Solubility of heavy metals added to MSW

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ABSTRACT

This paper aims to investigate the six heavy metal levels (Cd, Cr, Cu, Pb, Ni and Zn) in municipal solid waste (MSW) at different pHs. It intends to provide the baseline information of metals solubility in MSW co-disposed or co-digested with MSW incinerator ashes in landfill or anaerobic bioreactors or heavy metals contaminated in anaerobic digesters. One milliliter (equal to 1 mg) of each metal was added to the 100 ml MSW and the batch reactor test was carried out. The results showed that higher HNO_3 and NaOH were consumed at extreme pH of 1 and 13 compared to those from pH 2 to 11 due to the comparably higher buffer capacity. Pb was found to have the least soluble level, highest metal adsorption (%) and highest partitioning K_d ($l\,g^{-1}$) between pH 3 and 12. In contrast, Ni showed the highest soluble level, lowest metal adsorption (%) and lowest K_d ($l\,g^{-1}$) between pH 4 and 12. Except Ni and Cr, other four metals seemed to show the amphibious properties as comparative higher solubility was found in the acidic and basic conditions.

1. Introduction

Municipal solid waste (MSW) has been mostly treated with incineration accompanying with partly landfill disposal in Taiwan. MSW incinerator (MSWI) has the advantage of reducing MSW weight and volume while gaining the steam and energy recovery. However, residues such as bottom and fly ash generated in the incineration process still remain an environmental problem and need further treatment to prevent secondary pollution. Residues have been reused in several purposes such as aggregate, backfill, soil amendment and geotechnical application [1–6]. However, using residues as landfill cover have been a potential and particularly an aggressive option for fly ash. Specifically, the released heavy metals in the co-disposal or co-digestion process may affect the landfill and anaerobic bioreactors performance [7–15]. Further, released heavy metals and other hazardous materials have the potential to cause risk to human health and the ecological environment [16–20]. Therefore, heavy metal release and distribution in MSW is the key concern in landfill or anaerobic digester co-disposing or co-digesting MSW with MSWI ash or MSW anaerobic bioreactor containing possibly influential levels of heavy metals.

Metal levels and uptake in different media such as soil, sludge, forest, river, MSW, leachate, groundwater and landfill, etc. were reported by several researchers [21–34]. Partitioning of metals in different media, pH and organic matter was also investigated and its potential effects on the environment were presented. However, research regarding heavy metals distribution in MSW at different pHs was few [27,28]. Lo and Liao [7] and Lo [8] have presented the likely metal release in MSW co-disposed or co-digested with MSWI ash. In these reports, more than 30 metals ions were found in the released leachate. Thus, the effect of individual metal on the MSW digestion needs to be understood because of the possible synergistic or antagonistic effect by mixed metals. Therefore, this study aims to investigate the individual metal addition and its adsorption and solubility in MSW particularly focusing on the six heavy metals that may provide baseline information for MSW digestion in landfill or anaerobic bioreactor.

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2. Materials and methods

2.1. MSW and metal stock solution

This study aims to investigate the heavy metals adsorption and solubility in MSW. Thus, synthetic MSW typical of organic fraction was made with office paper (30%), newspaper (35%), hay (30%) and food waste (5%). Chemical constituents of C, H, O, N, etc. was analyzed by elemental analyzer (elementar vario EL III) and was found to be approximately ~46, ~6, ~41, ~1.4 and ~5.6%, respectively. In order to obtain the results quickly and without interference, MSW of dry basis was cut into pieces of ~5 mm and was blended with distilled water to get a high water content of total solid (TS) 6% of MSW. This TS content is typical of MSW treatment of anaerobic bioreactor [35,36] or similar to landfill sites with saturated water content that is easily for metal ions diffusion and microbial attack.

The added six heavy metals stock solution of 1000 mg l^{-1} were purchased from Merck Com. In these levels, 1 ml solution is equal to 1 mg metal content.

2.2. Experimental

This study aims to investigate the pH effect on the solubility of added heavy metals in MSW. The total content of added heavy metals of Cd, Cr, Cu, Ni, Pb and Zn was 1 ml (1 ml equal to 1 mg). One milliliter metal solution was added into 100 ml MSW substrate in 200 ml beaker leading to a total level of 10 mg l^{-1}. Thereafter, NaOH or HNO$_3$ with different normality was added and stirred completely with MSW to adjust the pH from 1 to 13. The pHs in MSW suspension were occasionally measured with a pH meter (pH330i) to ensure the desired exact pH values. The blank pH of MSW suspension without metal addition was measured to be 6.77 ± 0.21. The added quantity in mequiv. H$^+$ or mequiv. OH$^-$ was recorded and plotted as a function of pH. Further, MSW substrate in each beaker with adjusted different pH was filtered by 0.45 μm filter membrane with vacuum pump and the filtrate was analyzed with ICP-OES (Thermal Electron Corp.). Soluble and calculated adsorbed metals in MSW substrate versus pH were plotted. Similarly, control batch reactor test without metal addition was carried out for comparison.

2.3. Theoretical definition

In theory, metal hydroxides formation can be described as follows [24]:

$$M^{z+} + n(OH)^{-1} = M_z(OH)_n^{(z-n)} \tag{1}$$

$$[M_z(OH)_n^{(z-n)}] = \beta_{(OH)n}[OH^{-1}][M^{z+}], \qquad n = 1, 2, \ldots \tag{2}$$
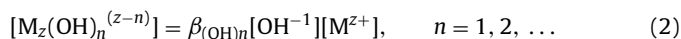
where $M_z(OH)_n^{(z-n)}$ and M^{z+} are metal hydroxide and free metal ion, respectively; $\beta_{(OH)n}$ is overall formation constant for metal hydroxide and n is the number of hydroxide ions associated with each metal ion.

In addition, solubility of metal oxides and hydroxides can be described as follows [21]:

$$p[M^{z+}] = -\log[M^{z+}] = -\log{}^cK_{so} - zpK_w + zpH \tag{3}$$

$${}^cK_{so} = [M^{z+}][OH^-]^z \tag{4}$$
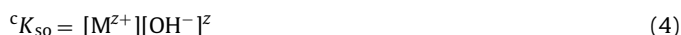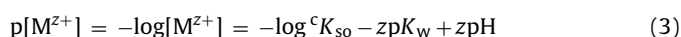
where M^{z+} is free metal ion and ${}^cK_{so}$ is conventional solubility product based on the levels of individual metal ion.

The above equations can be used to predict the free metal level and checked with experimental data analyzed by ICP-OES. However, metal adsorption (surface complex formation) onto MSW can

be calculated by the following definition:

$$M_{\text{adsorbed}(\%)} = \frac{M_{\text{total}} - M_{\text{solubility}}}{M_{\text{total}}} \times 100 \tag{5}$$

where $M_{\text{adsorbed}(\%)}$ is the adsorbed metal (%), M_{total} is the total metal content within MSW volume (mg l^{-1}) and $M_{\text{solubility}}$ is the metal solubility (metal ion levels) in MSW (mg l^{-1}). This definition is general and similar to $R = \{SOM^+\}/M_T$ derived by Wang et al. [23,24], where R, SOM^+ and M_T are ratios of adsorbed metal, metal–sludge (metal–MSW in this study) complex and total metal concentrations, respectively. M_T is the summation of adsorbed metal and free metal ion (M^{z+}).

Other expression of metal binding in mg g^{-1} TS can be calculated as below:

$$M_{\text{adsorbed}(mg/g\,TS)} = \frac{(M_{\text{total}} - M_{\text{solubility}}) \times V}{TS_{\text{MSW}}} \tag{6}$$

where $M_{\text{adsorbed}(mg/g\,TS)}$ is the adsorbed metal in MSW (mg g^{-1} TS), M_{total} is the total metal content within MSW volume (mg l^{-1}), $M_{\text{solubility}}$ is the metal solubility (metal ions levels) in MSW (mg l^{-1}), V is the working volume of MSW (l) and TS_{MSW} is total solid (TS) of MSW (g).

Through metal adsorption and free metal ions, metal partitioning can be evaluated by K_d coefficient (l kg^{-1}) which is the ratio of metal adsorption in solid phase such as soil over the concentration in soil solution [25]. K_d coefficient can be used in the similar condition with the replacement of soil by sludge or MSW and can be expressed as follow:

$$K_d = \frac{M_{\text{adsorbed}(mg/g\,TS)}}{M_{\text{solubility}(mg/l)}} \tag{7}$$

where $M_{\text{adsorbed}(mg/g\,TS)}$ is the adsorbed metal in MSW (mg g^{-1} TS) and $M_{\text{solubility}(mg/l)}$ is the metal solubility in MSW (mg l^{-1}).

Using the above equations and definitions, metal solubility, metal adsorption and K_d values in MSW over different pHs can be obtained.

3. Results and discussion

3.1. Acids or base addition as a function of pH

In this study, MSW with high water content (94%) similar to sludge (TS 6%) was used for batch reactor test by adding different quantity of NaOH or HNO$_3$. The results showed that pH increased as NaOH addition increased and pH decreased as HNO$_3$ addition increased. HNO$_3$ addition as mequiv. H$^+$ l^{-1} from pH 7 to 2 was found to increase linearly from 0 to ~30–60 for six heavy metals, respectively. In a similar way, NaOH added batch reactors from pH 7 to 12 increased from 0 to ~40–90 mequiv. OH$^-$ l^{-1} for six heavy metals, respectively. These trends are shown in Fig. 1. In addition, it is noted that HNO$_3$ or NaOH addition were found higher at extreme pH 1 and 13 than that at pH between 2 and 12 shown in Fig. 2. In particular, HNO$_3$ of ~320 mequiv. H$^+$ l^{-1} and NaOH of ~620 mequiv. OH$^-$ l^{-1} were added to reach pH 1 and pH 13 for Pb and Zn and Cd, respectively. These phenomena were due to the higher buffer capacity ($\beta = dV/dpH$ or d mequiv./dpH) found in the extreme pHs similar to the investigation [37].

3.2. Heavy metals solubility

Six heavy metals concentrations in MSW with metal addition showed that Cu, Zn, Pb and Cd had amphibious properties while that of Cr and Ni was not significant (Fig. 3). Metal levels appeared to increase as pH decreased. Soluble levels of Ni ion were found the highest while those of Pb ions were found the least as compared to
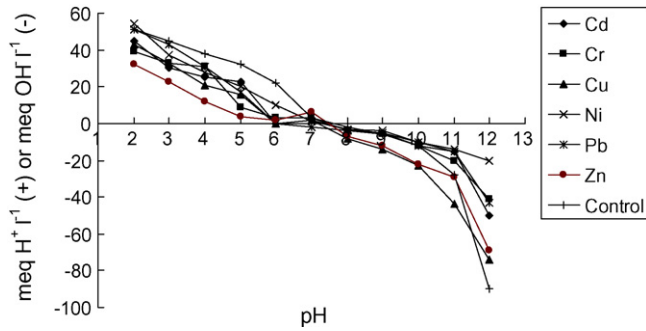
Fig. 1. HNO$_3$ (mequiv. H$^+$ l^{-1}) or NaOH (mequiv. OH$^-$ l^{-1}) needed to reach the pHs from 2 to 12 (Cd: ♦; Cr: ■; Cu: ▲; Ni: ×; Pb: ✱; Zn: ●; control: +).
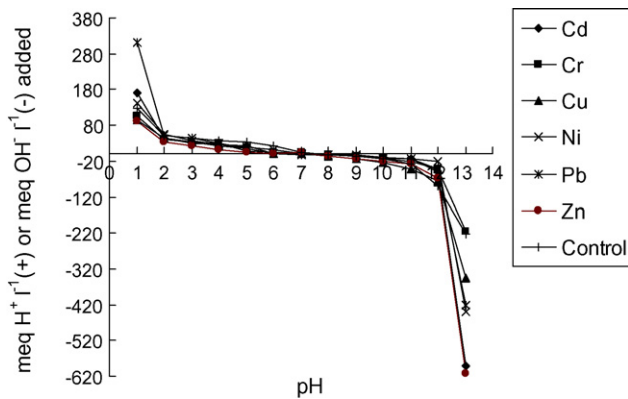
Fig. 2. HNO$_3$ (mequiv. H$^+$ l^{-1}) or NaOH (mequiv. OH$^-$ l^{-1}) needed to reach the pHs from 1 to 13 (Cd: ♦; Cr: ■; Cu: ▲; Ni: ×; Pb: ✱; Zn: ●; control: +).

other four heavy metals from pH 4 to 12. At extreme pH 1, Zn, Pb and Cd ions showed higher levels than those of Ni, Cu and Cr. However, Cu, Ni and Zn ions levels were found higher compared to those of Pb, Cd and Cr at extreme pH 13. Metal ions levels showed the order of Ni > Cu > Cr > Zn > Cd > Pb between pH 8 and 12. In other pH ranges, metal ions varied with pH as can been seen in Fig. 3. Results also indicated that Ni ion was the least adsorbable metal. This result was similar to the findings by Wang et al. [23].

Six heavy metals ion levels in MSW without metal addition showed the amphibious properties as listed in Fig. 4. Zn and Pb ions appeared to have higher levels compared to other four heavy metals. Higher levels of Zn and Pb ions might come from the MSW component that contained a comparatively higher content. In addition, Zn and Pb seemed to show a very similar concentration trend in MSW for both with and without metal addition.
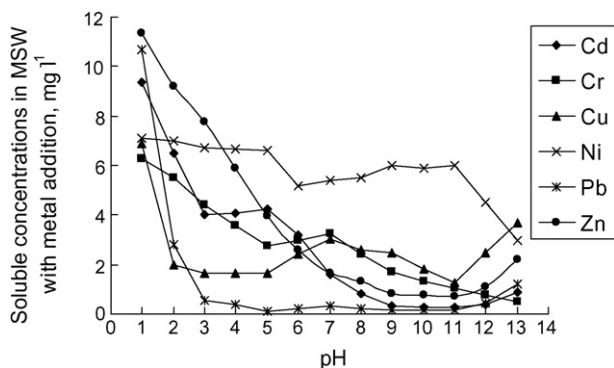

Fig. 3. Six heavy metal ions levels in MSW substrate at different pHs with 10 mg l^{-1} addition of individual metal (Cd: ♦; Cr: ■; Cu: ▲; Ni: ×; Pb: ✱; Zn: ●).
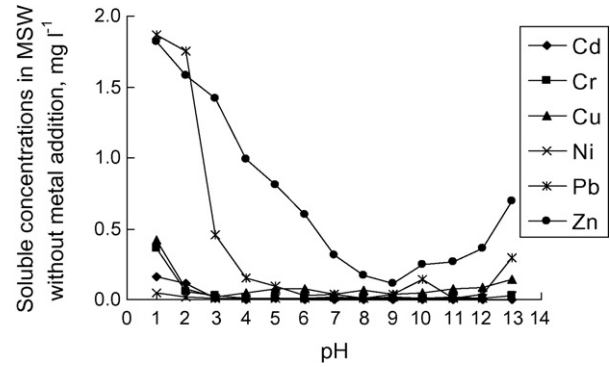
Fig. 4. Six heavy metal ions levels in MSW substrate at different pHs without metal addition (Cd: ♦; Cr: ■; Cu: ▲; Ni: ×; Pb: ✱; Zn: ●).
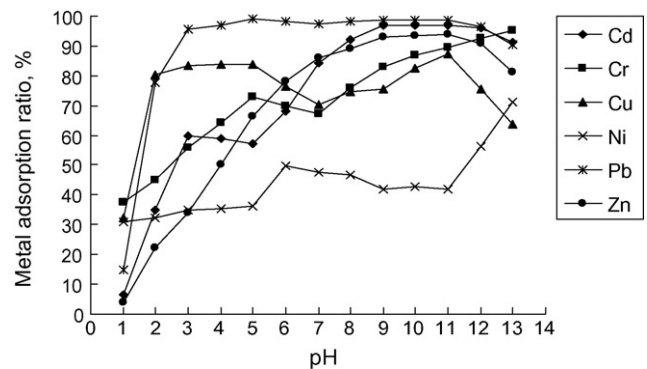
Fig. 5. The adsorption ratios (%) of six heavy metals at different pHs (Cd: ♦; Cr: ■; Cu: ▲; Ni: ×; Pb: ✱; Zn: ●).

3.3. Heavy metals adsorption

Metal adsorption appeared to show the complementary results with metal free ion level. Thus, the higher the Ni ion indicated that the lower the Ni adsorption in MSW (Fig. 5). Similarly, the least Pb ion showed the highest adsorption ratio (%) among the six heavy metals. This is because the total metal content added is 1 mg (10 mg l^{-1} in 100 ml MSW), then metal adsorbed in solid phase and metal dissolved in liquid was maintained constant with mass balance (1 mg plus MSW background content). Therefore, the metal ions concentration versus metal adsorption showed the complementary trends. It was noted that metal adsorption ratios exceeded around 60% for five metals for pH higher than 4 except for metal Ni. Ni adsorption ratios were found lower between ~30 and ~60% for all pH ranges. In addition, Pb showed higher adsorption ratios more than 90% between pH 3 and 13. In general, five heavy metals except Ni showed higher adsorption ratios of more than 70% between pH 7 and 12. Pb showed the highest adsorption higher than ~0.19 mg g^{-1} TS between pH 3 and 13 (Fig. 6). The adsorbed order of six heavy metals was found to be Pb > Zn > Cd > Cr > Cu > Ni in the pH range from 7 to 12. The adsorbed metals increases as pH increased from pH 1 to 11. Metal adsorption of Cd, Cu, Pb and Zn showed a slightly decrease for pH from 12 to 13 indicating the amphibious properties that metal solubility increased as pH increased.

3.4. Heavy metals partitioning

Pb showed the highest metal partitioning compared to other five heavy metals. Metal partitioning of Pb was found between 0.003 and 1.711 l g^{-1}. It was also noted that metal partitioning of six heavy metals increased as pH increased particularly found in the
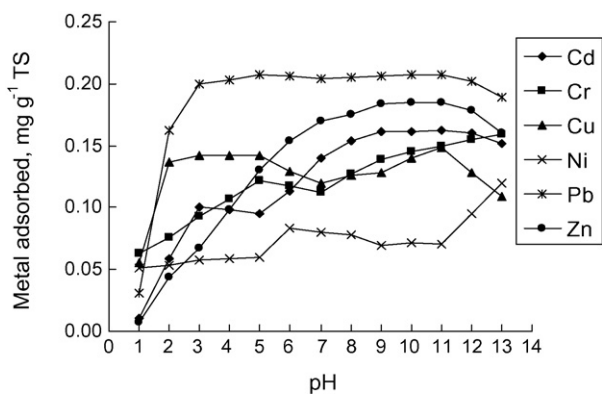
Fig. 6. The adsorbed metals (mg g^{-1} TS) of six heavy metals in MSW at different pHs (Cd: ♦; Cr: ■; Cu: ▲; Ni: ×; Pb: ✶; Zn: ●).
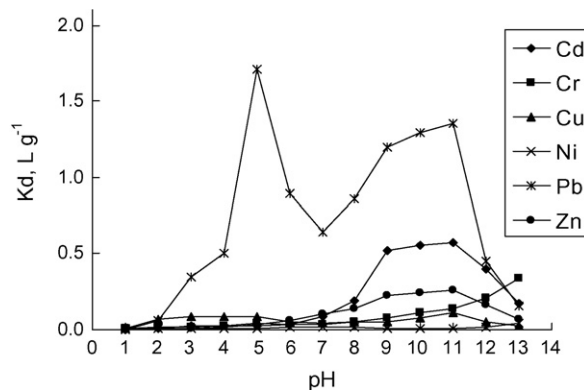
Fig. 7. The K_d values of six heavy metals at different pHs (Cd: ♦; Cr: ■; Cu: ▲; Ni: ×; Pb: ✶; Zn: ●).

pHs higher than 7 (Fig. 7). However, K_d values showed a decrease from pH 12 to 13 except Cr. According to the K_d definition, higher metal adsorption and lower metal solubility would cause the higher metal partitioning. This leads the result that Pb had higher K_d values than other five metals. Similar results were also found in Cd, Zn and Cr. These higher K_d values observed in higher pH was attributed to the higher metal adsorption and lower metal solubility. Metal adsorption versus metal solubility showed the different distribution patterns for six heavy metals. Pb and Zn were mostly found to have similar distribution patterns with slightly wider adsorption versus lower solubility while that of Ni was found narrower (Fig. 8.). In addition, Cr and Cu showed the similar distribution patterns.

3.5. Summary and implication

Metal solubility, distribution and adsorption in different media such as soil, sludge, river sediment, ground water, leachate and MSW were investigated by several researchers. These researches

indicated that environmental factors such as pH, dissolved organic matter, metals competition, adsorbent characteristics and the like might affect the metal adsorption [6,21,23–30,32]. In general, higher pH showed higher metal adsorption and higher K_d [23,25]. Metal adsorption also increased as the dissolved organic compounds increased [28]. Metal distribution was affected by the co-existing metals and different types of anions such as OH^{1-}, CO_3^{2-}, SO_4^{2-}, Cl^{1-}, NH_3-H^+, HCO_3^{1-}, etc. [22,23,29,30,38]. Except theoretical calculation, metal ion level can also be predicted by several models such as MINTEQA2 and WHAM model database [28], MINEQL$^+$ Chemical Equilibrium Program Version 3.0 [38], multivariate regression [31], Langmuir and Freundlich model [33] and the like.

Among the reported investigations, least adsorption of Ni [23] agreed with the results of this study. The results of Wang et al. [23] showed that higher sludge concentration at different wastewater plants exerted comparatively higher metal adsorption. Adsorption of Pb (5–103.7 mg g^{-1}), Cd (0.227–40.5 mg g^{-1}) and Cu (2.95–32.15 mg g^{-1}) by different adsorbents, pH and initial concen-

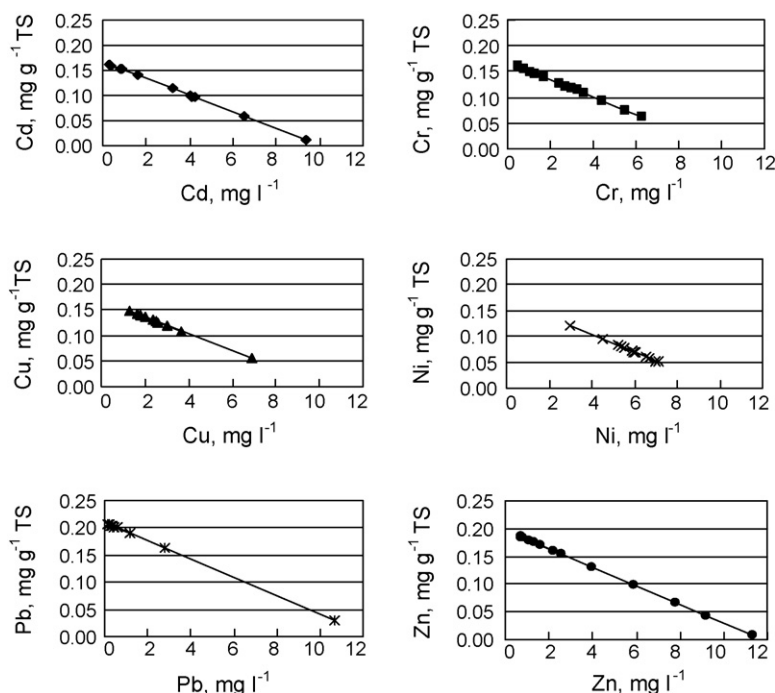**Fig. 8.** The distribution patterns of adsorbed metals vs. metal ions levels of six heavy metals with 13 different pHs (Cd: ♦; Cr: ■; Cu: ▲; Ni: ×; Pb: ✶; Zn: ●).

tration has been extensively reported [33]. The reported adsorbed metals were found higher than this study indicating MSW adsorption capacity was lower than these adsorbents such as peat, red mud, electric furnace slag and manganese nodules, etc. As expected, initial metal concentration ($\sim$10–100 mg l^{-1}) could affect the metal adsorption [33]. This phenomenon revealed that metal adsorption was found lower with initial metal levels of 10 mg l^{-1} in this investigation compared to the report by Das and Jana [33]. Sorption order of maximum uptake of Cd > Pb > Ni > Cu by Foil et al. [34] and Fe > Zn > Cu > Ni > Cd by Erses et al. [27] also showed the different results compared to this study of Pb > Zn $\approx$ Cr > Cd > Cu > Ni. Metal solubility and adsorption can also be affected by the stepwise formation constants [39] and functional groups such as carboxylic acids [28]; sulfhydryl groups, amino, carboxylate, imidazole and hydroxyl radicals of enzymes and other proteins [40]; amines, sulfate groups and carboxylic groups [41] and seven groups such as mercapto groups, uptake inside bacterial cells, carboxlic groups, phosphate and the like [42] therefore formed the different K_d values for different metals at each different pH. It was further noted that metal could be adsorbed in dissolved organic matter such as humic and fulvic acids [28] or solid matrix such as waste and sludge [23,27,34]. Thus, the metal adsorption constant (stability constant of metal–matrix complex) and K_d value with detailed continuation research could be used to explain the metal adsorption scenarios in different environmental conditions such as pH, dissolved organic matter, adsorbent characteristics, and metal ions competition. In addition, these results can provide the baseline information of heavy metal levels for co-disposal or co-digestion process in landfill or anaerobic digester.

4. Conclusions

Soluble concentrations and adsorption of six heavy metals (Cd, Cr, Cu, Pb, Ni and Zn) in MSW with different pHs were investigated. Results showed that higher HNO$_3$ and NaOH were needed to reach to the extreme pH of 1 and 13 compared to that from pH 2 to 11 due to the comparable higher buffer capacity. Pb was found to have the least soluble level, highest metal adsorption (%) and highest partition K_d (l g^{-1}) between pH 3 and 12. In contrast, Ni showed the highest soluble concentration, lowest metal adsorption (%) and lowest K_d (l g^{-1}) between pH 4 and 12. Results also showed that wider distribution patterns of metal adsorption versus soluble concentration were observed in Pb and Zn while that was found narrower in Ni. Except for metal Ni, other five metals seemed to show the amphibious properties as comparatively higher soluble concentration was found in the rather acidic and basic pHs.

Acknowledgments

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.jhazmat.2008.03.119.

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