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The use of cardboard factory sludge in the remediation of zinc-contaminated sediment

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Abstract: Paper and cardboard factory sludges are generated by various processes during pulp, cardboard and paper production, and the increasing quantities produced make the disposal of this sludge a problem. This study investigates the use of cardboard factory sludge as a stabilizing agent in the solidification/stabilization (S/S) treatment of zinc polluted sediment. Semi-dynamic and toxicity leaching tests were conducted to assess the effectiveness of the solidification/stabilization treatment and long-term zinc leaching behaviour. A diffusion-based model was used to elucidate the controlling leaching mechanisms. The applied S/S treatment was effective in immobilizing zinc, and the controlling leaching mechanism appeared to be diffusion, which indicates that a slow leaching of zinc could be expected when cardboard mill sludge is applied as a S/S agent.

Keywords: solidification/stabilization; leaching mechanism; leaching test.

INTRODUCTION

Zinc from various sources (industrial, mining, municipal sewage, agricultural and other activities) has entered water ways over time. Depending on their physico-chemical behaviour, heavy metals may adsorb onto suspended solids and subsequently accumulate in sediments, where they can pose a significant contamination problem.¹ However, heavy metals are not bound to sediments forever. With variations in the physical–chemical characteristics of the water conditions, some of the metals will re-enter the supernatant water and become available to living organisms.^{2,3} Symptoms of zinc toxicity in human beings include vom-

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ting, dehydration, electrolyte imbalance, abdominal pain, nausea, lethargy, dizziness and lack of muscular coordination. Zinc imparts an undesirable, bitter astringent taste to water in elevated concentrations. Concentrations of zinc above the recommended maximum value also cause adverse effects on the morphology of fish by inducing cellular breakdown of the gills.^{1,4}

It is now widely accepted that the toxicity and the mobility of elements depend strongly on their distribution in sediments. Hence, identification of the main binding sites and phase associations of heavy metals in sediments, based on the results of sequential extraction procedures, can help in understanding the remobilisation potential and the risks induced.⁵ Based on the Tessier procedure,⁶ the European Community Bureau of Reference produced a definition of an extraction protocol (the BCR protocol) and a purely operational definition of sequential fractionation. This method partitions the heavy metals into four fractions: exchangeable and carbonate bound, iron and manganese oxides bound, organic matter bound and residual metal. The distribution of metals in different phases obtained by the sequential extraction procedure offers an indication of their availability, which in turn allows a risk assessment of their presence in aquatic environments. The Risk Assessment Code (RAC) gives an idea of the possible risk by applying a scale to the percentage of metals present in the exchangeable and carbonate fractions, *i.e.*, labile phases.⁷ According to the RAC, if this fraction is < 1 %, there is no risk for the aquatic system, 1–10 % exhibits a low risk, 11–30 % medium risk, 31–50 % high risk and > 75% a very high risk.

In some cases, due to different factors, sediments may need dredging and remediation treatments. Currently, dredged sediment in Serbia is deposited in landfills whether it is contaminated or not. However, landfill capacity is limited, and can cause a serious threat if it is contaminated. In this study, stabilization/solidification (S/S) was used in order to remediate zinc-contaminated sediment, since this is a widely accepted treatment process for the immobilization of heavy metals contained in wastes.^{8,9} At the moment, there is not enough data about S/S treatments of contaminated sediments, especially those contaminated with zinc. There are a few studies regarding soil treatment with paper mill sludge but none for sediment treatment, and the conclusions obtained for soils that can be found in the literature may not be completely valid for sediments.

Paper and cardboard factory sludges are generated by various processes in the production of pulp, cardboard and paper, and the increasing quantities produced make the disposal of this sludge a problem.^{10,11} Waste is mainly generated from pulping, de-inking unit operations and wastewater treatment. The amount and composition of the waste depends on the paper grade produced, the raw materials used, the process techniques applied and the paper and cardboard properties to be achieved. The wastes can be reused and valorised in a safe and environ-

mental way, but landfill disposal of paper and cardboard sludges is the only waste disposal method used in Serbia.

The paper and cardboard factory sludges could be effective stabilizers due to their organic matter, silicate and carbonate contents. Organic matter is able to form stable complexes with several metals; the silicates are materials with high cation exchange capacity (CEC) and the bicarbonate/carbonate system is able to increase the pH value of soil. These chemical properties were able to reduce the harmful mobile metals in polluted soils when paper factory sludge was added to them.¹²

The long-term performance of S/S waste forms is difficult to predict. The mechanisms governing heavy metals leachability of contaminants from monolithic solidified waste forms, and the evaluation of the long-term behaviour of S/S wastes can be effectively examined using the American Nuclear Society's (ANS) semi-dynamic leaching test.¹³ The ANS 16.1 provides substantially more information regarding the "real time" rate at which heavy metals are released from a solidified product as compared to other leaching tests (toxicity characteristic leaching procedure – TCLP). The leaching results extend over a 90-day period instead of a single result at the end of the test (toxicity characteristic leaching procedure). The ANS 16.1 method was modified by including acetic acid (AA) and humic acid (HA) solutions as leachates instead of deionised water (DI). This was applied with the objective to mimic the worst possible conditions for S/S waste disposed in a landfill. It should be noted that this was used for the first time in the experiments with cardboard factory sludge and sediment.

The most commonly used leaching test, recommended by the USEPA, is the TCLP test, but it only provides one result to define waste toxicity.¹⁴ The TCLP was specifically designed to mimic acidic conditions in a sanitary landfill and identify wastes that have potential to contaminate ground water.

In view of the above, the objectives of this study were: 1) to define the zinc distribution in dredged sediment and evaluate its environmental risk based on the results of the sequential extraction procedure; 2) to assess the effectiveness of zinc immobilization using S/S treatments with cardboard factory sludge; 3) to evaluate the effectiveness of zinc immobilization by the S/S treatments under conditions which mimic a landfill environment and 4) to determine the controlling leaching mechanisms of zinc in the treated sediment samples.

EXPERIMENTAL

The cardboard factory sludge used was obtained after physical-chemical wastewater treatment from cardboard mill situated in Belgrade and its chemical characteristics were: pH 8.6 ± 0.3 , CEC $14.8 \text{ (cmol kg}^{-1}\text{)}$, water content 39 %, organic matter 31 %, carbonates 25 %, silicates 47 % and Zn 10.1 mg kg^{-1} . The sludge was dried, crushed and passed through a 2.0 mm sieve, then characterised and used in the experiments.

Fresh sediment was collected using an Eijkelkamp core sampler, from the middle sediment on a stretch of the Veliki Bački Canal (Serbia), and was placed in a sealed plastic, acid-rinsed box (15 cm×15 cm and 20 cm deep) immediately after sampling.

Sediment initial water content was 75 %. Organic matter content, 4.48 ± 0.1 %, ¹⁵ was determined as loss on ignition. Sediment pH of 7.3 ± 0.4 , ¹⁶ was measured according to ASTM D 4972-01. The given values represent the means of three measurements.

The pseudo-total zinc content was assessed in triplicate after *aqua regia* digestion¹⁷ and determined using an atomic absorption spectrometer (AAS, Perkin Elmer AAnalyst 700, USA) or an inductively coupled plasma mass spectrometer (ICP-MS, Perkin Elmer ELAN 5000, USA) according to the standard procedures.¹⁸ The standard deviations (% *RSD*) obtained ($n = 3$) were below 10 %. The results of the sediment zinc pseudo-total concentrations are discussed in reference to the Canadian guidelines¹⁹ and the Dutch regulations,²⁰ since Serbia has neither an established system of continual sediment quality monitoring nor regulations covering quality standards.

The microwave assisted sequential extraction procedure (MWSE) was performed using identical operating conditions for each individual BCR fraction.²¹ Mean values were used and the *RSD* values ($n = 3$) were below 5 %. The sum of the four steps of the sequential procedure and the pseudo-total zinc contents differed by less than 10 %. Zinc contents in the sequential extraction procedure steps were analyzed by AAS or ICP-MS according to standard procedures.¹⁸

Sediment and cardboard factory sludge were dried to constant mass at 105 °C. The sediment and cardboard factory sludge were mixed in proportions of 95:5 (M1), 90:10 (M2), 80:20 (M3), 70:30 (M4) and 50:50 (M5) by mass. The samples were prepared in the form of monolithic cubes ($(3\pm 0.1)^3$ cm³) by compaction. The compaction was performed according to ASTM D 1557-00,²² providing a compaction force of 2700 kN m⁻³. The samples were cured at 20 °C in sealed sample bags for 28 days and then subjected to the leaching tests, *i.e.*, the standard TCLP leaching method and ANS 16.1.

According to the USEPA protocol,¹⁴ a 0.1 M acetic acid solution of pH 2.88 was used to extract the control sample and the S/S-treated samples since the pH was above 5. The sediment samples were extracted at a liquid to solid (L/S) ratio of 20 in capped polypropylene bottles on a rotary tumbler at 30 rpm for 18 h. After extraction, the final pH of the leachate was measured and the liquid was separated from the solids by filtration through a 0.45 µm pore size membrane filter. This test was applied on every sample in triplicate and the *RSD* values were below 5 %.

The ANS 16.1 test was applied in duplicate only on the M2 sample using deionised water as the leachate, and the *RSD* values were below 10 %. The ANS 16.1 method was modified by including 0.014 M acetic acid pH 3.25 and humic acid solutions (20 mg TOC L⁻¹, pH 6.55) as leachate instead of deionised water (DI). All materials in contact with the leachate were pre-cleaned with HNO₃ and subsequently rinsed with deionised water. All results are expressed with respect to sediment dry matter.

Evaluation of the leaching behaviour diffusion model

The long-term leachability of zinc from the S/S treated sediments was evaluated using the ANS method 16.1.¹³ By applying this test, the cumulative fraction of zinc leached *versus* time was obtained. The ANS has standardized the Fick's Law-based mathematical diffusion model as follows:

$$D_e = \pi \left[\frac{\frac{a_n}{A_0}}{(\Delta t)_n} \right]^2 \left[\frac{V}{S} \right]^2 T_n \quad (1)$$

where a_n is the contaminant loss (mg) during the particular leaching period with subscript n ; A_0 is the initial amount of contaminant present in the specimen (mg); V is the specimen volume (cm^3); S is the surface area of the specimen (cm^2); $\Delta(t)_n$ is the duration of the leaching period in seconds; T_n is the elapsed time to the middle of the leaching period n (s), and D_e is the effective diffusion coefficient ($\text{cm}^2 \text{s}^{-1}$).

The relative mobility of zinc can be evaluated by this coefficient, which varies from $D_e = 10^{-5} \text{ cm}^2 \text{ s}^{-1}$, (very mobile) to $D_e = 10^{-15} \text{ cm}^2 \text{ s}^{-1}$ (immobile).²³

Determination of the controlling leaching mechanism

The type of leaching mechanism that controls the release of metals can be determined based on the values of the slope of the logarithm of the cumulative fraction release, $\log(B_t)$, versus the logarithm of time, $\log(t)$:²⁴

$$\log(B_t) = S \log(t) + \log \left[U_{\max} d \sqrt{\left(\frac{D_e}{\pi} \right)} \right] \quad (2)$$

where D_e is the effective diffusion coefficient in $\text{m}^2 \text{ s}^{-1}$ for component x ; B_t is the cumulative maximum release of the component in mg m^{-2} ; t is the contact time in seconds; U_{\max} is the maximum leachable quantity in mg kg^{-1} and d is the bulk density of the product in kg m^{-3} .

When the slope (S) is close to 1 (0.60–1.00), the process is defined as dissolution. In such a case, the dissolution of the material proceeds faster than diffusion. If the slope is around 0.50 (0.40–0.60), the release of heavy metals will be slow and diffusion will be the controlling mechanism. If the slope is less than 0.40, the release of metal is probably due to surface wash-off.²⁴

RESULTS AND DISCUSSION

Pseudo-total metal concentrations and sequential extraction of the untreated sample

The pseudo-total zinc concentration in the sediment was $1137 \pm 54 \text{ mg kg}^{-1}$. According to Dutch standards,²⁰ the sediment is polluted with zinc (class 4). Class 4 sediments are of unacceptable quality and have the highest urgency for action: dredging, disposal in special storage reservoirs and, if possible, sediment clean-up measures.²⁰ Compared with the Canadian Sediment Quality Guidelines for aquatic life protection,¹⁹ the metal contents are above the PEL value. Sediment concentrations above the *PEL* values are expected to be frequently associated with adverse biological effects. Although the *PEL* is considered to be applicable to a variety of sediment types, it cannot define uniform values of sediment pollution as the bioavailability (and hence toxicity) of the contaminants may be different.¹⁹

The percentage of extracted zinc in the most available, mobile fraction was 24.4 %. The percentages of zinc in the other modified BCR fractions according to their relative contents are as follows: 12.7 % in the reducible fraction, 1.9 % in oxidisable form, and 61.2 % in the residual fraction.

The sequential extraction results are not in full agreement with the results of the pseudo-total metal concentration in the sediment, which only confirms the opinion that the total metal concentration is not sufficient to define the real danger presented to the environment. According to the risk assessment code (RAC), 24.4 % of zinc in the carbonate phases comes under the medium risk category.⁷

The total non-residual fraction (exchangeable+reducible+oxidisable) of zinc was 39 %. This indicates that these zinc fractions in the sediment are potentially available for exchange and/or release into the environment.

Leaching tests: ANS 16.1 and TCLP

Samples of sediment-cardboard factory sludge mixtures were subjected to the semi-dynamic leaching test ANS 16.1.¹³ This test has not been previously applied to mixtures of metal-contaminated sediments with cardboard factory sludge. The total cumulative fractions of zinc leached (%) after completion of the ANS 16.1 test using deionised water, acetic acid and humic acid solutions as leachates are presented in Table I.

TABLE I. Total cumulative fraction of zinc leached (*CFL*) after completion of the ANS 16.1 test using deionised water, acetic acid and humic acid solutions as leachates and the mean effective diffusion coefficient D_e for untreated (SO) and treated sediment samples (M1, M2, M3, M4 and M5)

Parameter	SO	M1	M2	M3	M4	M5
Deionised water						
<i>CFL</i> / %	17.1	5.13	4.52	4.30	3.21	4.40
D_e / cm ² s ⁻¹	4.84×10^{-8}	7.94×10^{-13}	5.01×10^{-13}	3.16×10^{-12}	2.51×10^{-13}	6.34×10^{-13}
Acetic acid solution						
<i>CFL</i> / %	19.8	7.17	6.33	6.05	4.49	6.40
D_e / cm ² s ⁻¹	5.62×10^{-6}	6.62×10^{-10}	4.01×10^{-10}	2.04×10^{-10}	7.95×10^{-11}	3.12×10^{-10}
Humic acid solution						
<i>CFL</i> / %	18.3	6.16	5.42	5.16	3.85	5.34
D_e / cm ² s ⁻¹	2.72×10^{-7}	3.16×10^{-11}	2.52×10^{-11}	1.58×10^{-11}	7.93×10^{-11}	1.96×10^{-11}

The amount of zinc released during the ANS 16.1 tests for the untreated samples did not exceeded 20 % of the total mass of the contaminant in the waste, which is the upper limit for the diffusion model to be still applicable.¹³ The cardboard factory sludge exhibited good sorption properties, significantly reducing zinc leachability compared to the untreated sample. This is in agreement with literature data.^{10,12,25-30}

The humic acid leachate extracted more zinc than deionised water from all samples. Humic acids are very important in the formation of stable organo-mineral complexes because of their physico-chemical and biological stability. Due to their amphipathic nature and structural features, HAs play an important role in environmental processes governing the fate and transport of organic and inorganic pollutants in natural systems. They include binding sites with different complexation strength and are able to form inert and labile complexes with inorganic cations and organic compounds. Based on some previous studies, zinc shows a pronounced tendency for complexing with humic acids.^{31–34}

The amount of metal leached increased with high sorbent loading (M5). Similar results were reported by other authors.^{35–37} This may be attributed to two reasons: *i*) a large amount of adsorbent effectively reduces the unsaturation of the adsorption sites and correspondingly, the number of such sites per unit mass decreases resulting in comparatively lower adsorption at higher adsorbent amounts, and *ii*) higher amounts of adsorbent create particle aggregation, resulting in a decrease in the total surface area and an increase in diffusion path length, both of which contribute to decreasing the amount adsorbed per unit mass.

The diffusion coefficients D_e for the treated samples, calculated using Eq. (1), are listed in Table I. The mobility of zinc in this study was reduced by treatment possibly due to the high sorption capacity of the sludge.^{10,12,25–30} The diffusion coefficients are in agreement with the results of the cumulative release of zinc from the treated samples. The diffusion coefficients for metals in the treated samples ranged from 10^{-10} to 10^{-13} $\text{cm}^2 \text{s}^{-1}$ (low mobility).

At present, TCLP is used by the USEPA to evaluate whether a particular S/S process is effective in treating a given waste in terms of reduction of the contaminant mobility and toxicity.¹⁴ The results of the TCLP leaching test are presented in Table II.

TABLE II. Results of the TCLP leaching test (mg L^{-1} , mean value \pm SD) for untreated (SO) and treated sediment samples (M1, M2, M3, M4 and M5)

SO	M1	M2	M3	M4	M5
0.94 \pm 0.02 ^a	0.40 \pm 0.01	0.32 \pm 0.01	0.28 \pm 0.01	0.21 \pm 0.01	0.25 \pm 0.01

^adata obtained from triplicate, standard deviation

A comparison of the concentrations obtained in the TCLP test showed that the zinc concentrations in the treated samples were below the concentration obtained for untreated sediment, meaning the zinc had been successfully incorporated into the treated matrix.

Overall, bearing in mind the high initial zinc concentration of the sediment sample, the treatment was very effective. This may be difficult to readily explain due to the degree of complexity in natural sediment samples. In nature there are many constituents that could participate in and influence zinc leachability. Fur-

ther research is required to obtain some of this information in order to evaluate zinc speciation and subsequent mechanisms of incorporation and release in these samples.

Controlling leaching mechanism

The controlling leaching mechanisms were evaluated using the diffusion model (Eq. (2)). The slope (S) and R^2 values obtained from the diffusion model are presented in Table III.

TABLE III. Slope (S) and R^2 values obtained from the diffusion model for untreated (SO) and treated sediment samples (M1, M2, M3, M4 and M5)

Parameter	SO	M1	M2	M3	M4	M5
Deionised water						
S	0.22	0.43	0.47	0.47	0.48	0.47
R^2	0.99	0.99	0.99	0.98	0.99	0.99
Acetic acid solution						
S	0.20	0.41	0.40	0.53	0.54	0.60
R^2	0.98	0.99	0.96	0.98	0.96	0.92
Humic acid solution						
S	0.29	0.42	0.45	0.54	0.56	0.58
R^2	0.98	0.96	0.99	0.99	0.97	0.99

The slope values for the mixtures ranged from 0.41 to 0.60 for all the treated samples. This indicates that diffusion was the dominant leaching mechanism. In the untreated sediment sample, the slope values ranged from 0.20 to 0.29, indicating that the dominant leaching mechanism was surface wash-off.

Further research should be focused on a more detailed analysis aimed at elucidating the encapsulation of zinc into the sediment structure and its leaching mechanism, relying on studies of mineralogy (qualitative and quantitative X-ray diffraction) as well as micromorphology (scanning electron microscopy and optical microscopy).

CONCLUSIONS

Assessment of the sediment quality based on the pseudo-total zinc content showed it to be severely contaminated according to the corresponding Dutch standards and Canadian guidelines. The zinc seems to pose a medium risk based on the modified BCR sequential extraction procedure and the risk assessment code.

The immobilization treatment applying cardboard factory sludge appeared to be effective in the remediation of the zinc-contaminated sediment. Based on the cumulative fraction of zinc leached, the diffusion coefficients and the results of the TCLP tests, it appears that the treatment efficiently reduced zinc mobility. In all samples, the controlling zinc leaching mechanism after S/S treatment ap-

peared to be diffusion. Hence, only small amounts of zinc could be expected to leach into the environment over time from the treated sediment.

Due to the large volumes of waste generated in the paper, cardboard and pulp industry, it is necessary to continue to increase environmental awareness about the different applications of wastes, while taking into account the environmental and economic factors influencing these waste treatments. Conventional waste management methods, which might have been acceptable in the past, might not be optimal to meet present and future requirements. The utilisation of cardboard factory sludge to “remediate” a contaminated sediment could be an interesting approach.

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ИЗВОД

КОРИШЋЕЊЕ ОТПАДА КАРТОНСКЕ ИНДУСТРИЈЕ ЗА РЕМЕДИЈАЦИЈУ СЕДИМЕНТА КОНТАМИНИРАНОГ ЦИНКОМ

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Отпад у индустрији папира, картона и пулпе настаје као последица бројних процеса, а његова све већа продукција доводи до настајања проблема његове диспозиције. У овом раду је приказана могућност коришћења отпадног муља картонске индустрије као стабилизационог агенса у третману солидификације/стабилизације (С/С) седимента загађеног цинком. Да би се испитала ефикасност примењеног третмана и процена потенцијала излуживања цинка у дужем временском периоду примењени су семидинамички тест излуживања и тест излуживања токсичности. Модел базиран на дифузији је послужио за процену механизма који је доминантан током излуживања. Примењени третман се показао ефикасним за имобилизацију цинка, а доминантан механизам излуживања је дифузија. Резултати указују на то да би требало очекивати споро излуживање цинка када се отпадни муљ картонске индустрије примени као стабилизациони агенс у С/С третману.

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