Relating monolithic and granular leaching from contaminated soil treated with different cementitious binders

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Abstract

This work employed a clayey silty sandy gravel contaminated with a mixture of metals (Cd, Cu, Pb, Ni and Zn) and diesel. The contaminated soil was treated with 5 and 10% dosages of different cementitious binders. The binders include Portland cement, cement-fly ash, cement-slag and lime-slag mixtures. Monolithic leaching from the treated soils was evaluated over a 64-day period alongside granular leachability of 49 and 84-day old samples. Surface wash-off was the predominant leaching mechanism for monolithic samples. In this condition, with data from different binders and curing ages combined, granular leachability as a function of monolithic leaching generally followed degrees 4 and 6 polynomial functions. The only exception was for Cu, which followed the multistage dose-response model. The relationship between both leaching tests varied with the type of metal, curing age / residence time of monolithic samples in the leachant, and binder formulation. The results provide useful design information on the relationship between leachability of metals from monolithic forms of S/S treated soils and the ultimate leachability in the eventual breakdown of the stabilized/solidified soil.

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Abstract

This work employed a clayey silty sandy gravel contaminated with a mixture of metals (Cd, Cu, Pb, Ni and Zn) and diesel. The contaminated soil was treated with 5 and 10% dosages of different cementitious binders. The binders include Portland cement, cement-fly ash, cement-slag and lime-slag mixtures. Monolithic leaching from the treated soils was evaluated over a 64-day period alongside granular leachability of 49 and 84-day old samples. Surface wash-off was the predominant leaching mechanism for monolithic samples. In this condition, with data from different binders and curing ages combined, granular leachability as a function of monolithic leaching generally followed degrees 4 and 6 polynomial functions. The only exception was for Cu, which followed the multistage dose-response model. The relationship between both leaching tests varied with the type of metal, curing age / residence time of monolithic samples in the leachant, and binder formulation. The results provide useful design information on the relationship between leachability of metals from monolithic forms of S/S treated soils and the ultimate leachability in the eventual breakdown of the stabilized/solidified soil.

Keywords: blast furnace slag, fly ash, granular leaching, lime, monolithic leaching, Portland cement.

Introduction

Immobilisation of hazardous contaminants in soils through stabilisation/solidification (S/S) is an established technology for treatment of contaminated soils. S/S generally entails the addition of

cementitious binders to hazardous wastes and contaminated soils to physically encapsulate and chemically fix contaminants within the binder matrix. [1] The combined process of stabilisation and solidification usually results in a monolithic material with increased strength and decreased leachability, with the potential for eventual breakdown over a long period. Hence, contaminant leachability tests are normally performed on granular and/or monolithic samples of stabilized/solidified contaminated soils with a view to assess the long-term emission of contaminants from treated soils. The work of van der Sloot [2] provides details of several leaching test protocols. Nevertheless, very few attempts have been made to relate leaching results from different tests. A previous study in this direction is the work of Ogunro and Inyang [3], which related batch and column diffusion coefficients for leachable contaminants. The said work considered particulate waste materials (municipal solid waste incinerator-bottom ash amended with asphalt concrete) using Al and Cu diffusion coefficients as the target parameter. The study mainly observed no relationship between Al diffusion coefficients obtained through both leaching test methods. However, the study dealt with waste material other than contaminated soil.

Due to the paucity of literature in this area, little is known of how far leaching from stabilized/solidified monoliths is from granular forms of stabilized/solidified contaminated soils. Such knowledge is important, as it would provide useful design information on the relationship between the amounts of contaminants leached from monolithic forms of S/S treated soils and the ultimate leachability in the worst-case scenario that could occur over time. A number of degradation mechanisms of S/S treated materials, which could lead to the eventual breakdown of stabilized/solidified contaminated soils over time, have been identified. [4] There are two

leaching tests in common use, especially in the UK. These are the batch leaching test with de-ionised water extraction – BS EN 12457 [5] for granular samples, and the monolithic or tank leaching test – NEN 7375 [6] for monolithic samples. In the light of the above, it is desirable to establish relationships between leached contaminant concentrations from monolithic and granular forms of stabilized/solidified materials.

This work utilised data from extensive testing involving S/S treatment of contaminated soil with different binders. The binders, namely Portland cement, cement-fly ash, cement-slag and lime-slag mixtures, were deployed in studies aimed at developing operating envelopes for S/S treatment of contaminated soils. [7–11] It was the aim of this study to deduce relationships between leachability of metals in the granular and monolithic leaching tests using data generated from the above studies.

Materials and methods

The materials and methods used in this work have been described in detail in previous related publications. [9–11] Hence, only the most relevant details are summarised here.

Contaminated soil and binder

The studies employed a real site soil (65% gravel, 29% sand, 2.8% silt and 3.2% clay) from a Petrol station in Birmingham, UK. The soil is classified as clayey silty sandy gravel; it had a very low (0.22%, dry weight) organic carbon content. The soil was contaminated with very low

levels of metals and total petroleum hydrocarbons (TPH). Table 1 shows the total and leached concentrations of prime contaminants in the contaminated soil before spiking. In light of the above, the soil was spiked with 3000 mg/kg each of cadmium (using $Cd(NO_3)_2.4H_2O$), copper (using $CuSO_4.5H2O$), lead ($PbNO_3$), nickel ($Ni(NO_3)_2.6H_2O$) and zinc($ZnCl_2$), and 10,000 mg/kg of diesel. The same high concentration was used for all metals to increase the contaminant levels to relatively high values for monitoring compared to those typically found at contaminated sites. [12] The spiked contaminated soil had a highly alkaline pH of 9.83. The cause(s) of high soil alkalinity have been reviewed and such high alkalinity is mainly due to the association between sodium and carbonate species in the soil. [13] Further, one-third of the world's soils are alkaline. [14]

Four different binder formulations were used for S/S treatment of the contaminated soil. These include Portland cement (CEMI), and a mixture of CEMI and pulverised fuel ash (PFA) (CEMI:PFA = 1:4). Others are mixtures of CEMI and ground granulated blast furnace slag (GGBS) (CEMI:GGBS = 1:9), and hydrated lime (hlime) and GGBS (hlime:GGBS = 1:4). The contaminated soil was stabilized/solidified with 5 and 10% binder dosages (dry weight) of the aforementioned binders.

Preparation of stabilized/solidified products

Compaction using the 2.5 kg rammer was carried out on soil-binder mixtures in order to determine the dry density-moisture content relationship of the mixes. The stabilized/solidified products (50 mm diameter and 100 mm long) were prepared at the optimum moisture content

(OMC) and maximum dry density (MDD) for each soil-binder mixture. The OMC of the different soil-binder formulations ranged from 15 to 18%, while the MDD was in the range, 1.73 – 1.78 Mg/m^3. The moisture content determination excluded the moisture due to diesel content. [7, 9–10] The mixes were cured at 95% relative humidity and 20°C.

Testing of stabilized/solidified products

Monolithic leachability testing (also called the tank leaching test) was determined on 49-day mixes in accordance with NEN 7375. [6] The procedure used has been reported in a related paper on cement treatment of the same soil. [11] The ratio of the volume of leachant (de-ionised water) to the volume of the specimens (3 replicates) was kept constant at 3.5. The leachant was renewed at 8 time intervals of 0.25, 1, 2.25, 4, 9, 16, 36 and 64 days after commencement of the test. The leachate was analysed for only metal content, as the leached concentrations of hydrocarbons were negligible. Granular leachability testing was carried out using the Environment Canada acid neutralisation capacity test method, [15] albeit with zero acid addition to facilitate comparison with the monolithic leaching test. The particle size used was < 1.18 mm, the liquid-to-solid ratio was 10 and the contact time between samples and leachant (de-ionsed water) was 48 hours. Details of the test method can be found elsewhere. [9 – 10]

It should be noted that granular leachability testing was carried out on 49 and 84-day old mixes, to facilitate comparison with monolithic leaching test data. Especially, the cumulative leaching at 2.25 and 36 days, since the sample ages at those testing points were nearly the same. In other words, monolithic leaching test on 49-day old mixes submerged in the leachant for 2.25 days

141 nearly corresponds to granular leaching test on 49-day old mixes agitated in the leachant for 2
142 days. Similarly, leachate analysis of monolithic samples after 36 days (mixes then 85 days old)
143 nearly corresponds to granular leachability testing on 84-day old samples.
144
145 ***Statistics***
146
147 Regression modelling was carried out to find equations describing the relationship between
148 monolithic leaching and granular leaching using CurveExpert Professional 1.6.5 curve fitting
149 software. Monolithic leaching data was used as the predictor variable, while granular leaching
150 data was the response variable. The curve finder tool in CurveExpert was used to determine the
151 function that best fits the data among several inbuilt functions. Such functions include linear
152 regression, polynomial regression and non-linear regression models. The choice of the best fit
153 model was based on goodness of fit and applicability to representing granular leachability as a
154 function of monolithic leachability. MS Excel 2010 was then used to produce the resulting
155 graph(s) for consistency with other graphs in this work. Furthermore, one-way ANOVA was
156 used to test for statistically significant differences in metal leachability from monolithic forms of
157 the S/S treated soil due to the effect of sample curing age.
158
159 **Results and discussion**
160
161 It should be noted that some of the results presented here have been partly presented in previous
162 related publications. These include the monolithic leaching test results for the metals in CEMI-
163 treated soil presented in Kogbara et al. [11] The granular leachability results for the 5 metals at

zero acid addition in individual binders vis-a-vis CEMI-GGBS, hlime-GGBS and CEMI treated soil have also been published. [9–11] However, these are shown here in a different format and perspective to facilitate comparison between the different binders studied.

Monolithic leaching test results

The results of the cumulative measured and derived leaching in the four soil-binder systems considered are shown in Figures 1 – 5 for Cd, Cu, Pb, Ni and Zn, respectively. The data are for representative leachate samples from the entire leachant volume. Hence, there is no margin of error associated with the individual points. Generally, the leachate concentrations of the metals in all soil binder systems were very low even in 5% binder dosage mixes. The leachate concentrations were a little lower than those of Voglar and Lestan, [16] who used a higher (15% w/w) binder dosage, although the concentrations of some metals were far greater than the concentrations used here. The high alkalinity of the soil before stabilisation, coupled with the increase in pH compassed by addition of the binders, may be responsible for such low leachability. This is because the pH regime involved (see Table 2) corresponds to the region for minimum solubility of most of the metals considered. [9, 17]

The leachability trend was the same in all binders as higher concentrations of the metals were leached out in 5% than 10% dosage mixes. The only exception was the case of Pb, where the pH regime led to higher concentrations in 10% than 5% dosage mixes. However, the trend of Pb leachability was different in CEMI-GGBS mixes. The 5% dosage mix leached out higher concentrations of the metal as it is naturally expected (Fig. 3c). The effectiveness of the binder

for reduction of the granular leachability of Pb has been identified. [9, 18] This study extends the same to leachability in the monolithic leaching test. CEMI-PFA mixes also showed a similar trend. The 5% binder mix showed higher leachability than the 10% dosage mix during most of the sampling period. Nevertheless, the 5% dosage mix leached out marginally lower concentration of Pb than the 10% dosage mix after 64 days (Fig. 3b). The leaching behaviour of CEMI-GGBS mixes mentioned above differed from those in a similar study, [19] where there was no difference in contaminant leachability between ~14% and 22% (w/w) CEMI-GGBS dosages. However, the higher binder dosage leached out marginally higher amounts of Cd and Zn in the said study. This difference is mainly due to the difference in mix formulation of the binder as a far greater amount of GGBS was used here compared to their work.

The parameters used in determination of the leaching mechanisms involved in 5 and 10% dosage mixes of the soil-binder systems are shown in Tables 3 and 4, respectively. Generally, the slopes of the total increment (i.e. increments 2 - 7) were < 0.35 with a few exceptions. This indicates that surface wash-off of the metals otherwise physically encapsulated within the cementitious matrices was the predominant mechanism of contaminant release. This is similar to the findings of Voglar and Lestan. [16] All the same, there was evidence of diffusion-controlled contaminant release in some cases. Such instances include release of Cu in the 5% dosage CEMI mix (Table 3) and the 10% dosage mixes of CEMI-GGBS and hlime-GGBS (Table 4). Others are Ni release in the 5% dosage hlime-GGBS mix and the 10% CEMI-GGBS dosage mix (Table 4); and Pb release in the 10% dosage hlime-GGBS mix. Moreover, even though the total increment was not indicative of diffusion-controlled leaching, there were increments where the slopes and standard deviations suggested diffusion-controlled leaching (Tables 3 and 4).

Furthermore, the slopes of some increments indicate the possibility of dissolution of the components (Tables 3 and 4). This appears contrary to the finding that the specimens did not dissolve during the test. In such cases, viewed from the leaching mechanism of the matrix, the dissolution of the components has no permanent character. It is possible that dissolution was only occurring from the outer layer of the test piece. [6] Negligible amounts of hydrocarbons leached out from all binders. However, it is noteworthy that unlike other binders, there were traces of diesel film in leachates of CEMI-PFA mixes over time. This was more pronounced in the 5% dosage mix. This implies that of the four binders used, CEMI-PFA was the least effective in reducing the leachability of hydrocarbons in contaminated soil.

Relationship between monolithic and granular leachability

The granular leachability results presented in this section have been presented as part of leaching data in acid neutralisation capacity tests with zero acid addition in a different format and perspective for each of the individual binders in related publications. [9 – 11] In other words, metal leachability data in the above studies were presented against pH at 0, 1 and 2 meq/g HNO_3 addition. Hence, granular leachability data are only presented here in graphs relating them to monolithic leaching data (Fig. 6 – 8) in line with the object of this study. Figures 6 – 8 show the leachability of 49 and 84 day old granular samples agitated in deionised water leachant for 2 days versus the corresponding cumulative leaching of the same 49 day old (monolithic) samples above subjected to tank leaching test for 2.25 and 36 days. In the said graphs, the derived cumulative emissions from monolithic leaching tests were converted from mg/m^2 to mg/kg,

taking into account the total weight of the samples equivalent to the surface area. This was done in order to facilitate comparison with the granular leachability values.

In Figures 6 – 8, there are two graphs for each metal. The first graph for a given metal shows the contribution of each of the four different binder formulations to the leaching trend observed. It also shows the leachability at the two different curing ages considered. While the second graph shows the regression model identifying the relationship between granular and monolithic leaching, with data from different binders and curing ages combined. The following summarises the major findings from the relationship between the monolithic and granular leachability.

i. In most cases, the trend in the relationship between both leaching tests at 84 days was clearer than that observed for 49 day old samples (Fig. 6 – 8 a and b). Hence, the 84-day data would make more sense when comparing leaching trends. However, the difference in leachability of the metals in the monolithic leaching test due to differences in curing age was not statistically significant. The only exception was for Cu leachability, which was highly significant at the 0.001% probability level.

ii. It is thought that the above is linked to the fact that the 49-day data came from monolithic samples that were just 2.25 days old in the leachant. Hence, there has not been sufficient ingress of the leachant into the monolith causing release of reasonable contaminant concentrations compared to 36 days residence time in the leachant for the 84-day data.

iii. Looking at the relationship between both leaching tests for the different metals among different binder formulations, there was no clear general trend for Cd and Cu leachability as it differed among the different binders at 49 and 84 days [Fig. 6 and 7 (a and b), and

Fig. 8a]. With the exception of CEMI-PFA mixes, the leaching trend for Cd recorded at 84 days was opposite the trend observed for 49-day samples (Fig. 6a). Taken all mixes together, there is an apparent inverse relationship for Cu at 84 days (Fig. 6b).

iv. Similarly, there was an inverse relationship between both leaching tests for Ni and Zn at 84 days although CEMI mixes were an exception to this for Ni as there was no difference in granular leaching due to a slight increase in monolithic leaching (Fig. 7b and 8a). However, there was a direct relationship between leaching from monolithic and granular samples for Pb at 84 days (Fig. 7a).

v. In practice, an inverse relationship between both leaching tests implies that granular leachability did not increase with increase in cumulative metal emission from monolithic samples. A direct relationship shows increase in granular leachability with increase in cumulative derived metal emission.

vi. The relationship between monolithic and granular leachability was complicated, generally following higher order polynomial regression models. Specifically, degree 6 polynomials were the best-fit model relating granular and monolithic leaching of Cd and Pb (Fig. 6c and 7c). While the best fit for Ni and Zn were degree 4 polynomials (Fig. 7d and 8b). The dose-response multi-stage 4 model was the best fit for Cu (Fig. 6d). The best-fit model for Cu probably followed a different regression family due to significant differences observed in Cu leachability at both curing ages considered.

vii. The above is supported by a similar observation in a previous related study. Polynomial approximation was found to be the most convenient for modelling cumulative amount of radionuclides, ^{137}Cs and ^{60}Co, leached from a cement composite matrix. [20] Polynomial fits have also been used in describing dynamic leaching of metal contaminants from

279 solidified wastes [21] and Pb solubilisation with pH in cement-stabilized synthetic waste.
280 [22]

281 viii. As with most polynomial models, an implication of polynomial fit to the leaching data is
282 that extrapolation to values outside the range of measurement is not advisable. This is
283 because the optimized constants in the equations in Figures 6, 7 and 8 do not necessarily
284 have any physical significance.

285

286 Furthermore, the finding that there was no defined relationship between both leaching tests for
287 Cd is similar to the observation of Ogunruo and Inyang. [3] They observed that there was no
288 defined relationship between Al diffusion coefficients obtained through batch and column
289 leaching tests. The observed inverse relationship for Ni and Zn among different binders stems
290 from the fact that in the granular leaching test, the pH of the mixes fell within the pH zone (ca.
291 10.8 – 12.8) where leachability of the metals increases with pH. [9 – 11] Higher binder dosage led
292 to higher pH (compare data in Table 2) and hence increases in granular leachability of the
293 metals, whereas in the monolithic leaching test higher binder dosage led to decrease in metal
294 leachability. Conversely, in the case of Pb, although its leachability increases with pH around the
295 afore-stated pH range, there was a direct relationship. The previously mentioned unique leaching
296 behavior of Pb in monolithic samples, where higher binder dosage led to higher cumulative
297 metal emission is partly responsible for the behavior. The above, coupled with the effectiveness
298 of CEMI-GGBS and CEMI-PFA in immobilising the metal, led to a situation in which increase
299 in cumulative metal emission from monoliths corresponded to increase in granular leachability.

300

301 The relationships between leaching from both forms of the treated material varied with type of
302 metal, curing age / residence time of monolithic samples in the leachant and binder formulation.
303 It is suspected that the variations are due to the different mechanisms involved in both leaching
304 tests. The variations could also be caused by differences in leaching mechanisms across different
305 increments in the monolithic leaching test (see Tables 3 and 4). In support of the above, in
306 majority of the monoliths, surface wash-off of contaminants was the dominant leaching
307 mechanism. However, there were cases of diffusion-controlled leaching in some increments.
308 Whereas, advection (i.e. water percolating through or along the material) is the dominant
309 leaching mechanism for granular materials. [23]
310
311 **Conclusions**
312
313 Leaching of five of the most common metallic contaminants found in soils from monolithic
314 samples of contaminated soil treated with different binders consisting of mixtures of cement, fly
315 ash, blast furnace slag and lime was considered in this study. These were later related to leaching
316 from granular forms of the treated materials. The results showed that the leachate concentrations
317 of the metals in all soil-binder systems in the monolithic leaching test were very low even in 5%
318 binder dosage mixes. Generally, the predominant mechanism of release in all soil-binder systems
319 was surface wash-off of contaminants, although diffusion-controlled leaching was observed in
320 some cases. CEMI-GGBS mixes demonstrated a unique effectiveness for Pb. Mixes with the
321 other binders leached out higher concentrations of Pb with 10% binder dosage than with 5%
322 dosage in line with the amphoteric behaviour of the metal but CEMI-GGBS mixes did not.
323

The results showed that the relationship between both leaching tests was more meaningful when monolithic samples in the tank-leaching test have spent more residence time in the leachant. The relationship between both leaching tests for the different metals among different binder formulations was not straightforward. It varied with the type of metal, curing age / residence time of monolithic samples in the leachant and the binder formulation. With surface wash-off as the predominant leaching mechanism for monolithic samples, granular leachability as a function of monolithic leaching, with data from different binders and curing ages combined, generally followed degrees 4 and 6 polynomial functions. The only exception was for Cu leachability, which followed the multistage dose-response model. These results show that the relationship between both leaching tests is complicated and difficult to interpret. Nevertheless, they provide useful design information on the relationship between leachability of metals from monolithic forms of S/S treated soils and the ultimate leachability in the eventual breakdown of the stabilized/solidified soil. The results would be helpful in estimating granular leachability of metals in near neutral-pH environments from monolithic leaching test data. Especially, when used together with future in-depth studies on the subject matter involving more binder types and dosages, curing ages, etc. This work has already set the stage for such future studies.

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437
438

FIGURE CAPTIONS

Figure 1. Cumulative measured and derived leaching of Cd in contaminated soil treated with (a) CEMI, (b) CEMI-PFA, (c) CEMI-GGBS and (d) hlime-GGBS binders.

Figure 2. Cumulative measured and derived leaching of Cu in contaminated soil treated with (a) CEMI, (b) CEMI-PFA, (c) CEMI-GGBS and (d) hlime-GGBS binders.

Figure 3. Cumulative measured and derived leaching of Pb in contaminated soil treated with (a) CEMI, (b) CEMI-PFA, (c) CEMI-GGBS and (d) hlime-GGBS binders.

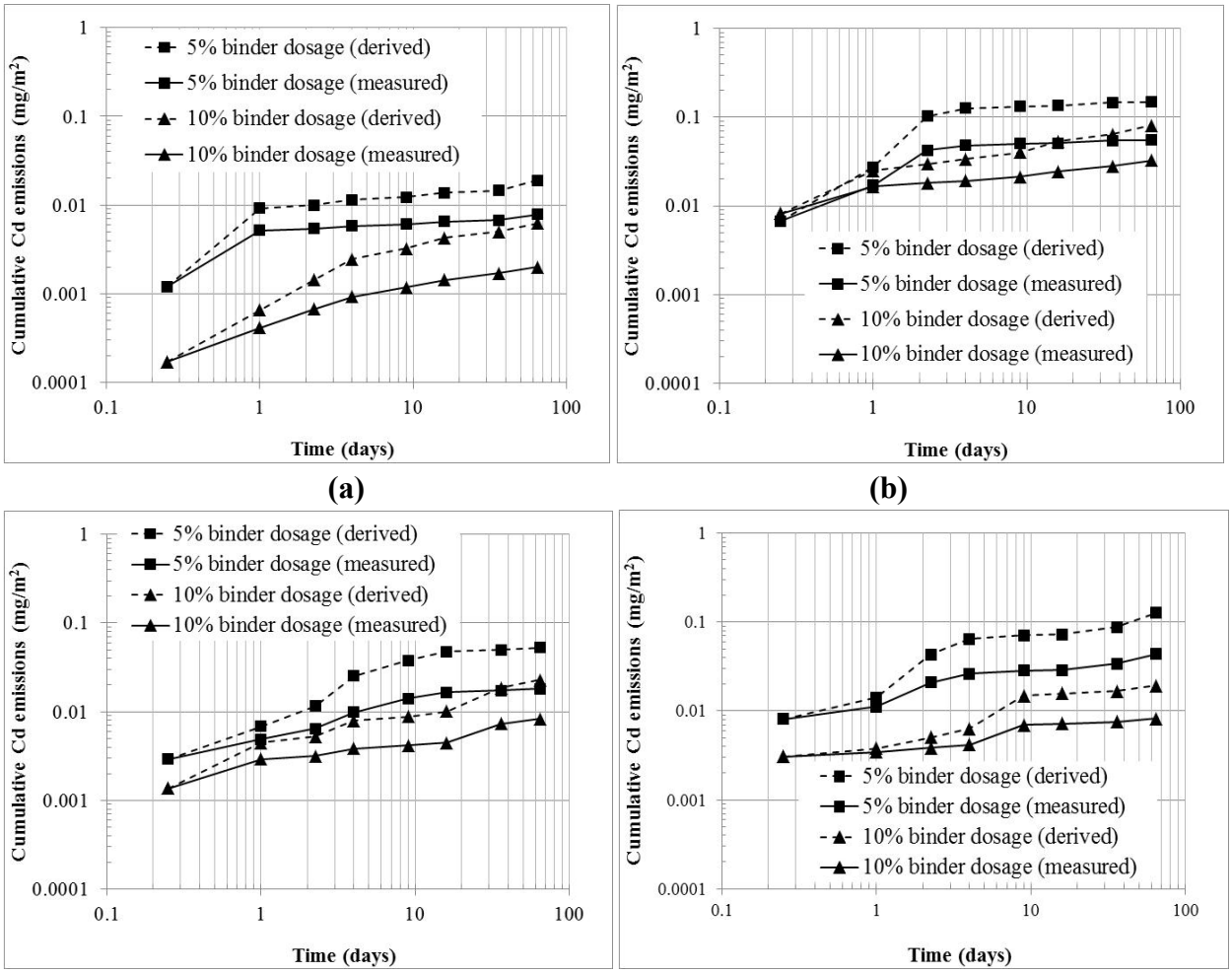
Figure 4. Cumulative measured and derived leaching of Ni in contaminated soil treated with (a) CEMI, (b) CEMI-PFA, (c) CEMI-GGBS and (d) hlime-GGBS binders.

Figure 5. Cumulative measured and derived leaching of Zn in contaminated soil treated with (a) CEMI, (b) CEMI-PFA, (c) CEMI-GGBS and (d) hlime-GGBS binders.

Figure 6. (a) & (b) Relationship between monolithic and granular leaching for Cd and Cu; (c) & (d) regression models relating both leaching tests for Cd and Cu, respectively.

Figure 7. (a) & (b) Relationship between monolithic and granular leaching for Pb and Ni; (c) & (d) regression models relating both leaching tests for Pb and Ni, respectively.

Figure 8. (a) Relationship between monolithic and granular leaching for Zn and (b) polynomial regression model relating both leaching tests for Zn.


5% binder dosage (derived)
5% binder dosage (measured)
10% binder dosage (derived)
10% binder dosage (measured)
Cumulative Cd emissions (mg/m²)
Time (days)
1
0.1
0.01
0.001
0.0001
0.1
1
10
100

(a) **(b)**

(c) **(d)**

Fig. 1

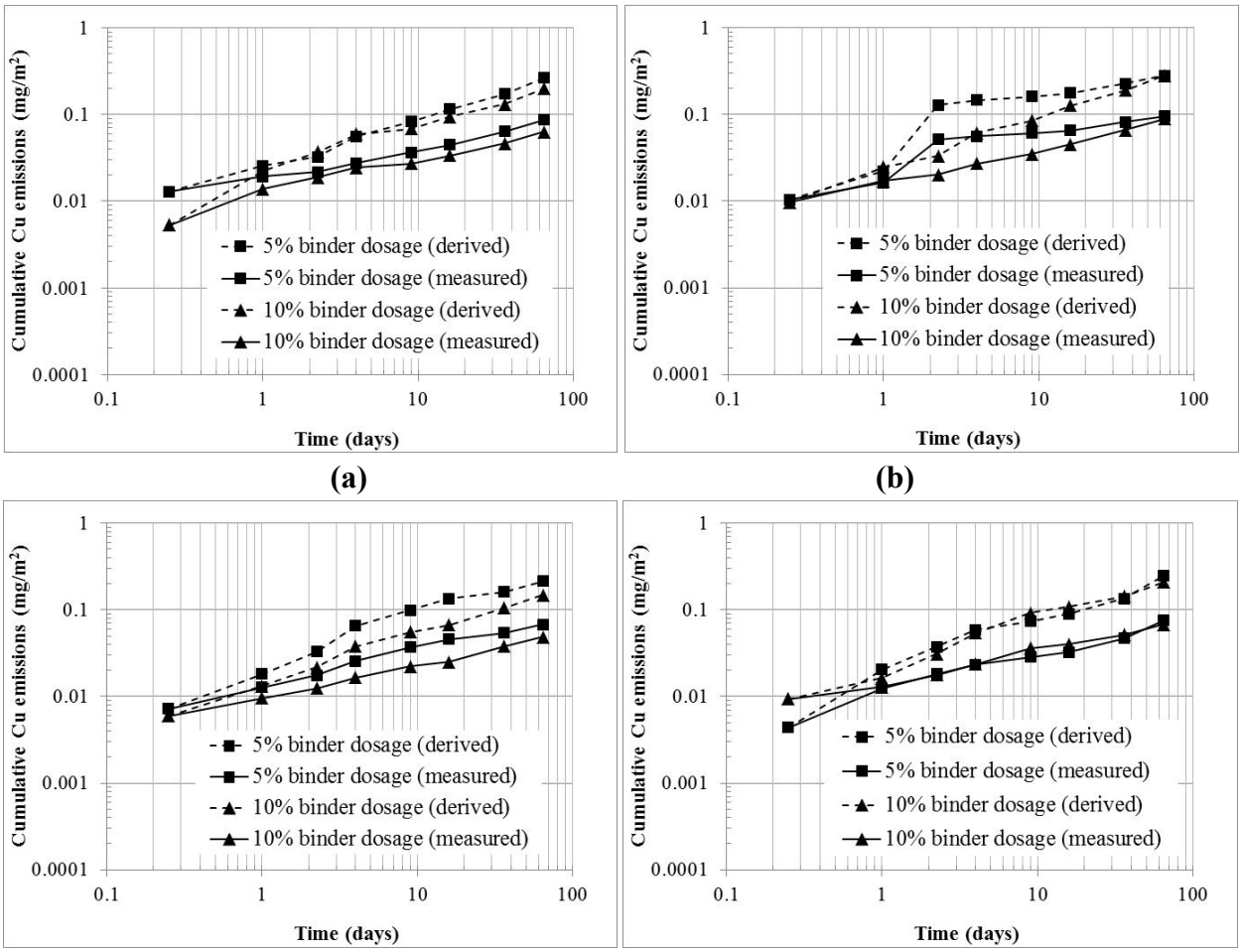

Cumulative Cu emissions (mg/m²)
Time (days)
5% binder dosage (derived)
5% binder dosage (measured)
10% binder dosage (derived)
10% binder dosage (measured)
(a)
(b)
(c)
(d)

Fig. 2

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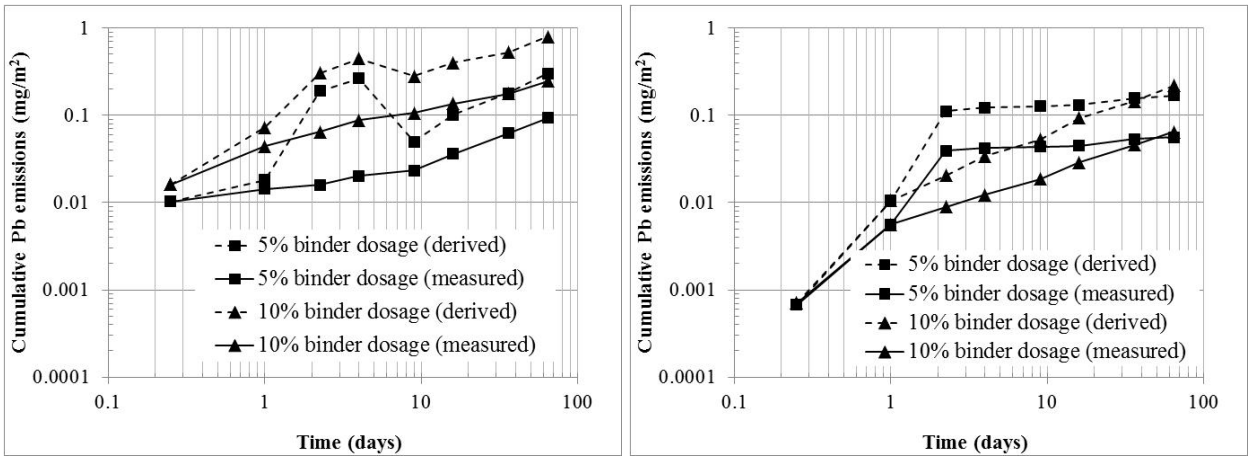

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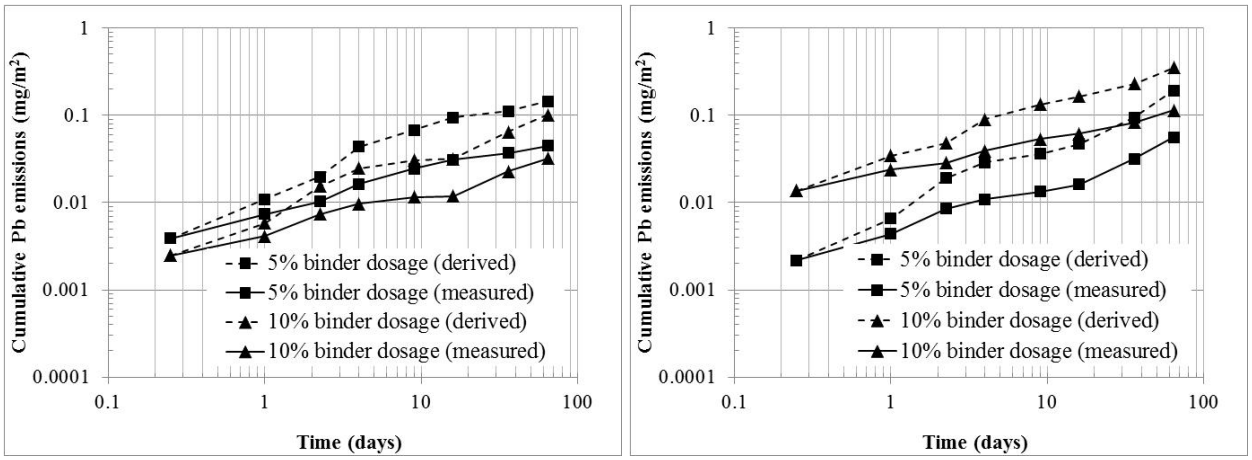

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493 **Fig. 3**
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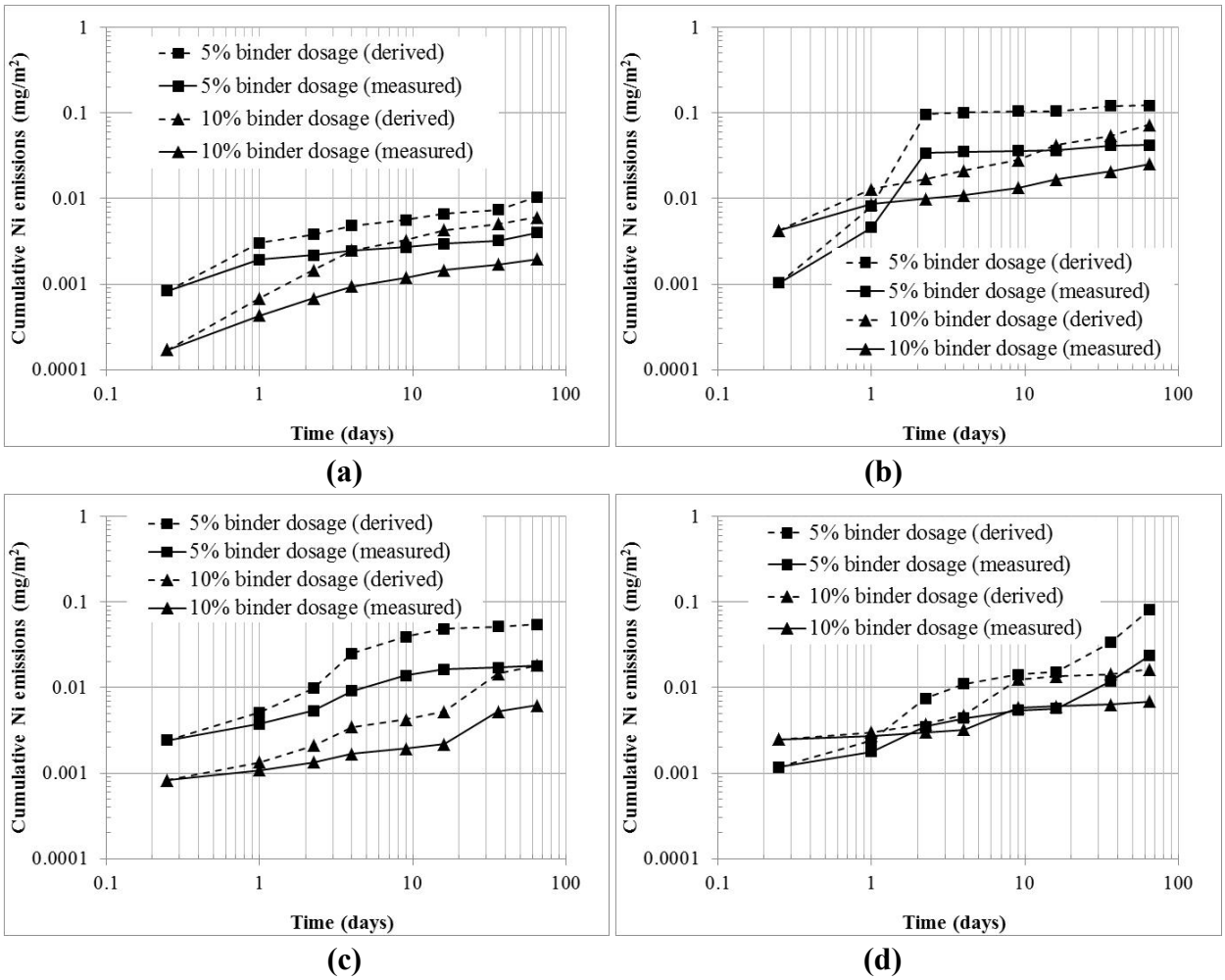

5% binder dosage (derived)
5% binder dosage (measured)
10% binder dosage (derived)
10% binder dosage (measured)
Cumulative Ni emissions (mg/m²)
Time (days)
(a)
(b)
(c)
(d)

Fig. 4

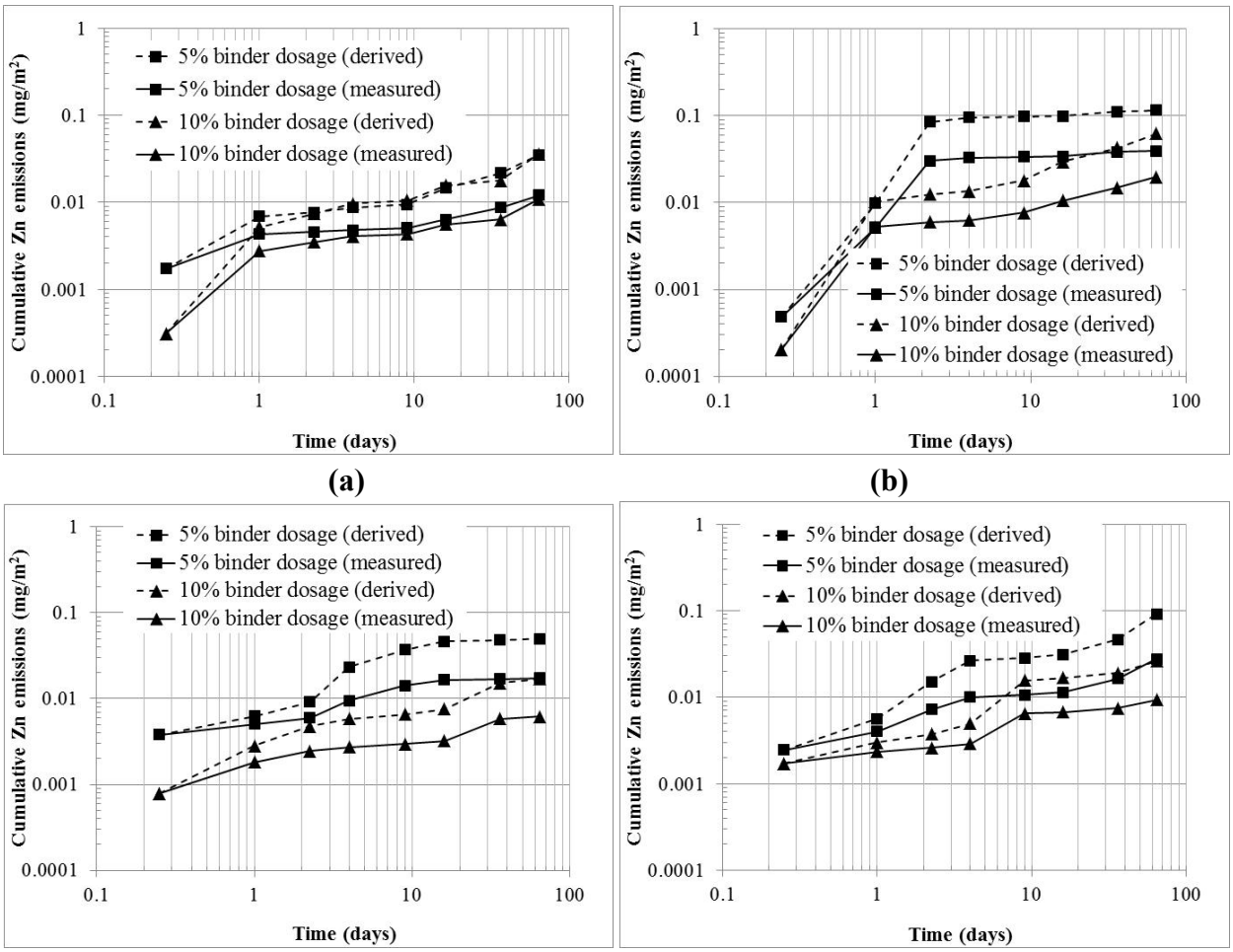

5% binder dosage (derived)
5% binder dosage (measured)
10% binder dosage (derived)
10% binder dosage (measured)
Cumulative Zn emissions (mg/m²)
Time (days)
1
0.1
0.01
0.001
0.0001
0.1
1
10
100
(a)
(b)
(c)
(d)

Fig. 5

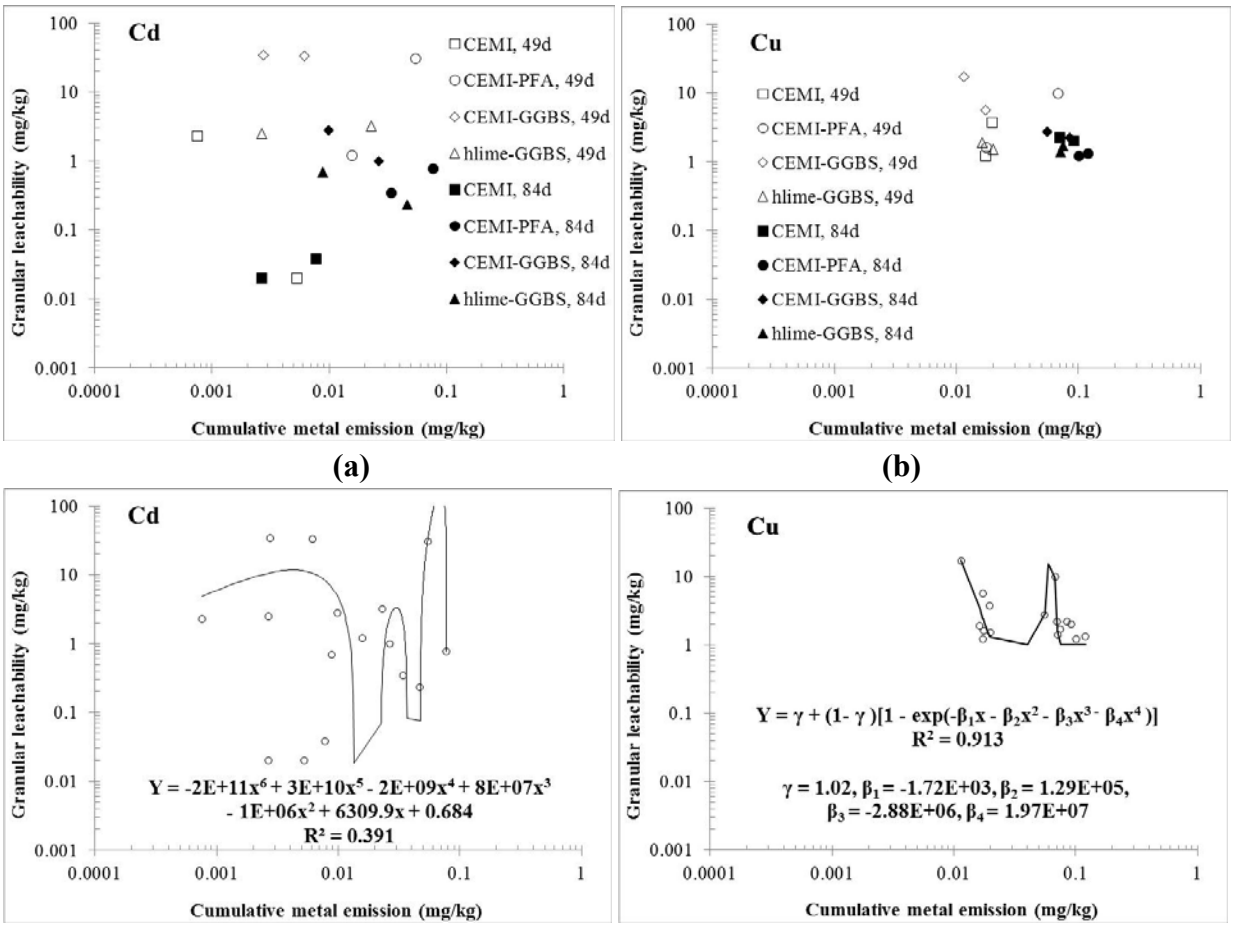

Cd
Granular leachability (mg/kg)
Cumulative metal emission (mg/kg)
CEMI, 49d
CEMI-PFA, 49d
CEMI-GGBS, 49d
hlime-GGBS, 49d
CEMI, 84d
CEMI-PFA, 84d
CEMI-GGBS, 84d
hlime-GGBS, 84d
(a)
Cu
(b)
Y = -2E+11x⁶ + 3E+10x⁵ - 2E+09x⁴ + 8E+07x³ - 1E+06x² + 6309.9x + 0.684
R² = 0.391
(c)
Y = γ + (1- γ)[1 - exp(-β1x - β2x² - β3x³ - β4x⁴)]
R² = 0.913
γ = 1.02, β1 = -1.72E+03, β2 = 1.29E+05, β3 = -2.88E+06, β4 = 1.97E+07
(d)

Fig. 6

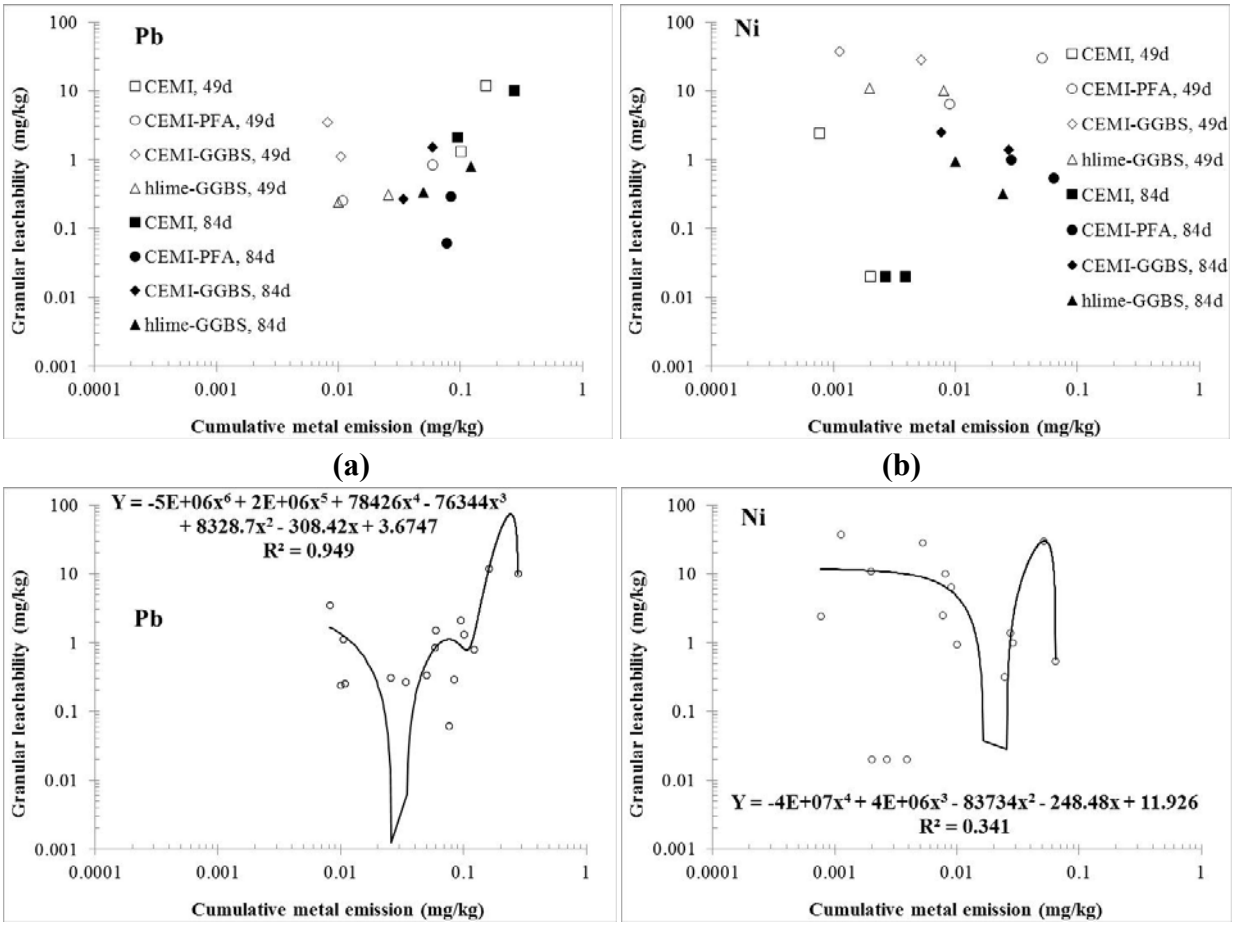
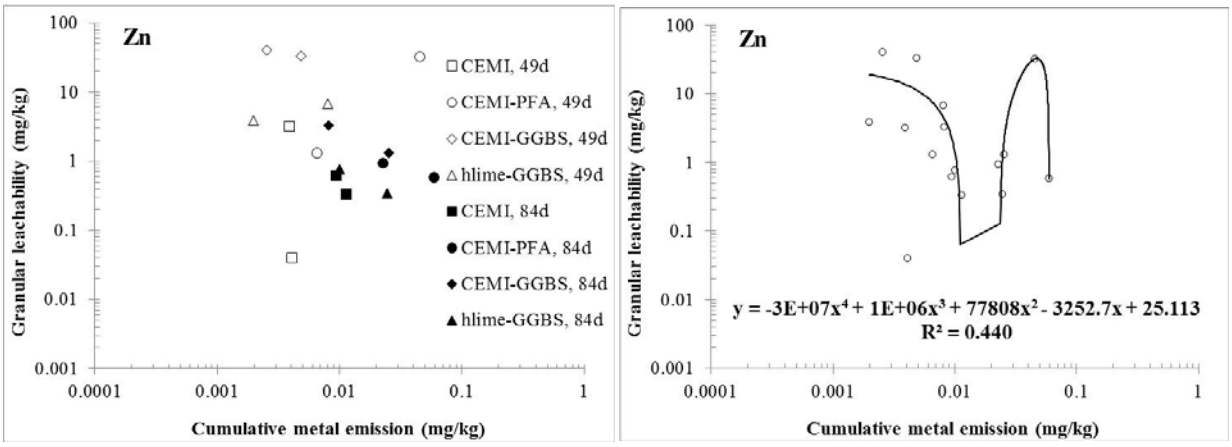

Pb
Granular leachability (mg/kg)
Cumulative metal emission (mg/kg)
CEMI, 49d
CEMI-PFA, 49d
CEMI-GGBS, 49d
hlime-GGBS, 49d
CEMI, 84d
CEMI-PFA, 84d
CEMI-GGBS, 84d
hlime-GGBS, 84d
(a)
Ni
(b)
Y = -5E+06x⁶ + 2E+06x⁵ + 78426x⁴ - 76344x³ + 8328.7x² - 308.42x + 3.6747
R² = 0.949
(c)
Y = -4E+07x⁴ + 4E+06x³ - 83734x² - 248.48x + 11.926
R² = 0.341
(d)

Fig. 7


Zn
Granular leachability (mg/kg)
Cumulative metal emission (mg/kg)
CEMI, 49d
CEMI-PFA, 49d
CEMI-GGBS, 49d
hlime-GGBS, 49d
CEMI, 84d
CEMI-PFA, 84d
CEMI-GGBS, 84d
hlime-GGBS, 84d
y = -3E+07x⁴ + 1E+06x³ + 77808x² - 3252.7x + 25.113
R² = 0.440

(a) **(b)**

Fig. 8

586 **Table 1. Concentrations of prime contaminants in the soil before spiking**

Contaminant	Total concentration (mg/kg)	Leached concentration (mg/kg)
Cadmium	0.3	< 0.1
Copper	28	0.4
Lead	401	< 0.1
Nickel	18	0.1
Zinc	179	0.1
TPH	43	< 10

587 TPH: Total hydrocarbon content

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591 **Table 2. Leachate pH data of the soil binder systems**

Approx. curing age (days)	Binder	Leachate pH in monolithic leaching test		Leachate pH in granular leaching test	
		5% dosage	10% dosage	5% dosage	10% dosage
49	CEMI	11.71	11.98	11.87	12.25
	CEMI-PFA	11.40	11.54	10.45	11.42
	CEMI-GGBS	11.64	11.76	11.25	11.57
	hlime-GGBS	11.51	11.79	11.12	11.63
84	CEMI	11.82	12.04	12.06	12.38
	CEMI-PFA	11.36	11.48	10.37	10.92
	CEMI-GGBS	11.79	11.85	11.20	11.51
	hlime-GGBS	11.78	11.92	11.05	11.83

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Table 3. Determination of the leaching mechanisms involved in 5% binder dosage mixes during the monolithic leaching test

Increment*	Mix detail	Cadmium		Copper		Lead		Nickel		Zinc		Significance of Slopes (rc) of increments		
a – b		rc	Sd_{rc}	rc	Sd_{rc}	rc	Sd_{rc}	rc	Sd_{rc}	rc	Sd_{rc}	≤ 0.35	0.35 < rc ≤ 0.65	> 0.65
2 - 7	CEMI	-0.43	0.34	0.49	0.17	0.70	0.28	-0.20	0.15	0.26	0.48	Surface wash-off	Diffusion	Dissolution
	CEMI-PFA	-0.59	0.49	0.07	0.42	-0.20	0.56	-0.41	0.74	-0.51	0.68			
	CEMI-GGBS	-0.02	0.34	0.28	0.16	0.31	0.19	0.07	0.38	0.04	0.47			
	hlime-GGBS	-0.21	0.54	0.19	0.15	0.33	0.25	0.37	0.46	0.11	0.40			
5 - 8	CEMI	0.61	0.32	0.63	0.05	1.20	0.23	0.54	0.24	1.30	0.26	Depletion	Diffusion	Dissolution
	CEMI-PFA	-0.62	0.62	0.80	0.10	0.64	0.34	0.11	0.74	0.55	0.58			
	CEMI-GGBS	-0.85	0.18	0.14	0.14	0.05	0.14	-0.84	0.18	-1.19	0.21			
	hlime-GGBS	1.23	0.53	1.04	0.14	0.59	0.10	1.71	0.49	1.66	0.14			
4 - 7	CEMI	-0.20	0.19	0.41	0.06	0.82	0.32	-0.09	0.08	1.02	0.31	Depletion	Diffusion	Dissolution
	CEMI-PFA	-0.42	0.54	0.45	0.21	0.34	0.42	0.31	0.67	-0.05	0.71			
	CEMI-GGBS	-0.75	0.20	-0.09	0.06	-0.13	0.08	-0.79	0.19	-0.98	0.27			
	hlime-GGBS	-0.32	0.66	0.32	0.20	0.15	0.27	0.57	0.58	0.16	0.52			
3 - 6	CEMI	0.20	0.19	0.70	0.18	0.90	0.33	0.07	0.08	0.77	0.33	Depletion	Diffusion	Dissolution
	CEMI-PFA	-1.86	0.04	-0.85	0.32	-1.55	0.32	-2.20	0.36	-2.25	0.16			
	CEMI-GGBS	0.30	0.21	0.39	0.13	0.50	0.16	0.30	0.25	0.51	0.32			
	hlime-GGBS	-1.58	0.21	-0.07	0.06	0.37	0.11	-0.74	0.17	-0.83	0.25			
2 - 5	CEMI	-0.92	0.37	0.46	0.24	0.20	0.26	-0.41	0.17	-0.78	0.30	Depletion	Diffusion	Dissolution
	CEMI-PFA	-0.64	0.44	-0.14	0.54	-0.50	0.64	-0.67	0.72	-0.73	0.63			
	CEMI-GGBS	0.61	0.16	0.57	0.11	0.63	0.15	0.84	0.18	0.92	0.24			
	hlime-GGBS	0.00	0.43	0.00	0.07	0.43	0.22	0.36	0.27	-0.18	0.44			
1 - 4	CEMI	-0.09	0.54	0.07	0.26	0.06	0.27	0.01	0.25	-0.30	0.40	Surface wash-off	Diffusion	Delayed diffusion or dissolution
	CEMI-PFA	0.63	0.34	0.48	0.48	1.33	0.68	0.97	0.75	1.41	0.64			
	CEMI-GGBS	0.48	0.19	0.49	0.13	0.58	0.15	0.59	0.24	0.34	0.37			
	hlime-GGBS	0.46	0.27	0.56	0.13	0.29	0.14	0.51	0.23	0.59	0.15			

rc: slope of the relevant increment; * These are data points on Figures 1 – 5; Sd_{rc}: standard deviation of the slope of the relevant increment

Criteria for diffusion controlled leaching in increment a-b: $CF_{a-b} \geq 1.5$, $Sd_{rc} \leq 0.5$, $0.35 < rc \leq 0.65$

CF_{a-b}: concentration factor in increment a-b, it was > 1.5 in all cases, hence it is not shown here

Table 4. Determination of the leaching mechanisms involved in 10% binder dosage mixes during the monolithic leaching test

Increment*	Mix detail	Cadmium		Copper		Lead		Nickel		Zinc		Significance of Slopes (rc) of increments		
a – b		rc	Sd_{rc}	rc	Sd_{rc}	rc	Sd_{rc}	rc	Sd_{rc}	rc	Sd_{rc}	≤ 0.35	0.35 < rc ≤ 0.65	> 0.65
2 - 7	CEMI	0.11	0.11	0.19	0.21	0.21	0.13	0.10	0.10	-0.12	0.33	Surface wash-off	Diffusion	Dissolution
	CEMI-PFA	0.05	0.28	0.46	0.17	0.52	0.10	0.23	0.20	0.31	0.44			
	CEMI-GGBS	0.21	0.42	0.38	0.16	0.23	0.51	0.60	0.32	0.19	0.39			
	hlime-GGBS	0.11	0.42	0.36	0.20	0.46	0.17	0.22	0.44	0.25	0.44			
5 - 8	CEMI	0.15	0.10	0.95	0.12	0.70	0.13	0.07	0.08	1.22	0.43	Depletion	Diffusion	Dissolution
	CEMI-PFA	0.42	0.13	0.66	0.04	0.64	0.09	0.39	0.11	0.67	0.13			
	CEMI-GGBS	1.01	0.30	0.62	0.19	1.38	0.55	1.07	0.37	0.66	0.45			
	hlime-GGBS	-0.54	0.49	0.37	0.26	1.40	0.59	-0.60	0.46	-0.03	0.57			
4 - 7	CEMI	-0.09	0.08	0.35	0.28	0.21	0.18	-0.09	0.08	0.20	0.40	Depletion	Diffusion	Dissolution
	CEMI-PFA	0.50	0.14	0.40	0.12	0.65	0.09	0.51	0.13	1.14	0.20			
	CEMI-GGBS	0.53	0.46	0.32	0.23	0.38	0.68	0.86	0.42	0.89	0.36			
	hlime-GGBS	-0.39	0.52	0.05	0.20	0.72	0.14	-0.33	0.54	0.02	0.57			
3 - 6	CEMI	0.07	0.08	0.08	0.25	0.20	0.18	0.07	0.08	0.17	0.41	Depletion	Diffusion	Dissolution
	CEMI-PFA	0.51	0.14	0.64	0.19	0.67	0.09	0.64	0.09	0.97	0.27			
	CEMI-GGBS	0.02	0.30	0.13	0.17	-0.94	0.23	0.03	0.14	-0.33	0.14			
	hlime-GGBS	0.25	0.51	0.14	0.22	-0.12	0.23	0.49	0.52	0.52	0.60			
2 - 5	CEMI	0.23	0.12	-0.23	0.17	0.03	0.13	0.20	0.11	-0.78	0.12	Depletion	Diffusion	Dissolution
	CEMI-PFA	-0.45	0.25	0.32	0.23	0.31	0.06	-0.08	0.20	-0.42	0.45			
	CEMI-GGBS	-0.39	0.34	0.45	0.09	0.23	0.24	0.23	0.17	-0.49	0.09			
	hlime-GGBS	1.04	0.24	0.78	0.04	0.19	0.21	1.19	0.26	0.95	0.42			
1 - 4	CEMI	0.65	0.04	0.48	0.13	0.60	0.10	0.65	0.05	0.70	0.44	Surface wash-off	Diffusion	Delayed diffusion or dissolution
	CEMI-PFA	-0.29	0.28	0.26	0.23	1.06	0.29	-0.04	0.20	0.62	0.76			
	CEMI-GGBS	0.07	0.34	0.32	0.12	0.54	0.14	0.15	0.19	0.15	0.23			
	hlime-GGBS	-0.29	0.25	0.33	0.19	0.62	0.21	-0.33	0.29	-0.20	0.12			

rc: slope of the relevant increment; * These are data points on Figures 1 – 5; Sd_{rc}: standard deviation of the slope of the relevant increment

Criteria for diffusion controlled leaching in increment a-b: $CF_{a-b} \geq 1.5$, $Sd_{rc} \leq 0.5$, $0.35 < rc \leq 0.65$

CF_{a-b}: concentration factor in increment a-b, it was > 1.5 in all cases, hence it is not shown here