C<0.4) and the hydrophobic effect produced by the additive allow a dense, low porous, and mechanically resistant granular material to be obtained, which is further characterized by an extremely low permeability in comparison with traditional S/S materials. The strengthened mechanical properties widened the remediated soil reuse range, not only as in situ excavation filler but also as a resource for other ex situ purposes (e.g. medium traffic intensity road foundation or aggregate in concrete production). Besides, this technique reduces the amount of the resultant materials that require landfill disposal, and considering the obtained performance, the cost is very competitive.

## 6 Recommendations and perspectives

Regarding the long-term performance, there have been few investigations into the long-term effectiveness and durability of S/S contaminated soils (Harbottle et al. 2007), but some case studies investigated the long-term behavior of S/S-treated soils from contaminated sites after 3 to 5 years and noted no significant deterioration over this period (Board et al. 2000; Al-Tabbaa and Boes 2002). This offers some level of confidence in the medium-term behavior of S/S-treated soils. Some predictive modeling approaches can use the results of leaching tests, and the durability of S/S waste materials could be based on a measurement of the estimated long-term leaching of hazardous components from the S/S materials exposed to external influences (Felix et al. 2000). The projection of the leaching of the contaminated soil (of the case study area) stabilized using this S/S technique over a 100-year period has already been reported by Surico et al. (2003) and indicates that the cumulative leaching is far below the limits stated in the Dutch Building Material Decree (VROM 1999).

However, a long-term monitoring plan over several years is currently running in order to monitor the S/S treatment durability. Various monitoring piezometers installed in the remediated site will allow any change in leachate characteristics to be checked, tracing the effectiveness of the considered remediation procedure on a long-term basis. Further research will be conducted on all the component materials used during S/S treatment to assess their specific ecotoxicological effects.

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