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Extraction of heavy metals from e-waste contaminated soils using EDDS

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Abstract

Environmental contamination due to uncontrolled e-waste recycling activities is drawing increasing attention in the world. Extraction of these metals with biodegradable chelant [S,S]-ethylenediaminedisuccinic acid (EDDS) and the factors influencing extraction efficacy were investigated in the present study. Results showed that the addition of EDDS at low pH (5.5) produced higher metal extraction than that at high pH (8.0) solution. Metal speciation analysis indicated that Cu was completely complexed with EDDS at different pH conditions with various amounts of EDDS applied. For Pb and Zn, at low EDDS dose of 0.304 mol/kg soil, they were present as Pb- and Zn-EDDS. However, at high EDDS dose of 1.26 mol/kg soil, most of Pb was bound with dissolved organic matter. Ca and Al were found to be strong competitors for trace metals to EDDS at low application dose and low pH condition.

Key words: e-waste; soil washing; biodegradable chelant; heavy metal; EDDS

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Introduction

E-waste as one of the fastest growing waste types of the 21st century is flooding into the environment because of large number of electronic products and shortened life span. Estimated e-waste is currently growing at a rate of 4% per year in the world. It was estimated that China was the second largest e-waste-producing country with 2.3 million tons in 2010, following the U.S. of about 3 million tons (Focus, 2005). In addition, large quantities of e-waste were exported to China from developed countries due to the lacking enforcement of law, and the temptation of the profits behind the uncontrolled recycling of precious metals from these materials. In some places within China, e-waste recycling was quite active in the past two decades (Li et al., 2007; Shen et al., 2008; Wong et al., 2007a). Primitive recycling techniques, such as melting circuit board over a coal grill and strong acid leaching, were often used in the e-waste processing leading to large quantities of toxic metals and organic substances released into air, waters, soils, vegetables and other crops nearby, threatening the health of local residents (Bi et al., 2007; Fu et al., 2008; Yu et al., 2006; Zhao et al., 2009). The combustion of the unsalvaged e-waste scraps is most often adopted to reduce the volume prior to the recovery of precious metals. High concentrations of trace metals, such as more than 20 g/kg Cu and 4 g/kg Pb and Zn, were reported in the e-waste open burning sites (Luo et al.,

2011; Sepúlveda et al., 2010; Wong et al., 2007b), and these locations have been identified as major pollution sources which may pose a significantly potential risk of metal leaching to the surrounding environment. Therefore, the open burning sites should be set as the priority of the remediation task in the e-waste contaminated area (Luo et al., 2011).

Metal-contaminated soils can be remediated by *in situ* and *ex situ* techniques, such as flushing, phyto-extraction, soil washing and heap leaching. Chelating agents have been extensively studied and used in various clean-up methods (Hauser et al., 2005; Kos and Leštan, 2003b; Leštan et al., 2008; Luo et al., 2005, 2006a; Tandy et al., 2004). The *ex situ* soil washing is proposed as a useful technique due to its high potential extraction efficiency. The method poses less risk of metal leaching than *in situ* methods as most of the chelant is removed from the soil before returning to the field. EDTA (ethylenediaminetetraacetic acid) has been the most often used chelating agent, but it is recalcitrant in the environment so that residual EDTA and metal-EDTA complexes would remain exist in soil with high mobility and leading to potential adverse human health and environmental effects (Luo et al., 2006b, 2006c; Nowack, 2002). Thus, EDDS has recently emerged as a promising substitute of EDTA because it can efficiently extract heavy metals, and is also readily biodegradable in soils. In addition, EDDS is less toxic than EDTA to plants, and microorganisms, which would keep the fertility and function of the original soils (Kos and Leštan, 2003a; Grčman et al., 2003). How to apply this

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technique in a cost-effective way is a big concern in the real practice. Many factors such as the pH of chelant, extraction time, soil metal concentration and metal composition were found to influence the metal extraction efficiency in various extents. Hence, finding out the major factors controlling metal extraction and optimizing the operation procedure are of great importance.

The high concentration of trace metals in e-waste open burning sites and the relatively small scale in the field may make *ex situ* soil washing a potential clean-up method. The objectives of the present study are to evaluate the efficiency of EDDS on metal extraction of the e-waste contaminated soils, and to identify major factors influencing effective metal extraction.

1 Materials and methods

1.1 E-waste contaminated soil

The soil used in the present study was collected from a past e-waste open burning site (top 20 cm soil layer), located in a small town of Guangdong Province, South China (Luo et al., 2011). After being dried at 30°C, stones and plastic debris were removed, and the soil was passed through a 2-mm sieve. Total concentrations of metals were determined by ICP-AES (Optima 3300 DV, Perkin Elme, USA) after strong acid digestion (4:1 concentrated HNO₃ and HClO₄ (V/V)) (Luo et al., 2005). The metal concentrations and soil properties are given in Table 1.

1.2 Metal extraction with EDDS

The soil/solution ratio was set at 1:10 (W:V) by mixing 2 g of air-dried soil with 20 mL of deionized water (control) or EDDS solution in 50 mL polypropylene centrifuge tubes. EDDS (as Na₃EDDS salt, Fluka Chemie GmbH, Switzerland) was added to the soil at concentrations of 0, 0.304, 1.26 and 6.3 mol/kg soil, corresponding to 0-, 1-, 4- and 20-folds the molar sum of Zn, Cu, Pb and Cd in the soil, respectively. Hereafter, the four treatments were referred as to control, MR1, MR4 and MR20, respectively. The solution pH was adjusted with HNO₃ and maintained at pH 5.5 with 2 mmol/L MES (2-morpholinoethane-sulfonic acid), and pH 8 with 2 mmol/L HEPES (4-(2-hydroxyethyl)-piperazine-1-ethane-sulfonic acid). The two buffers added have very weak complexing properties and do not affect EDDS adsorption and complexation with metals in soil (Yip et al., 2009). The solution pH varied by at most 0.5 after the experiments. Three replicates were conducted for each treatment. Samples were shaken in a style of end-over-end at 200 r/min on a shaker, and all the tubes were wrapped with aluminium foil to avoid possible photodegradation during the extraction process.

The pH of the extraction solution was measured at the end of every reaction interval within 7 days (0.5, 1, 2, 4,

8, 16, 24, 48, 72, 120 and 168 hr), and the suspension was centrifuged for 10 min at 4000 ×g followed by filtration through 0.45 μm filter paper (Waterman, England). The solutions used for metal analysis were acidified with concentrated HNO₃, and metal concentrations in the extracts were determined by ICP-AES. The total dissolved organic carbon (DOC) concentrations were measured by TOC analyzer (TOC-5000A, Shimadzu, Japan). The EDDS concentrations were measured with the colorimetric method described by Vandevivere et al. (2001b) and Yip et al. (2009) using UV-visible spectrophotometer (Libra S35, Biochrom, UK), in which the absorbance at 670 nm was measured, and the detection limit was 0.1 mmol/L EDDS.

1.3 Speciation of metals with EDDS

The EDDS and metal speciation in the supernatants obtained from the batch extraction experiments was calculated with ECOSAT (Version 4.8, Wageningen University, the Netherlands). The input parameters include the measured EDDS concentrations, the dissolved metal concentrations (Cu, Zn, Pb, Cd, Al, Ca, Mn and Fe), the DOM concentrations and the pH of soil solutions. The stability constants of metal-EDDS complexes were cited from previous publications (Tandy et al., 2006; Martell et al., 2001; Orama et al., 2002). The concentration of DOM was calculated by multiplying the DOC concentrations (40 mg/L at pH 5.5 and 52 mg/L at pH 8.0) by two where the DOC coming from EDDS has been subtracted (Koopmans et al., 2008). The composition of DOM was assumed to be twice the DOC, and to be composed of 50% fulvic acid, and 50% humic acid (Tandy et al., 2006). Parameter values for metals binding to DOM were taken from Milne et al. (2003), and the binding was modeled using the NICA-Donnan model (Kinniburgh et al., 1996). Al and Fe oxides Al(OH)₃ and amorphous Fe(OH)₃ may be precipitated in solution, which was considered in the calculation with the model (Koopmans et al., 2008; Yip et al., 2009).

1.4 Statistical analysis

The data were statistically analyzed using statistical package, SPSS 17.0 (SPSS, USA). Variance analysis ($p < 0.05$) of metal concentrations among different treatments was performed using a one-way ANOVA test (Tukey HSD), and the two-sample *t*-test was employed to examine the statistical significance of differences in the mean concentration of these metals between (or among) different treatment groups.

2 Results and discussion

2.1 Heavy metal extraction

The effect of EDDS application on metal extraction of e-waste contaminated soils was assessed by a 7-day batch

Table 1 Physicochemical properties of used soil

Al	Ca	Cd	Metal concentration (mmol/kg)						pH	Texture (%)		
			Cu	Fe	Mg	Mn	Pb	Zn		Sand	Silt	Clay
1530 ± 49	563 ± 7.1	0.170 ± 0.004	199 ± 3.1	246 ± 5.8	143 ± 2.1	3.52 ± 0.06	50.6 ± 1.0	54.3 ± 0.2	7.8	70.7	23.5	5.80

extraction experiment. In view of the nature of soil contamination, Cu, Pb, Zn, and Cd were selected as the target metal pollutants for soil washing. As shown in Fig. 1, when EDDS was absent, no Cd, and only very low concentrations of Cu, Zn and Pb (< 0.0187 mmol/L) were extracted at pH of 5.5 and 8.0, and there was no significant difference observed in metal extraction between these two pH conditions ($p > 0.05$). The addition of EDDS to the soil dramatically increased the concentrations of Cu, Pb, Zn, and Cd, with a quick increase in the first 8 hr, and an apparent equilibrium reached after 48–72 hr (Fig. 1). The reaction between chelant and metals in soil took place in a short time after the chelant was applied to soil. This was observed in many other studies (Tandy et al., 2004; Yip et al., 2009), and it was generally thought the 24 hr was an optimum extraction time for metals in tested soils (Tandy et al., 2004). In the present study, more than 80% of the maximum levels of metals extracted in the whole experiment period have been extracted in the first 24 hr (Fig. 1).

The amount of chelant applied to soil plays a great role in the metal extraction efficiency. More metals were extracted at a higher dose of EDDS addition (1.26 mol/kg soil) with 36%, 60%, 19%, and 68% increase of Cu, Zn, Pb

and Cd extracted from the soil, respectively, in comparison with the low dose of application (0.304 mol/kg soil) (Fig. 1). The pH of extraction solution was another crucial factor possibly influencing the metal extraction from soil. There was no difference observed in metal extraction between the two pH conditions in the control groups (with no EDDS addition). However, the difference between the two pH conditions was obvious when EDDS was present in soil. In general, for a given chelant concentration, low pH (5.5) resulted in higher metal extraction than the high pH (8.0) ($p < 0.05$). The influence of pH on metal extraction was more significant for Pb and Cd when EDDS was applied at high concentration ($p < 0.05$). At high EDDS application, more than about 19% and 51% Pb and Cd were leached out at pH 5.5, in comparison with pH 8.0 (Fig. 1). The difference was only 6% and 21% at low dose of EDDS. This result was different from the previous study by Tandy et al. (2004), where less pH dependence of Pb extraction was shown in high dose of chelant application.

Among the four target metals, Cu was the highest in extraction solution, followed by Pb, Zn, and Cd. The trend was consistent with the original concentrations of metals in the contaminated soil (Tables 1 and 2). The metal extraction efficiencies at different chelant/metal ratios are

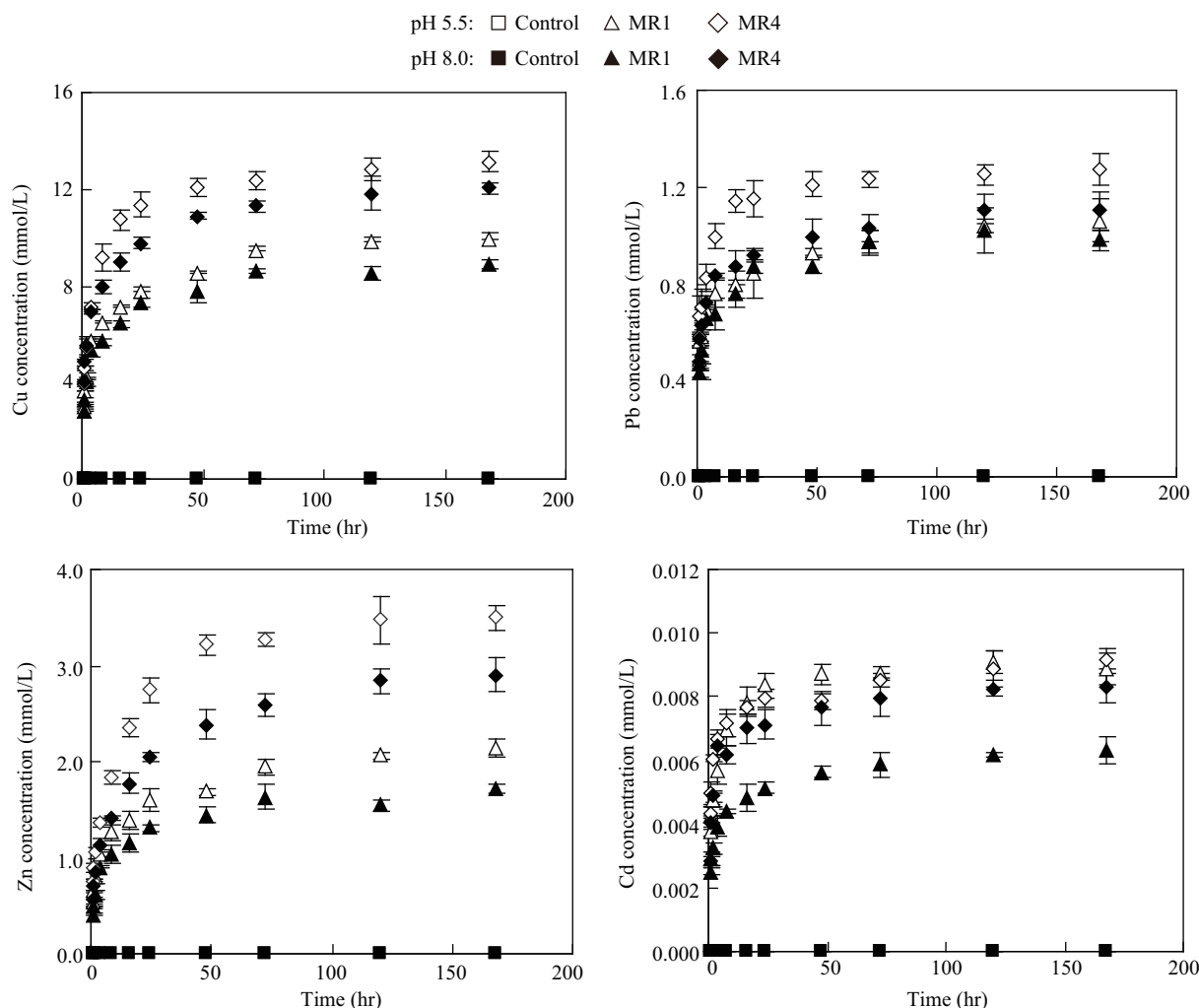


Fig. 1 Kinetics of Cu, Pb, Zn, and Cd extraction from e-waste contaminated soil with different EDDS concentrations (MR1: 0.304 mol/kg soil; MR4: 1.26 mol/kg soil) at pH 5.5 and pH 8.0.

Table 2 Metal extraction efficiencies from the e-waste contaminated soil after 7-day extraction at different EDDS concentrations

Metal	pH	Metal extraction efficiency (%)		
		MR1	MR4	MR20
Cu	5.5	49.8 ± 0.7 a	66.0 ± 2.3 b	64.2 ± 2.3 b
	8.0	44.7 ± 1.4 a	60.4 ± 3.6 b	61.1 ± 1.4 b
Pb	5.5	40.7 ± 1.0 a	48.8 ± 1.2 b	39.7 ± 3.2 a
	8.0	39.1 ± 0.5 a	42.3 ± 1.7 b	36.5 ± 2.7 a
Zn	5.5	39.4 ± 1.2 b	64.4 ± 0.8 b	64.4 ± 2.3 b
	8.0	31.6 ± 1.0 a	53.3 ± 1.7 b	54.2 ± 0.2 b
Cd	5.5	30.8 ± 1.3 a	52.2 ± 1.4 b	54.0 ± 1.7 b
	8.0	25.7 ± 0.9 a	37.2 ± 1.6 b	38.6 ± 2.9 b

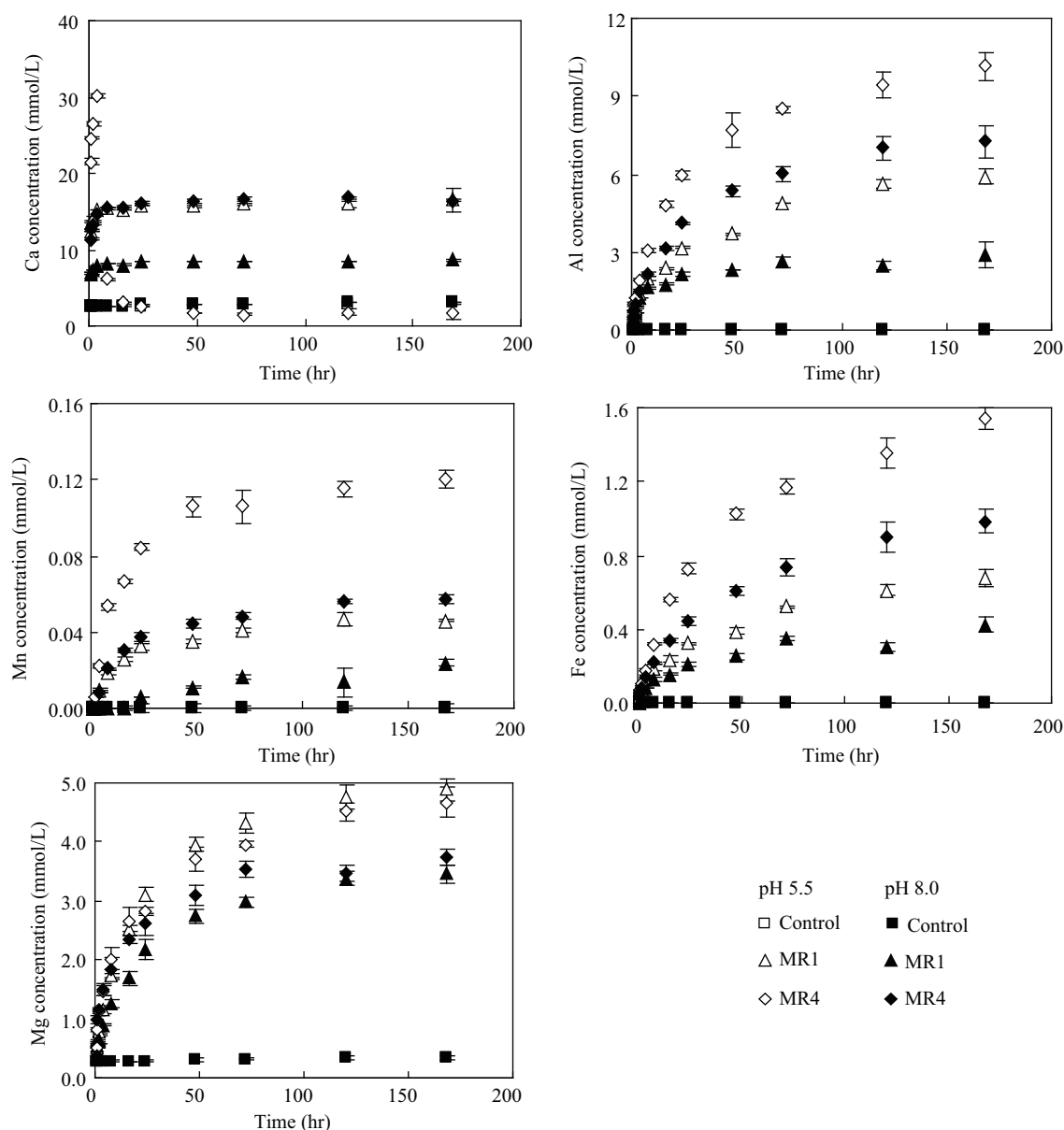
Values shown are mean ± S.D. Values followed by different letters are significantly different at $p = 0.05$. MR1, MR4 and MR20 means EDDS concentration of 0.304, 1.26 and 6.3 mol/kg soil.

given in Table 2. At the end of the experiment (168 hr), the EDDS application for MR4 at pH 5.5 showed the highest metal extraction, with 66.0%, 48.8%, 64.4% and 52.2% for Cu, Pb, Zn and Cd, respectively. No additional metals were extracted at MR20 in comparison with MR4 (Table 2).

2.2 Major element extraction

Once applied to soils, chelant not only extracts the target trace metals, but also the major soil elements such as Ca, Fe, Al and Mn. The dissolution of major elements with the addition of chelant could influence the efficiency of target metals extraction. Increased metal extraction at decreasing pH was observed to be offset by the increase of Ca extraction (Tandy et al., 2004). Apart from EDDS induced dissolution of major elements, the newly formed metal-EDDS complexes, such as Zn-EDDS and Pb-EDDS, were also able to dissolve iron oxide in a similar manner as the free EDDS, yet at a slow rate (Tandy et al., 2004).

In the current study, substantial amounts of Ca and Mg were extracted with deionized water when no chelant was applied, while the dissolution of Al, Fe and Mn was negligible during the extraction (Fig. 2). This might be attributed to higher percentages of Ca and Mg in the exchangeable and carbonate adsorbed phases (61.1% for Ca and 65.7% for Mg).

**Fig. 2** Kinetics of mineral extraction from e-waste contaminated soil with different EDDS concentrations at pH 5.5 and pH 8.0.

When EDDS was present, the extraction of Al, Fe and Mn increased at low pH and high ratio of chelant to soil. For Mg, the pH of the extraction solution had a big influence on its extraction and the EDDS dose had very little effect, with only less than 10% difference in the extractable Mg between the two doses of EDDS at the same pH. However, Ca showed a different scenario (Fig. 2). For Ca, at MR1, the extraction was high at low pH (5.5). However, for MR4 at pH 5.5, a different pattern was shown. During the initial extraction (within 4 hr), the concentration of Ca in the extracts at MR1 (pH 5.5) increased rapidly with the highest concentration reaching 29.9 mmol/L at 4 hr, then followed by a sharp decrease, and at 24 and afterwards, the concentration of Ca was even lower than the control (with no EDDS addition). This result was not possibly caused by the competition of EDDS with other metals because the Ca-EDDS complex was a very minor speciation for Ca (Fig. 3). The effect of pH at low MR was consistent with that observed in the study of Tandy et al. (2004), but did not agree with Yip et al. (2009), reflecting the complication of the factors on the dissolution of minerals, including soil pH, the concentration and speciation of metal contaminants, etc.

The dissolution of Ca was low after 24 for all the

treatments. However, from 24 to 168 hr the dissolution of Al and Fe increased gradually, and the concentrations of these two major elements at 168 hr were 2-fold of those at 24 hr. While the increase of target trace metals concentrations during this time interval was relatively minor (Figs. 1 and 2). The results indicated that after 24 hr Al and Fe could become more competitive for EDDS than trace metals. Prolonged extraction could facilitate the dissolution of major elements, and in turn, would mobilize soil colloids and fine particles, which could clog soil columns in flushing studies (Hauser et al., 2005; Zhao et al., 2007). Among the major elements, Ca dissolution was by far the most significant one (except treatment of MR4 at pH 5.5) (Fig. 2), followed by Al. The concentrations of Al and Fe in the extraction solution seemed to be controlled by the presence of amorphous and poorly crystalline oxides in soils (Komórek et al., 2009).

2.3 Metal and EDDS speciation

The reactions of EDDS in the environment occur predominantly in the form of metal-EDDS complexes. The reactivity of metal-EDDS complexes may reveal the changes in soil after EDDS application (Nowack, 2002). The distribution of EDDS species varied with the amount

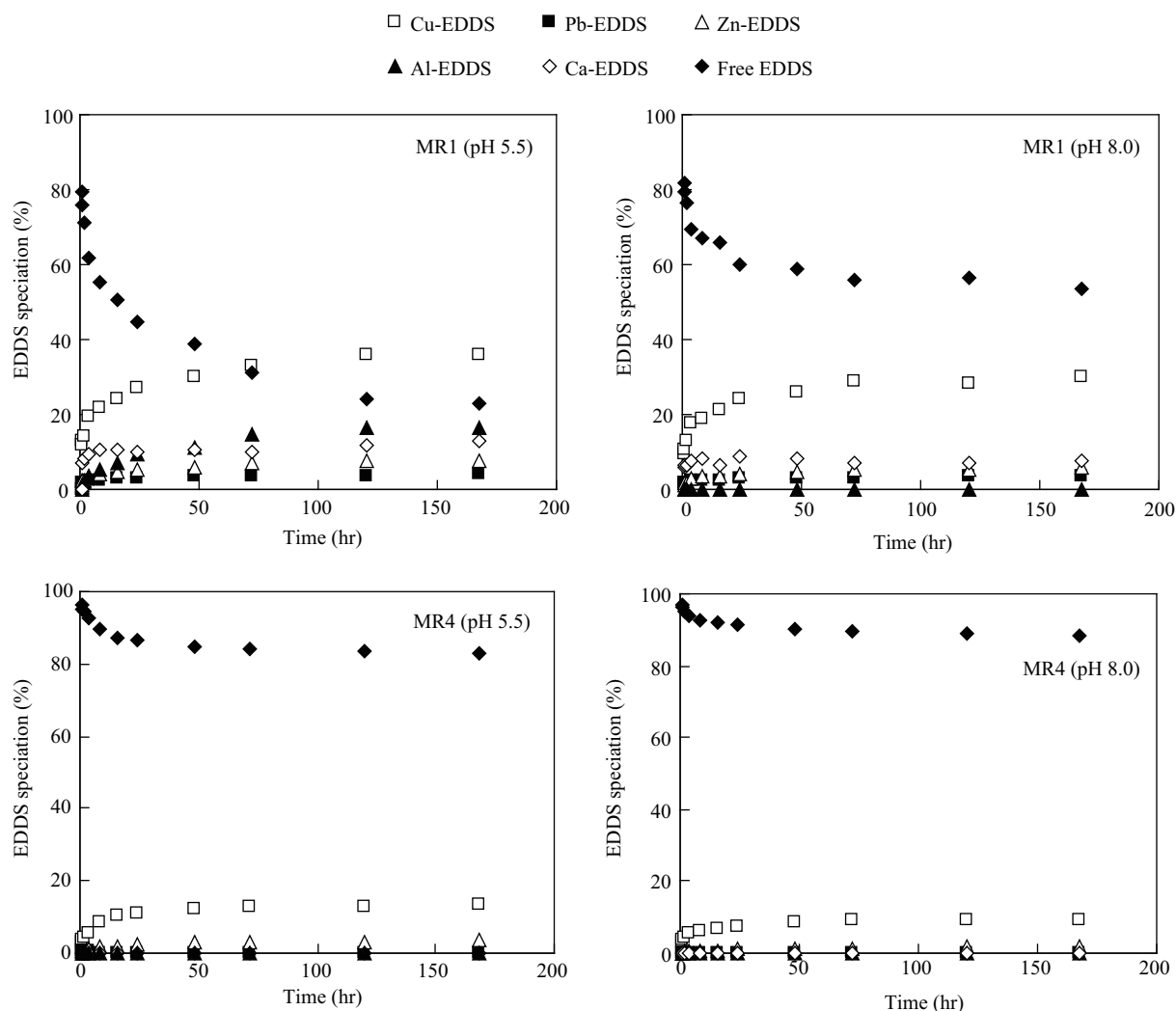


Fig. 3 Metal-EDDS complex speciation in solution at MR1 and MR4 during 168 hr extraction time.

of chelant added, pH and extraction time. Virtually all the Cu in soil solution was preferentially complexed with EDDS as Cu-EDDS (Fig. 4). Cu-EDDS was not easily exchanged due to its high ionic potential and its four strong equatorial bonds between Cu and EDDS (Tsang et al., 2009). EDDS is suitable for Cu-contaminated soils remediation due to its high affinity for the metal. Pb and Zn, appeared mainly in Pb- and Zn-EDDS complexes at MR1. However, different results were observed for MR4 (Fig. 4). At pH 5.5, Zn-EDDS accounted for about 95% of the total Zn; Pb-EDDS showed an initial increase within the first 4 hr being about 89% (at 4 hr) of the total Pb, then followed by a fast decrease and dropped to less than 10% of the total after 8 hr. At pH 8.0, Zn-EDDS varied between 48%–76% of the total, and very minor of Pb (< 1.5%) was in the form of Pb-EDDS (Fig. 4). Even though Zn- or Pb-EDDS complexes were low in solutions for MR4 (Fig. 3), the total dissolved Zn and Pb were still higher than that in the MR1 (Fig. 1). Further speciation analysis showed that Zn and Pb were complexed with DOM in addition to metal-EDDS forms (Fig. 5). Metal dissociation from the newly formed metal-EDDS complexes could dissolve minerals, such as iron oxides, which probably promoted the mobilization of colloids and organic matters, and solubilized the metals again with the increased DOM.

EDDS, like other chelants, not only complexes heavy metals, but also extracts organic matter from soil (Yang et al., 2001). Excess of free EDDS in the extraction solution could increase the extraction of organic matter from soils (Fig. 6). Pb was more easily to detach from Pb-EDDS than Zn-EDDS because of high sorption strength of Zn-EDDS, which was in agreement with that observed the less Pb-EDDS than Zn-EDDS in soil solution (Fig. 5).

Major element dissolution plays a great role in the EDDS speciation, in particular Al-EDDS at pH 5.5 contributed 10% of EDDS speciation for MR1, and Al-EDDS accounted for 80% of the total Al with the other 20% being Al-DOM. At pH 8.0, however, Al-EDDS dropped to < 5% of the total Al, and hydrolyzed Al, colloidal Al(OH)₃ and Al-DOM were the major species in the extraction solution. Ca-EDDS reached about 20% of the total Ca at MR1 at pHs of 5.5 and 8.0, and the remaining 80% was binding with DOM (Fig. 7). At MR4, Al and Ca were completely complexed with DOM. Therefore, the role of DOM in mineral species needs to be considered at MR4. In light of this, the relative amount of metal-DOM was analyzed (Fig. 6). At pH 5.5, during the initial 4 hr, the Ca-DOM increased rapidly to 42% of the total DOM, followed by an abrupt decrease to less than 2% at the end of the experiment. Al-DOM increased gradually from less

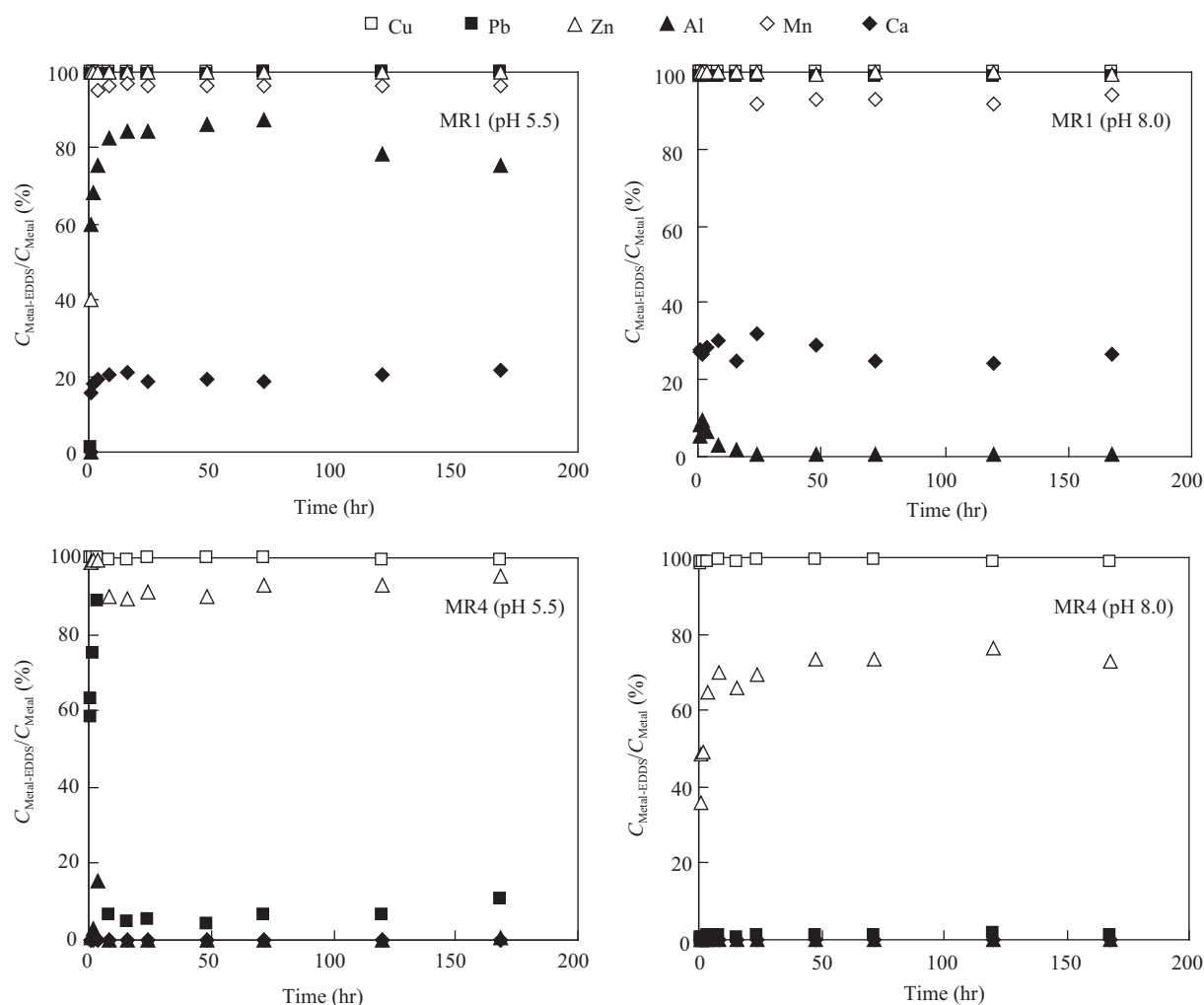


Fig. 4 Metal-EDDS complex as the percentage of total concentrations of dissolved metals at pH 5.5 and pH 8.0.

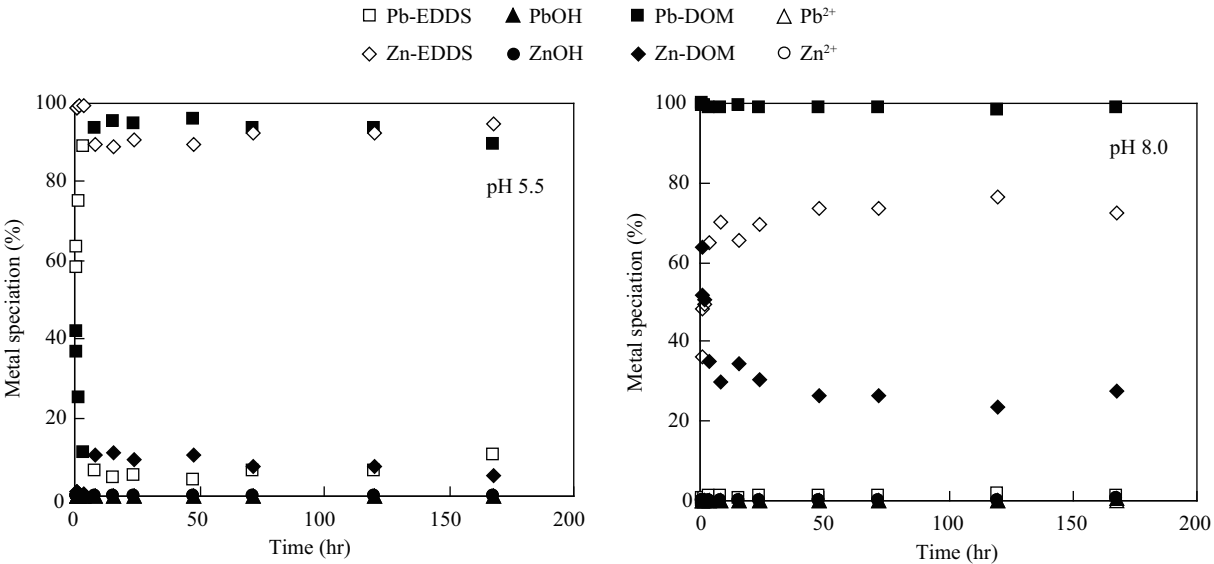


Fig. 5 Speciation of Pb and Zn in the soil solution at MR4 over 168 hr of the extraction time at pH 5.5 and pH 8.0.

than 1% to 15% during this period, which provides further evidence on that the fast decrease of Ca in the extraction solution was mainly caused by the competition of DOM

with Al (Fig. 6). At pH 8.0, the amount of Ca-DOM was higher than Al-DOM for MR1 (Fig. 6). Although Mn was totally complexed with EDDS, due to the low

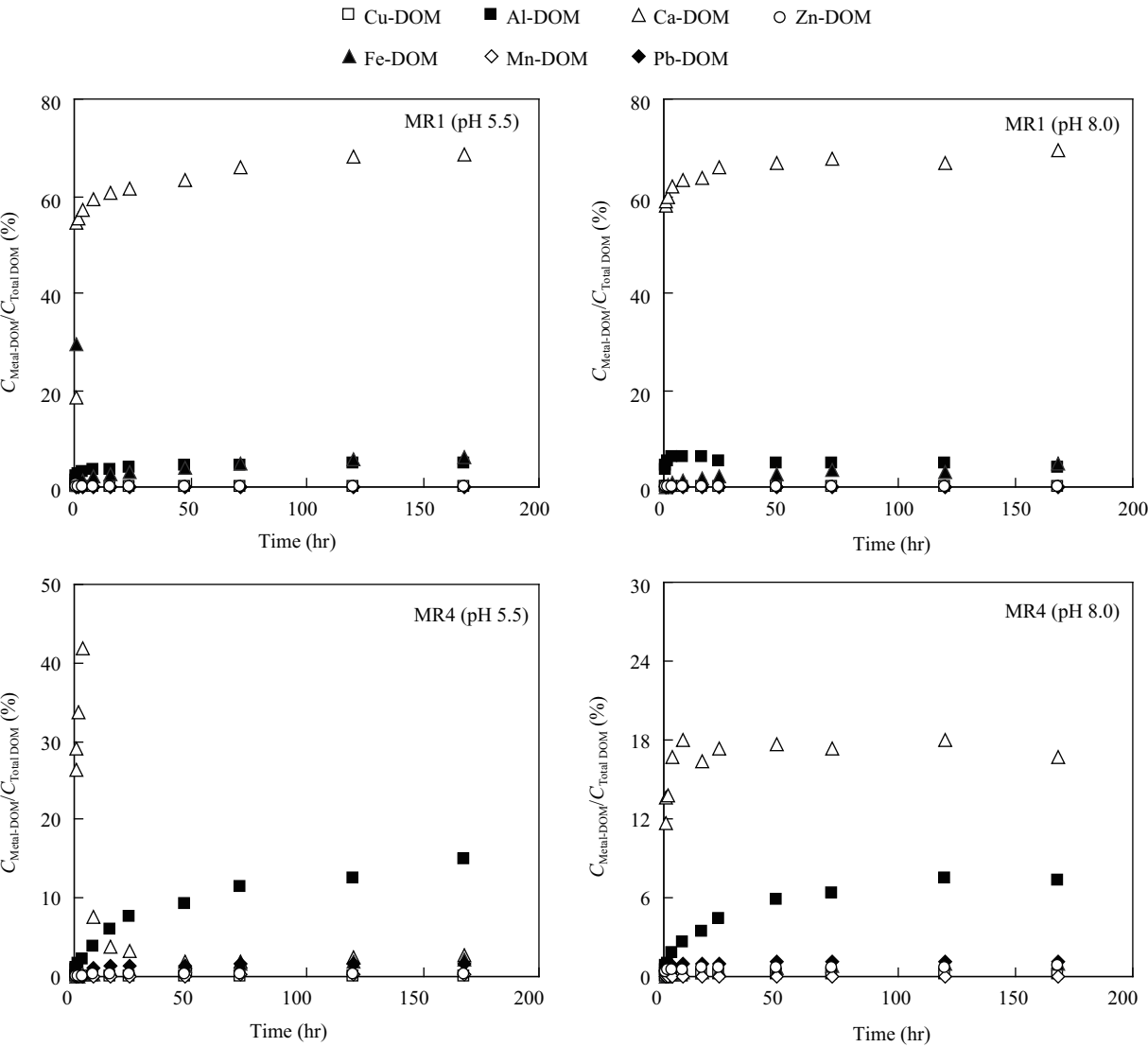


Fig. 6 Metal-DOM complex as the percentage of total DOM at pH 5.5 and pH 8.0.

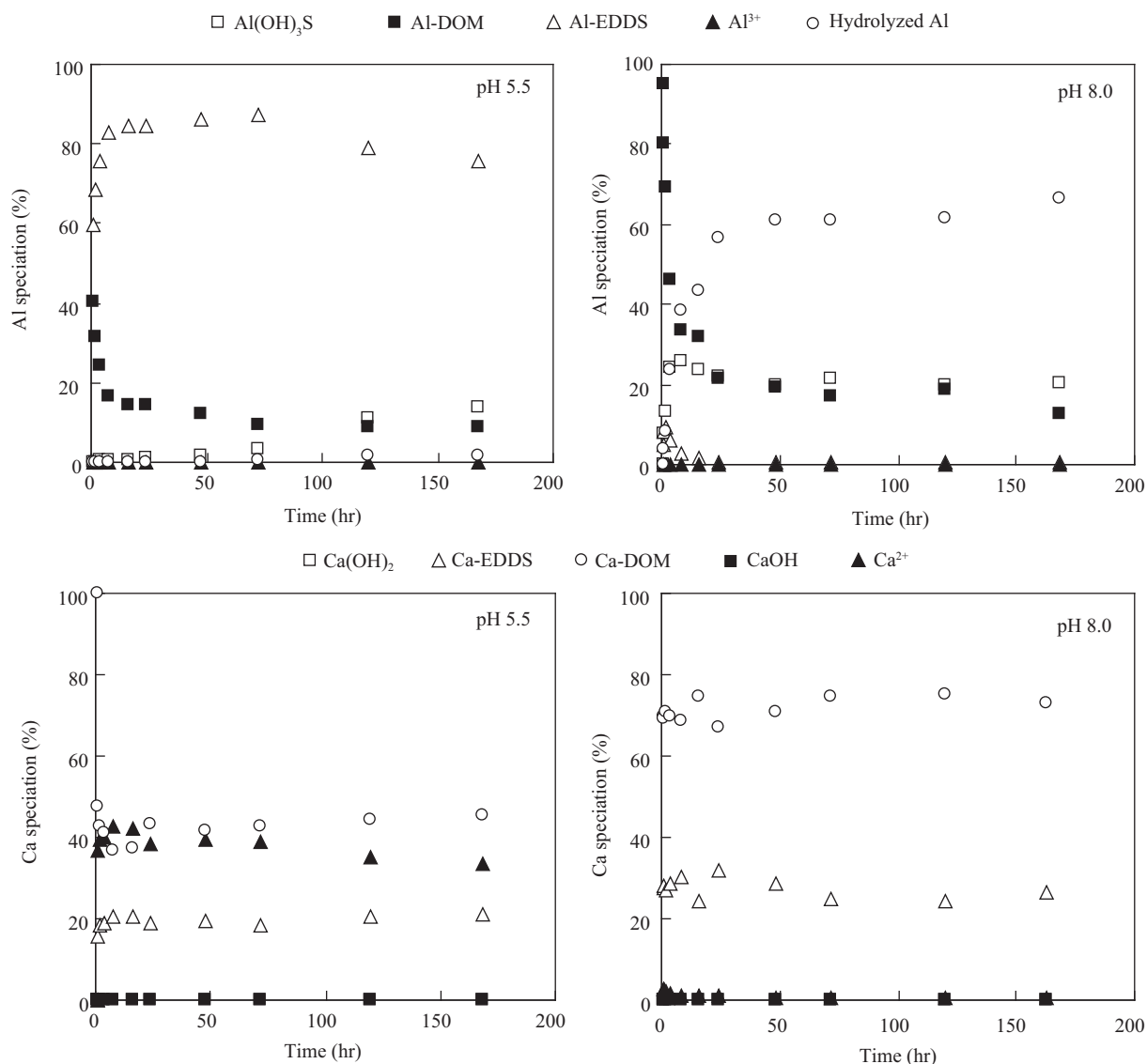


Fig. 7 Speciation of Al and Ca in the soil solution at MR1 over 168 hr of the extraction time at pH 5.5 and pH 8.0.

concentrations in soil, it showed a very limited role in EDDS speciation (Fig. 4). Fe was dominantly presented as Fe-DOM in the extraction solution for all the treatments, but much lower than Ca-DOM (Fig. 6).

2.4 E-waste contaminated soil clean-up implication

The effectiveness of EDDS in metal extraction from e-waste contaminated soil in the present study was comparable with previous studies on other soil types (Finžgar et al., 2006; Hauser et al., 2005; Kos and Leštan, 2004; Luo et al., 2005; Tandy et al., 2004; Vandevivere et al., 2001a). EDDS extracted metal contaminants almost exclusively from the exchangeable, mobile, Mn oxides, and the organic fractions (data not shown). Although the total metal concentration was still high in the e-waste impacted soil after the treatment by EDDS, the potential toxicity of metals to organisms should be evaluated by means of the bioavailability of metals and not by the total concentrations. Therefore, further toxicity tests are needed to evaluate the quality of EDDS-washed soils by means such as physiologically based extraction test (Ruby et al., 1993).

Disturbance of soil minerals on metal extraction can be of concern during the soil washing process using chelant. More information on the change of mineral compositions in soil is needed for an optimum extraction design, and selecting parameters, such as chelant dosage and extraction time. Based on the present study, it is suggested after the soil is extracted by EDDS for 24 hr, leaching solution could be circulated to extract other fresh soils which would reduce the operation cost of chelant usage. When the EDDS left in soils is degraded completely, soil extraction is suggested to carry out again with fresh EDDS in order to minimize the dissolution of Al and Fe from soil minerals. Further study using heap leaching is needed because this technology is much less destructive to soil property, and is easy to upscale for field applications.

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