



Extractabilities of heavy metals in chemically-fixed sewage sludges

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Abstract

Chemically-fixed sewage sludges, including lime-treated sludge (LS), lime–sodium silicate-treated sludge (LSS), cement-treated sludge (CS), and cement–sodium silicate-treated sludge (CSS), were produced from sewage sludge by bench processes using lime, cement, and sodium silicate as additives. The extractabilities of heavy metals (Cu, Pb, Cr, and Zn) in these chemically-fixed sludges were studied using sequential extraction and single extraction. For comparative objectives, untreated sewage sludge (S) was also studied. Sequential extraction revealed that the percentages of the heavy metals of organically-bound form in S were in the order of $\text{Cu} > \text{Pb} > \text{Cr} > \text{Zn}$ ($73.7 > 46.6 > 35.2 > 21.4\%$) and those of exchangeable form, the most labile form, in four chemically-treated sludges investigated were also in this order ($\text{Cu} > \text{Pb} > \text{Cr} > \text{Zn}$), implicating that the heavy metals (e.g. Cu) having higher affinity for organics become more labile in the sewage sludge after chemically-fixed, but those (e.g. Zn) having less affinity for organics become more stable. The results of single extraction conducted at various pH suggested that this effect was attributed to the irreversible dissolution of the metals (e.g. Cu) having higher affinity to organics at very high pH (10.64–12.05) during processing and air drying (20–25°C) of these chemically-fixed sludges. © 1998 Elsevier Science B.V.

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1. Introduction

A chemical fixation process of sewage sludge is a modified lime stabilization process: quicklime and an additive are processed with sewage sludge to produce a friable, soil-like material that is odorless and easily handled [1]. The additive can be

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Portland cement, sodium silicates, cement kiln dust, and lime kiln dust [2]. The process has become an alternative to incineration, composting, and digestion. It has been implemented in over 100 cities in the United States.

Sewage sludge often contains high levels of heavy metals that are harmful to the environment. Guidelines of heavy metals have been designed to limit the application of sewage sludge to agricultural land [3]. Regarding to the characteristics of heavy metals, many studies have focused on sewage sludges and sludge-amended soils rather than chemically-fixed sewage sludges. Thus, little information is available on the extractabilities and fractionation of heavy metals in chemically-fixed sewage sludges. The objectives of this study were to determine and compare the fractionation and extractabilities of heavy metals (Cu, Pb, Cr, and Zn) in the sewage sludge before and after being chemically-fixed.

Analytical limitations imposed by interference, selectivity, and sensitivity do not allow a complete differentiation of metals between all chemical forms in sewage sludges. However, a number of schemes have been proposed to fractionate metals on the basis of extractability in various chemical reagents either discretely or sequentially [4]. Metal forms in sediments, sludges, and soils have been investigated by many authors through selective chemical extraction techniques to obtain fractions corresponding to soluble, exchangeable, organic, adsorbed, precipitated, and residual forms [5–7]. Soon and Bates [8] mentioned that metals existing as exchangeable fractions in soils are considered to be the most available to plants, allowing us to evaluate the harmful effects of heavy metals through sequential extraction test. Of many sequential extraction protocols, the one developed by Tessier et al. [9] has been widely used for sediments and soils [10,11].

After a preliminary extraction test of chemically-fixed sludges and considering that chemically-fixed sludges mostly consisted of inorganic matter and look like a soil [1], the Tessier procedure of sequential extraction was used to fractionate the heavy metals in the sludges into five forms (exchangeable, carbonate-bound, oxide-bound, organically-bound, and residual). Single extraction conducted at various pH was also carried out to obtain more information on the extractability of organics and heavy metals in the sludges.

2. Experimental

The aerobically digested sewage sludge cake (13.61% solids) used in this study was sampled from the filter press outlet of the Min-Sheng Wastewater Treatment Plant, Taipei, Taiwan. The additives used were sodium silicate (Merck, $\text{Na}_2\text{O} = 7.5\text{--}8.5\%$; $\text{SiO}_2 = 25.5\text{--}28.5\%$; density = $1.296\text{--}1.396 \text{ g ml}^{-1}$; water content = 62%), lime (reagent grade $\text{Ca}(\text{OH})_2$), and Portland cement (Type I). Altogether, four types of sample were obtained from the chemical processing of sewage sludge: lime-treated sludge (LS), lime–sodium silicate-treated sludge (LSS), cement-treated sludge (CS), cement–sodium silicate-treated sludge (CSS). The sludge treated involving the addition of sodium silicate was prepared in three steps: mixing of sewage sludge cake (200 g) with sodium silicate (15 ml^{-1} for LSS; 10 ml^{-1} for CSS), further mixing with added lime (20 g for

Table 1
Procedure of sequential extraction

Step fraction	Extraction
1. Exchangeable	1 mol l ⁻¹ MgCl ₂ (pH 7), 20°C
2. Carbonate-bound	1 mol l ⁻¹ NaOAc + HOAc (pH 5), 20°C
3. Oxide-bound	0.04 mol l ⁻¹ NH ₂ OH · HCl + 25% HOAc, 95°C
4. Organically-bound	30% H ₂ O ₂ + 0.02 mol l ⁻¹ HNO ₃ (pH 2), 85°C; 2 mol l ⁻¹ NH ₄ OAc + 20% HNO ₃
5. Residual	HF/HClO ₄

LSS) or cement (25 g for CSS), and air drying (20–25°C). The sludge treated using only lime or cement as an additive was prepared by direct mixing the sewage sludge (200 g) with lime (20 g for LS) or cement (25 g for CS), followed by air drying (20–25°C). Untreated sludge (S) was prepared by drying in a forced-air oven (60°C). The products from the above treatment were powdered to pass through an 80-mesh sieve prior to X-ray diffractometry (XRD) analysis and extraction tests.

XRD was performed with the powdered samples (80-meshes) of above products. XRD patterns were obtained with a X-ray diffractometer (Shimadzu, XD-5) using CuK α radiation. The operation conditions for the X-ray examinations were the following: 45 keV accelerating voltage, 35 mA current, 10°–70° scanning range, 0.02° step-width, and 4° min⁻¹ scanning speed.

After a preliminary test of chemically-fixed sludges and considering that chemically-fixed sludges mostly consisted of inorganic matter and look like a soil [1], the Tessier procedure of sequential extraction (Table 1) was used to fractionate the heavy metals in the samples (duplicate) into five forms (exchangeable, carbonate-bound, oxide-bound, organically-bound, and residual). Additionally care was taken during the extraction steps for carbonate-bound and organically-bound fractions to prevent foam over.

The single extraction of duplicate samples conducted at various pH (pH 1–13) were carried out in 50 ml centrifuge tubes shaken in a reciprocal shaker bath (Yihder BT350R) at 125 rpm, 25°C. Each tube contained 1 g of sieved sample diluted to 20 ml with distilled water, of which the pH was adjusted with 1 M NaOH or 1 M HCl prior to full dilution. Each extraction lasted for 24 h, after which the supernatant from the centrifugation (Kubota 6800) at 12 000 \times g for 10 min was decanted, then analyzed with an atomic adsorption spectrometer (Perkin-Elmer 4000) and a total organic carbon analyzer (O · I Corporation 700) for concentrations of heavy metals and dissolved organic carbon (DOC). The residues from the centrifugation were discarded.

3. Results and discussion

3.1. Basic characteristics

The percentages of heavy metals (Cu, Pb, Cr, and Zn) in LS were about one-half those in S (Table 2). It resulted from the diluting effect of lime added (reagent grade Ca(OH)₂), which was free from heavy metals. The percentages of metals (Cu, Pb, Cr,

Table 2
Basic characteristics of chemically-fixed sewage sludges

Parameter	LS ^a	LSS ^a	CS ^a	CSS ^a	S ^a
pH ^b	12.05	11.82	10.78	10.64	7.65
Water (%)	6.99	14.93	11.0	10.83	1.5
Bulk density (g ml ⁻¹)	0.70	0.58	0.65	0.42	0.62
Cr (mg kg ⁻¹ sludge ^c)	52.16	62.95	73.4	86.3	89.89
Pb (mg kg ⁻¹ sludge ^c)	81.12	82.34	93.98	85.6	138.4
Zn (mg kg ⁻¹ sludge ^c)	621.6	517.5	625.2	498.8	1089
Cu (mg kg ⁻¹ sludge ^c)	119.4	96.11	112.0	96.87	202.0

^aS: untreated sludge; LS: lime-treated sludge; LSS: lime–sodium silicate-treated sludge; CS: cement-treated sludge; CSS: cement–sodium silicate-treated sludge.

^bMeasured in a 1:10 sample:0.01 M CaCl₂ solution.

^cOven-dry base for untreated sludge; air-dry base for chemically-fixed sludges.

Because sodium silicate solution and cement inherently contained heavy metals and both the weight and combined water content of the sludges varied after treatment, mass balance was not calculated.

and Zn) in LSS, CS, and CSS were larger than those in LS (Table 2), because the Portland cement and sodium silicate added both contained heavy metals. The pH of the samples, from the most to the least alkaline, was in the order of LS > LSS > CS > CSS > S (Table 2). The pH of the samples heavily influenced the extraction of heavy metals and organics, as will be discussed in later sections.

3.2. XRD graphs

The XRD patterns (Fig. 1a) of S indicated that quartz (26.90°2θ) was the main mineral component. Chemical treatment reduced the peak intensity of quartz (26.90°2θ) in sewage sludge. It resulted from the diluting effect of additives added to sewage sludge. The appearance of Ca(OH)₂ peaks at various degrees for LS (Fig. 1b) and tricalcium silicate (C₃S) for CS (Fig. 1d) implicated that lime and cement added to the sludges were not exhausted, respectively. The addition of sodium silicate to the LSS and CSS, on the other hand, allowed Ca(OH)₂ and C₃S to be incorporated, as evidenced by the absence of these peaks in Fig. 1c,e. Vail [12], Gowman [13], and Conner [14] have mentioned that sodium silicate contains both polymeric silicates and colloidal silica which can interact with divalent calcium to form silica/silicate gels. Kalousek [15] reported that a reaction between lime, sodium, and water formed Na–C–H–S gels. Thus, the sodium silicate added to the sludge was suggested to interact with Ca(OH)₂ and C₃S, respectively, to form noncrystalline matter.

The CaCO₃ peaks (29.46°2θ) in the XRD graphs (Fig. 3) for chemically-fixed sludges suggested the occurrence of carbonation. These peaks were much more enhanced for LSS and CSS than for LS and CS, indicating that adding sodium silicate resulted in further carbonation of sludges. CaCO₃ is formed through the reaction of hydrated lime or cement hydrate gels with CO₂ in the presence of air. Enhanced carbonation of LSS and CSS was suggested to be due to their porousness, which gave them increased contact area with air. The porousness of LSS and CSS corresponded to

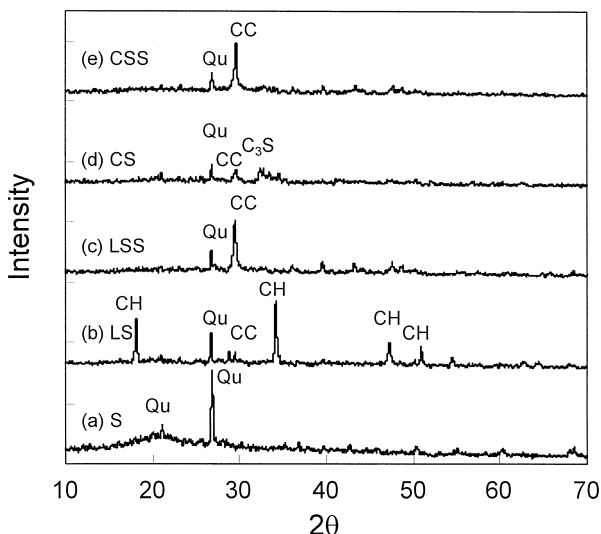


Fig. 1. XRD patterns of: (a) Untreated sewage sludge; (b) Lime-treated sludge; (c) Lime–sodium silicate-treated sludge; (d) Cement-treated sludge; (e) Cement–sodium silicate-treated sludge. * C_3S : tricalcium silicate ($3CaO \cdot SiO_2$); CH: calcium hydrate; CC: calcium carbonate; Qu: quartz (SiO_2); S: untreated sludge; LS: lime-treated sludge; LSS: lime–sodium silicate-treated sludge; CS: cement-treated sludge; CSS: cement–sodium silicate-treated sludge.

their lower density (Table 2), compared to LS and CS, respectively. The pH of LSS and CSS were lower than those of LS and CS, respectively (Table 2), supporting the enhanced carbonation in LSS and CSS. Water film was observed on the surface of LSS and CSS during air drying. This also supported the enhanced occurrence of carbonation concluded above: $Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O, \Delta H_1 = -69 \text{ kJ mol}^{-1}$.

3.3. Fractionation of heavy metals

Sequential extraction indicated that the percentages of Zn, Cu, and Pb of carbonate-bound form in LSS and CSS were greater than those in LS and CS, respectively (Fig. 2a,b,d). This result corresponds to the XRD patterns which show the enhanced peak intensities of $CaCO_3$ for LSS and CSS as discussed earlier.

The percentages of Pb, Cu, Zn, and Cr of organically-bound form in chemically-fixed sludges were consistently lower than those in S (Fig. 2a–d), which was attributed to the irreversible dissolution of organics and organically-bound metals at very high pH during processing and air drying of chemically-fixed sludges. In the sludge after chemically-fixed, Cu shifted to the exchangeable fraction (Fig. 2a), Pb to the oxide-bound fraction (Fig. 2b), and Cr to the residual (LSS and CSS) or oxide-bound (LS and CS) fraction (Fig. 2c). Because little amount of Zn was of organically-bound form in S (21.4%) (Fig. 2d), the shift of Zn from organically-bound fraction to other fractions was the least obvious.

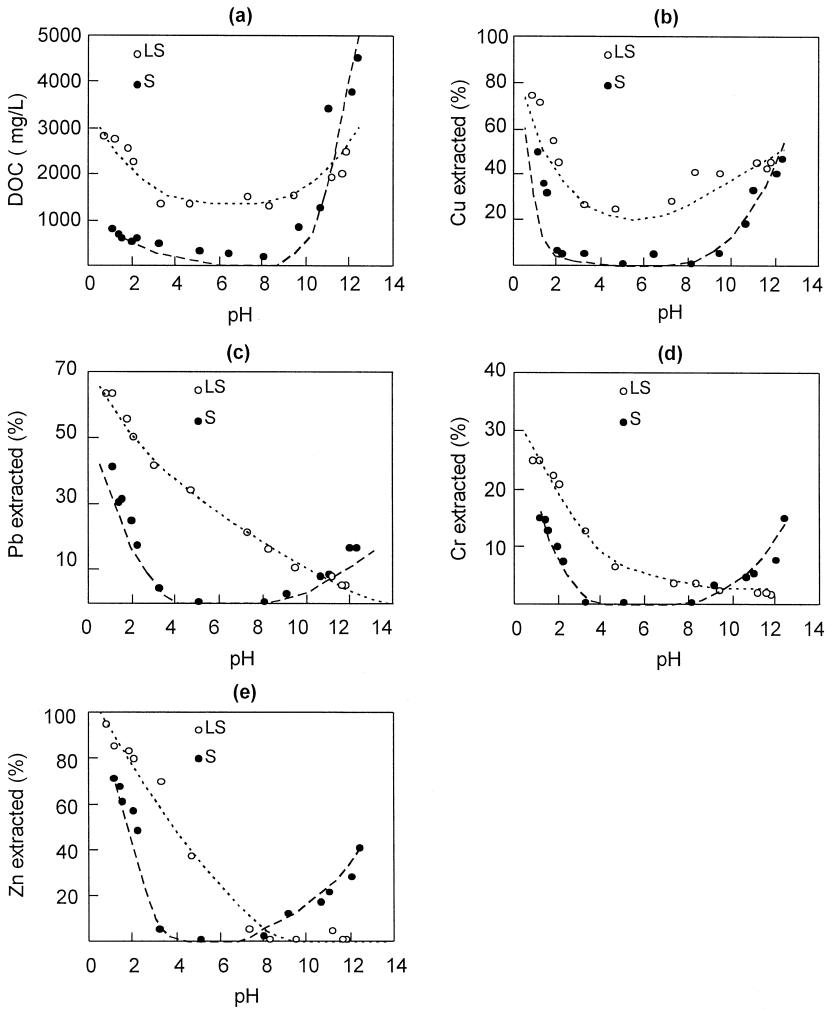


Fig. 2. The results of single extraction conducted at various pH. (a) Dissolved organic carbon (mg l^{-1}) extracted from lime-treated sludge (LS). (b)–(e) Percentages of metals (Cu, Pb, Cr, Zn) extracted from lime-treated sludge (LS) and untreated sludge (S).

The addition of sodium silicate also led to the shifts of Cu, Pb, and Cr from the organically-bound fraction in lime (cement)-treated sludge to the other fractions in lime–sodium silicate (cement–sodium silicate)-treated sludge (Fig. 2a–c). It is assumed that the shift was due to the competition between silica/silicate gels and organics for metals ions.

According to Bishop [16], Cr and Pb can be bound into the silica matrix and become resistant to leaching. The fractionation of Cr in this investigation was the most unique. The addition of sodium silicate led to the shift of Cr from the organically-bound to the residual fraction (Fig. 2c). Sequential extraction in this study indicated that the majority

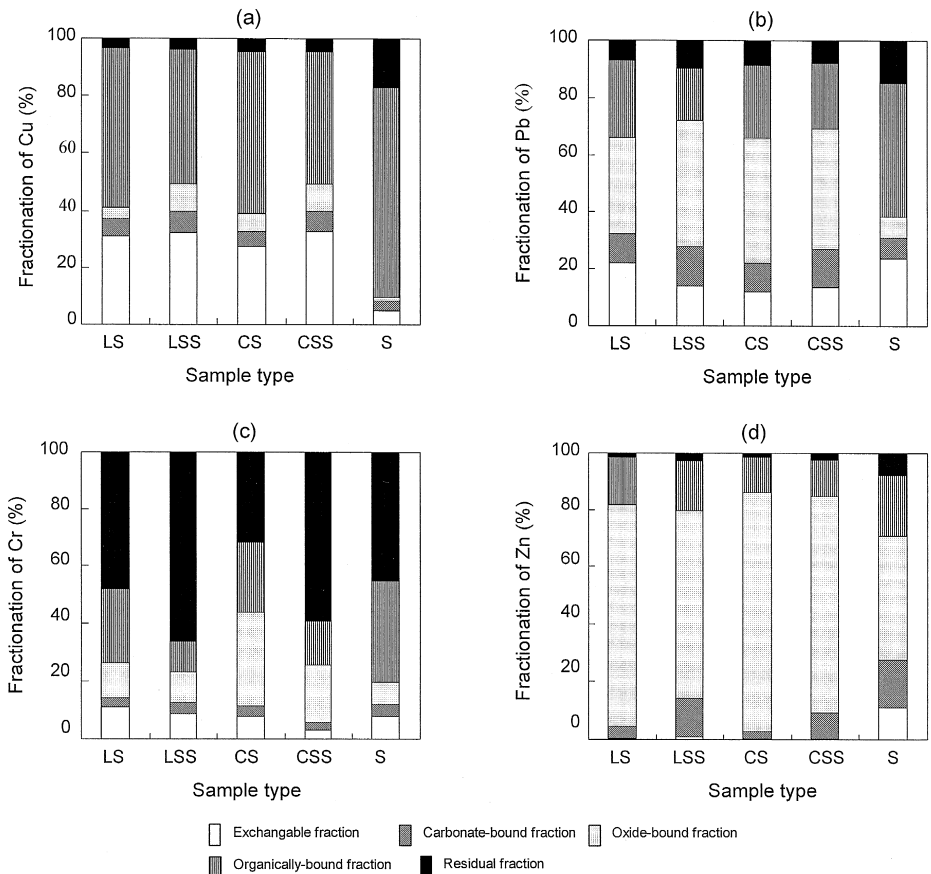


Fig. 3. The fractionations of (a) Cu, (b) Pb, (c) Cr, and (d) Zn in untreated sewage sludge (S) and chemically-fixed sewage sludges, including lime-treated sludge (LS), lime–sodium silicate-treated sludge (LSS), cement-treated sludge (CS), and cement–sodium silicate-treated sludge (CSS).

of Si (> 90%) was of residual form (not presented schematically), which was similar to the report by Tessier et al. [9], suggesting that the above shifted Cr was bound into silicates. Cocke et al. [17] concluded that Cr, in cement-based systems, was different from heavy metals for its size and charge and allowed it to be incorporated throughout the solid material during cement hydration. Bishop [16] reported that Cr could be bound into the silica matrix. Mollah et al. [18] reported that Cr ions substituted for silicon in the C–H–S gels and was resistant to be extracted.

3.4. Single extraction conducted at various pH

The pH strongly affected the extraction of heavy metals from both untreated and chemically-fixed sludges, as shown in Fig. 3. For the objective of simplification, the results for only LS are shown in Fig. 3, since all chemically-fixed sludges investigated gave similar trends. Heavy metals were extracted from chemically-fixed and SS more

than their hydroxides were at $\text{pH} > 5-6$ but less at $\text{pH} < 5-6$ (Fig. 3b–e) using the calculated solubilities of metal hydroxides for comparison (not presented schematically). At high pH, the percentages of heavy metals extracted from S were higher than those from chemically-fixed sludge (Fig. 3b–e). The trend of metals extracted from S was consistent with that of DOC (Fig. 3a) at high pH, indicating that heavy metals were extracted from S in proportion to organics at high pH. The organic content in chemically-fixed sludges was less than that in S, while the DOC extracted from chemically-fixed sludge was much more than that from S (Fig. 3a), suggesting the occurrence of the irreversible dissolution of organics at very high pH during processing and air drying of chemically-fixed sludge. The processing conditions of chemically-fixed sludge were very similar to the extraction conditions of S extracted at very high pH, leading to the enhanced amounts of organics, and some metals (e.g. Cu) having higher affinity to organics, to be extracted. Of the four metals investigated, Cu was extracted from chemically-fixed sludges much more than that was from S because a great amount (73%) of Cu was of organically-bound form, giving the best example of the irreversible dissolution concluded above. Pb and Cr showed similar trends at $\text{pH} < 8$, but to a lesser extent (Fig. 3c–e).

The amounts of heavy metals of organically-bound form in S influenced the amounts of heavy metals extracted from chemically-treated sludges. Regarding the percentages of heavy metals of organically-bound form in S, Cu was the most (73.7%), Zn was the least (21.4%), and Pb and Cr were in-between (46.6% and 35.2%, respectively). Several authors have reported that Cu was mostly distributed in organically-bound fraction [19] and that it has a great affinity for organic ligands [20,21]. Recombining the data of sequential extraction (Fig. 2), we found that the percentages of metals of exchangeable form (1 M MgCl_2) in chemically-fixed sludges depended on their amounts of organically-bound form in S, irrespective of metals species (Fig. 4). Their relationship equation of regression was: $E = 0.5884 \times (\text{OR}) - 12.417$, where E = percentages of metals of exchangeable form (1 M MgCl_2) in chemically-fixed sludges; OR = percentages of

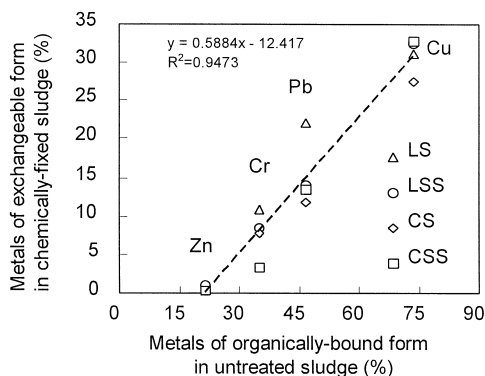


Fig. 4. The relationship between percentages of metals of exchangeable form in chemically-fixed sludges and percentages of metals of organically-bound form in untreated sludge. * S: untreated sludge; LS: lime-treated; LSS: lime-sodium silicate-treated sludge; CS: cement-treated sludge; CSS: cement sodium silicate-treated sludge.

metals of organically-bound form in S, irrespective of metal species. According to this equation, metals (e.g. Cu) become labile in the sludges after chemically-fixed when they have high affinity for organics in S, since the exchangeable form of metal is the most labile. On the other hand, metals (e.g. Zn) become stable in the sludges after chemically-fixed when they have low affinity for organics in S. This effect was suggested to be attributed to the irreversible dissolution of organics, and the heavy metals having higher affinity for organics, at very high pH during processing and air drying of chemically-fixed sludges, as concluded early.

Although the objectives of the works introduced by other authors differed from this study, they can still help us explain the effects of chemical processing on the extractability of metals in sewage sludge. Conner [14] stated that the cement-based organic immobilization process, also a kind of chemical treatment, of organic wastes can increase the leaching of heavy metals if the pH of the process exceeds the normal range. The experiments of Sims [22] and Little et al. [23] showed an increasing plant uptake of Cu, but a decreasing plant uptake of Zn, according to increasing liming rate to the soils amended with lime and with composted sewage sludge + lime, respectively. The experiments of Stehouwer et al. [24] showed that leachate concentration of Zn decreased with increasing alkaline by-product amendment + sewage sludge added to soils, but that of Cu increased. Some other authors have reported that liming increased the extractabilities of some metals in soils [5,25]. Organic matter is usually alkali-extractable. Many authors, such as Stover et al. [6] and Sposito et al. [26], have extracted organically-bound heavy metals from sewage sludge with NaOH as an extractant. Leita and Nobili [27] mentioned that the higher solubility of heavy metals at alkaline pH was attributed to the enhanced formation of organic matter–metal complexes.

4. Conclusions

The data of this study indicated that heavy metals (e.g. Cu) with higher affinity to organics in sewage sludge become unstable in the sludge after chemically-fixed. This explains why the order (Cu > Pb > Cr > Zn) of the percentages of exchangeable metals in chemically-fixed sludges was the same as that of organically-bound metals in S. This effect was thought to be attributed to the irreversible dissolution of organically-bound metals at very high pH during processing and air drying of chemically-fixed sludges. The processing conditions of chemically-fixed sludges were very similar to the extraction condition of S extracted at very high pH, leading to strong amounts of organics, and the metals having higher affinity for organics, to be extracted. While some metals (e.g. Zn) become more stable in the sludge after chemically-fixed, resulting from their less affinity for organics.

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