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Removal behaviors of Cu and Pb from heavy metal contaminated silts flushed by citric acid

Yan Wang[1a], Lin Tang[*2] and Likui Liu[3]

[1]School of Civil and Environmental Engineering, Ningbo University, Ningbo 315211, China
[2]School of Ocean Engineering, Harbin Institute of Technology at Weihai, Weihai 264209, China
[3]China Construction Third Bureau Engineering Design Co. Ltd, Wuhan 430074, China

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Abstract. A range of Pb(II) and Cu(II) contaminated silt columns were prepared to simulate heavy metal contaminated site, and citric acid was employed to flush the silt columns. The concentration of citric acid, flushing time and concentrations of Cu(II) and Pb(II) were considered to study the removal behaviors of heavy metals. The removal efficiency of Cu(II) was found much better than that of Pb(II). The total removal ratio of Cu(II) could reach 59.9-73.4% when using 0.005mol/L citric acid, whereas the removal efficiency decreased with increasing the concentration of citric acid because higher concentration of chelating agent could lead to decrease of permeability in soil. The removal efficiency of Pb(II) was not as good as Cu(II), with the maximum removal ratio only 16.7%, and higher citric acid concentration led to higher removal efficiency, because lower concentration of citric acid could be adsorbed on soil surface and caused inhibitory effect on Pb(II) removing. The removal ratio of Cu(II) was greater when the initial Cu(II) concentration was lower in the contaminated soil, however, the same 0.005 mol/L concentration of citric acid was not effective to remove Pb(II), and the removal behaviors of Cu(II) and Pb(II) from contaminated silts were rather different. Therefore, appropriate concentration of citric acid should be carefully chosen to flush the heavy metal contaminated site, and this study can provide some theoretical basis for remediating heavy metal contaminated site by soil flushing.

Keywords: citric acid; contaminated silt; flushing; heavy metal; removal

1. Introduction

Nowadays an increasing amount of soil has been contaminated with the rapid development of global economy in modern society. During the past few years, soil pollution has been a focus point and many researchers have paid much attention to this environmental problem(Gusiatin and Klimiuk 2012, Sriraam *et al.* 2019, Ghiyas and Bagheripour 2020). Once polluted by inorganic pollutants especially heavy metals, the soil is hard to be remediated (Maity *et al.* 2013). The heavy metals can enter the soil environment through both non-anthropogenic and anthropogenic behaviors (Torres *et al.* 2012). Among the inorganic pollutants, lead and copper are encountered very frequently and are quite toxic. Therefore, effective treating technology is urgently needed to solve this problem. Lead can stay for a very long period in the soil without degradation and migration, and can generate potential toxic effect even at rather low concentrations (Freitas *et al.* 2013). When the concentration of copper in the soil is above the allowable value, it may have phytotoxic effects (Navarro *et al.* 2011, Zaheer *et al.* 2015), and be environmental threat (Kos and Leštan 2004).

As an in-situ remediation method to treat heavy metal contaminated soil, soil flushing has gained broad attention throughout the world. Some excellent extractants which are helpful to leach heavy metals from contaminated soils have been proved very effective (Khalid *et al.* 2017). In order to facilitate the process of heavy metals mobilizing and removing from contaminated soils, a lot of reagents have been carefully studied. Ethylenediaminetetraacetic acid (EDTA), a chelating agent, is considered one of the most efficient reagent for soil washing. EDTA has very strong chelating ability for a wide range of heavy metals, and does nearly no harm to the properties of soils compared with inorganic acids (Deng *et al.* 2017, Qiao *et al.* 2017). However, EDTA has the risk of leading to the deterioration of soil functions and the contamination of groundwater because of poor bio-degradability and high persistence in the soil environment. (Nowack 2002, Guo *et al.* 2016, Suanon *et al.* 2016, Jelusic and Lestan 2014). Biodegradable flushing reagents are then taken into account to replace the nondegradable extractants. The low molecule weight organic acids, for instance, citric acid, is reported to be a promising replacement due to the biodegradability, the effectiveness of mobilizing heavy metals and environmental friendliness (Di Palma and Mecozzi 2007, Freitas *et al.* 2013, Zhang *et al.* 2017, Zaleckas *et al.* 2013).

As a nontoxic tricarboxylic acid, citric acid can easily form strong complexes with heavy metals over a wide pH

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[*]Corresponding author, Assistant Professor
E-mail: tanya_tang3@hit.edu.cn
[a]Professor
E-mail: wangyan@nbu.edu.cn
[b]Assistant Professor

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range (Lenhart *et al.* 2000), which may make it a good reagent to flush the contaminated soil. Citric acid has been found very effective in phytoextraction of soil contaminated by heavy metals by facilitating the mobilization of heavy metals in soil (Chen *et al.* 2003, Meers *et al.* 2005, Lesage *et al.* 2005, Aderholt *et al.* 2017, Muhammad *et al.* 2009).

Also, batch experiments of soil washing in laboratory suggest the effectiveness of citric acid as a washing reagent to remove heavy metals in contaminated soil (Jean *et al.* 2007, Liang *et al.* 2019, Ke *et al.* 2020). However, studies on behaviors of heavy metal contaminated soil flushed by citric acid are very limited (Bassi and Prosher 2000, Jean-Soro *et al.* 2012, Perez-Esteban *et al.* 2013, Cheng *et al.* 2011, Ash *et al.* 2016), accordingly, it is now not easy to apply this technique in situ, and it is necessary to further study the effect of citric acid on the removal behavior of heavy metals from contaminated soil.

In this study, the contaminated silts were filled into the columns and then were compacted to simulate the contaminated site, and citric acid with different concentrations were used to leach different concentrations of Cu(II) and Pb(II) contaminated soil. The removal behaviors of Cu(II) and Pb(II) were systemically analyzed and the removal mechanisms were discussed.

2. Materials and methods

2.1 Preparation of samples

The silt samples used in this study were taken from the nearby area of Qiantang River, in Hangzhou city. The silts were dried in an oven at temperature of 105oC for 24h to lose moisture, and passed through a sieve with the pore diameter of 2 mm. Then, the basic properties of the soil were obtained through a series of laboratory tests, and Table 1 shows the basic parameters of the silts. According to the plastic and liquid limits, the soil is identified as silt. The chemical reagents utilized in this study include citric acid (CA), $CuCl_2 . 2H_2O$, $PbCl_2$, and all are analytical pure reagents.

Table 1 Basic parameters of silt samples

Parameters	Value
Dry Density	1.53 g.cm^{-3}
Maximum Dry Density	1.61 g.cm^{-3}
Optimum Moisture Content	16%
Liquid Limit	34%
Plastic Limit	26.3%
pH	8.8

The heavy metal contaminated silts in this study were prepared in the laboratory. At first, a certain amount of dry silts were accurately weighed, and specific amount of $CuCl_2 . 2H_2O$ or $PbCl_2$ was dissolved by deionized water. Then, the solutions of heavy metal copper or lead were added to the dry silts and the samples were stirred evenly. Based on this method, three different concentrations (i.e., 100, 200 and 400 mg/kg) of heavy metals (i.e., copper and lead) contaminated silts were prepared. The moisture content in the contaminated silts was 16% which is the optimal water content obtained by proctor compaction test. All the prepared samples of contaminated silts were stored in sealed bags for 7 days. A certain amount of different concentrations (i.e., 0.005, 0.015 and 0.03 mol/L) of citric acid were also prepared in stock.

2.2 Contaminated silt columns flushed by citric acid

The prepared contaminated silts were filled into a series of columns to simulate virtual heavy metal contaminated site. The weight of silt samples were determined according to the maximum dry density and the size of the column, and then were divided into three equal parts. Each part of the samples were filled into the column and were compacted well, which means the whole silt column was compacted in three layers. Finally, several groups of compacted silt columns were prepared, and the dry density of all the silt columns was 1.53 g/cm^3 through calculation, which was 95% of the maximum dry density (i.e., 1.61 g/cm^3). The inner diameter of the column was 5 cm, and the length was 5 cm.

All the silt columns were leached by deionized water to make sure the air in the pores of the soils were eliminated and the soils were in a state of saturation before leaching with citric acid. For all the tests, at the time of the first two pore volumes, deionized water was used to flush the silt columnsto remove free ions in the soil pores. The flushing agent citric acid solutions were stored in markov bottles, which can help to keep a stable waterhead during the tests. The flushing agent solution flowed through the silt column from the bottom to the top. The leachates from the outlet were collected at certain intervals, and the concentrations of Cu(II) and Pb(II) were detected by the atomic absorption spectrophotometry.

3. Results and discussion

3.1 Cu(II) contaminated silts flushed by different concentrations of citric acid

Fig. 1 shows the variation of Cu(II) concentration in leachate from 200 mg/kg Cu(II) contaminated silts flushed with different concentrations of citric acid. At the time of the first two pore volumes, deionized water was used to flush the silt columns. As can be seen in the figure, there is a small peak concentration, indicating that just a very small number of heavy metals could be removed by deionized water. In detail, as leached by different concentrations of citric acid, the concentration of Cu(II) in leachate grew rather rapidly with the rise of pore volumes at first, and reached the maximum at about 3 to 4 pore volumes, then as the time elapsed, the concentration began to drop and reached the bottom. The peak value of the concentration of Cu(II) in leachate under different citric acid was 93.3, 151.0 and 170.5 mg/L when using citric acid of 0.005, 0.015 and 0.03 mol/L, respectively. As for the soil flushed by 0.03 mol/L citric acid, it terminated at about the fourth pore

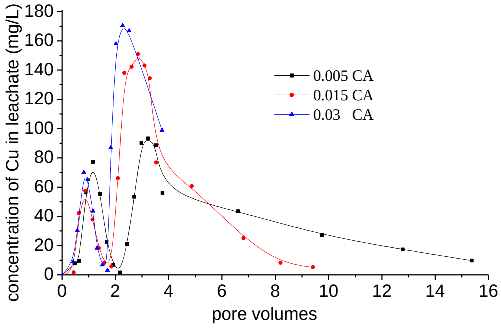

Fig. 1 Variation of Cu(II) concentration in leachate from 200 mg/kg Cu(II) contaminated silt soil flushed with different concentrations of citric acid

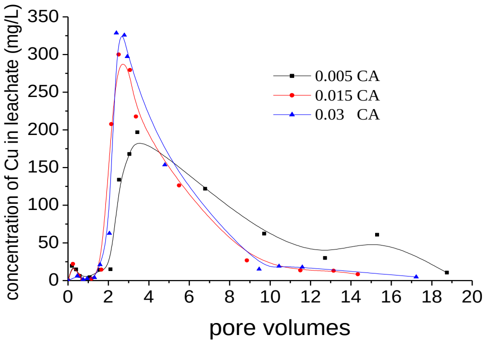

Fig. 2 Variation of Cu(II) concentration in leachate from 400 mg/kg Cu(II) contaminated silt soil flushed with different concentrations of citric acid

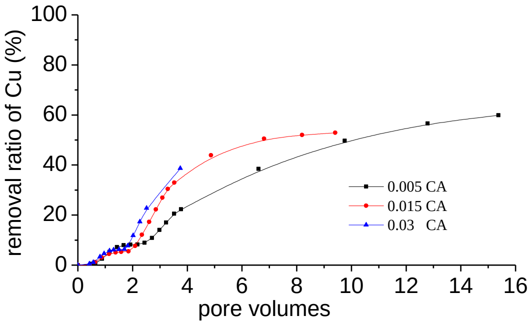

Fig. 3 Removal ratio of Cu(II) from 200 mg/kg Cu(II) contaminated silt soil flushed with different concentrations of citric acid

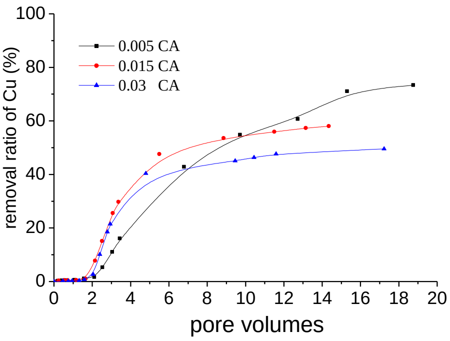

Fig. 4 Removal ratio of Cu(II) from 400 mg/kg Cu(II) contaminated silt soil flushed with different concentrations of citric acid

volume because of very low permeability. Hydraulic conductivity can easily change when the physical or chemical properties of soil change (Gao *et al.* 2021). During the experiment, the hydraulic conductivity of the soil columns were found to decrease as increasing the concentration of citric acid, and this phenomenon was also reported in other researchers work. (Mancini *et al.* 2011) and (Luciano *et al.* 2012) used chelating agents to leach heavy metals from soil columns, and also found the soil permeability decreased in this process. Clogging may be a major problem in the application of chelant-enhanced flushing when keeping constant water head during the flushing process.

As can be seen from Fig. 2, the concentration of Cu(II) in leachate for 400 mg/kg Cu(II) contaminated soil had the similar tendency and the peak value was 196.8, 300.1 and 328.7 mg/L when using citric acid of 0.005, 0.015 and 0.03mol/L, respectively. Based on the facts found from both Figs. 1 and 2, the maximum concentration of Cu(II) in the leachate became higher with the increase of concentration of citric acid, and the peak value seemed to occur at about the same time.

Fig. 3 shows the tendency of Cu(II) removal ratio for silt soil contaminated by 200 mg/kg Cu(II) with the increase of pore volumes when flushed with different concentrations of citric acid. The removal ratio of Cu(II) went up sharply flushed by citric acid with concentrations of 0.005 and 0.015mol/L with the increase of pore volumes initially. While with the continuing growth of pore volumes, the removal ratio became stable and finally reached 59.9 and 52.9% for the soil flushed by 0.005 and 0.015mol/L citric acid respectively.

As Fig. 4 shows, the removal ratio for soil contaminated by 40 0mg/kg Cu(II) also had the similar tendency compared with the 200 mg/kg Cu(II) contaminated soil. However, the final removal ratio was 73.42, 58.03 and 49.59% for the soil flushed by 0.005, 0.015 and 0.03mol/L citric acid respectively.

In general, with the development of the pore volumes, the removal ratio increased and finally became stable under different concentrations of citric acid. At the initial stage, lower concentration of citric acid had lower removal ratio of Cu(II). While as the flushing continued, the lower concentration of citric acid, the higher final removal ratio was. Acid leaching could strongly affect soil structure and cause important losses (up to 50%) of the soil mineral substances (Tampouris *et al.* 2001). With the increase of citric acid concentration, the structure of the soil became weaker and soil permeability also decreased which may lead to the decrease of the removal ratio.

Fig. 5 and 6 show the residual concentrations of Cu(II) in 200 and 400 mg/kg Cu(II) contaminated soils respectively flushed by different concentrations of citric acid. The concentration of Cu(II) in soils flushed by different concentrations of citric acid declined rapidly with the proliferation of the pore volumes. In addition, the residual concentration of Cu(II) in soil at the end decreased with decreasing the concentration of citric acid, in other words, the removal efficiency of Cu(II) was higher when using lower concentration of citric acid. The final

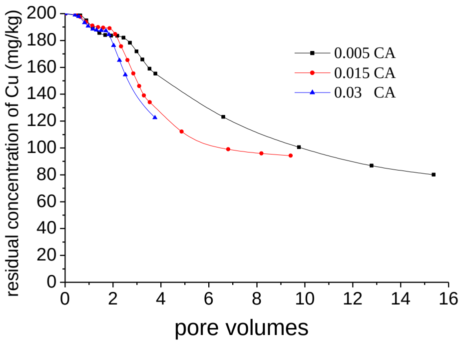

Fig. 5 Residual concentration of Cu(II) in 200 mg/kg Cu(II) contaminated soil after flushed by different concentrations of citric acid

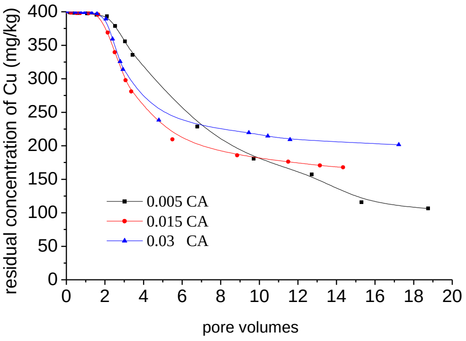

Fig. 6 The residual concentration of Cu(II) in 400 mg/kg Cu(II) contaminated soil after flushed by different concentrations of citric acid

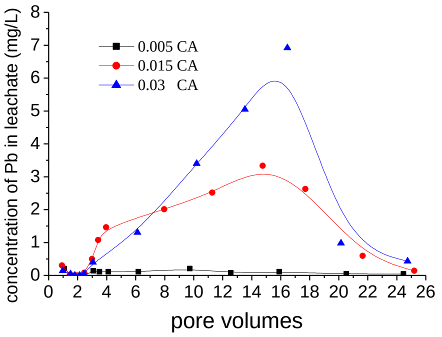

Fig. 7 Variation of Pb(II) concentration in leachate from 200 mg/kg Pb(II) contaminated silt soil flushed with different concentrations of citric acid

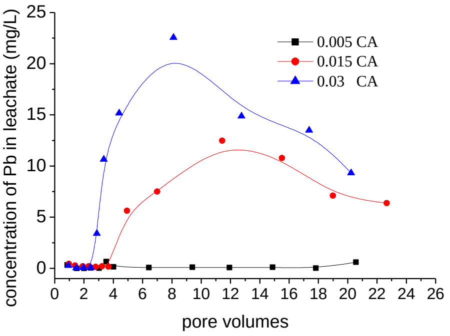

Fig. 8 Variation of Pb(II) concentration in leachate from 400 mg/kg Pb(II) contaminated silt soil flushed with different concentrations of citric acid

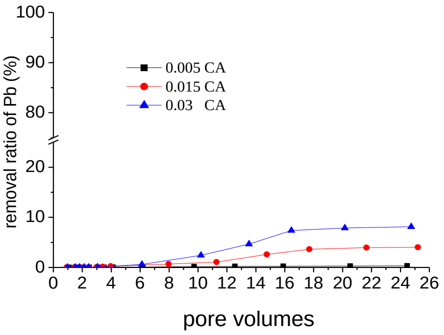

Fig. 9 Removal ratio of Pb(II) from 200 mg/kg Pb(II) contaminated silt soil flushed with different concentrations of citric acid

concentration of Cu(II) in contaminated soil with 200 and 400 mg/kg Cu(II) was 80.1 and 106.3mg/kg respectively after leached by 0.005 mol/L citric acid.

3.2 Pb(II) contaminated silt soil flushed by different concentrations of citric acid

Fig. 7 shows the variation of Pb(II) concentration in leachate from 200 mg/kg Pb(II) contaminated silt soil flushed with different concentrations of citric acid. In detail, as flushed by different concentrations of citric acid, the concentration of Pb(II) in leachate grew slowly with increasing pore volumes at initial stage, and there was a small peak concentration of 3.3 and 6.9mg/L when using citric acid of 0.015 and 0.03mol/L respectively. However, the development of Pb(II) concentration when using 0.005mol/L citric acid was not obvious and the concentration was very small.

It can be found from Fig. 8, the concentration of Pb(II) in leachate for 400mg/kg Pb(II) contaminated soil had the similar tendency and the peak value was 12.6 and 22.6 mg/L when using citric acid of 0.015 and 0.03mol/L, respectively. The concentration of Pb(II) in leachate using 0.005mol/L citric acid was still very small and had no obvious variation.

Compared with Cu(II) contaminated soil flushed by citric acid, the peak value of the concentration of Pb(II) occurred much later than that of Cu(II), and was much smaller than that of Cu(II).

Fig. 9 shows the tendency of Pb(II) removal ratio for silt soil contaminated by 200mg/kg Pb(II) with the increase of pore volumes as flushed by different concentrations of citric acid. The removal ratio went up extremely gradually with the development of pore volumes and became stable with the pore volume continuing increase which finally reached 0.3, 4.7 and 8.2% when using citric acid with concentrations of 0.005, 0.015 and 0.03mol/L respectively. The removal ratio of Pb(II) seemed very small flushed by citric acid, especially for the soil flushed by 0.005mol/L citric acid.

As Fig. 10 shows, the removal ratio for soil contaminated by 400mg/kg Pb(II) also had the similar tendency compared with the 200mg/kg Pb(II) contaminated soil. And the final removal ratio was 0.2, 3.8 and 16.7% for

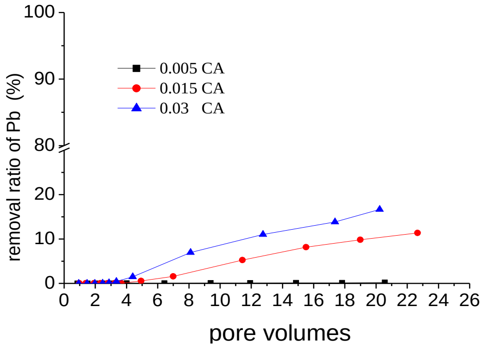

Fig. 10 Removal ratio of Pb(II) from 400 mg/kg Pb(II) contaminated silt soil flushed with different concentrations of citric acid

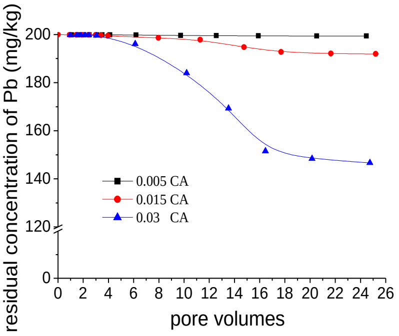

Fig. 11 Residual concentration of Pb(II) in 200 mg/kg Pb(II) contaminated soil after flushed by different concentrations of citric acid

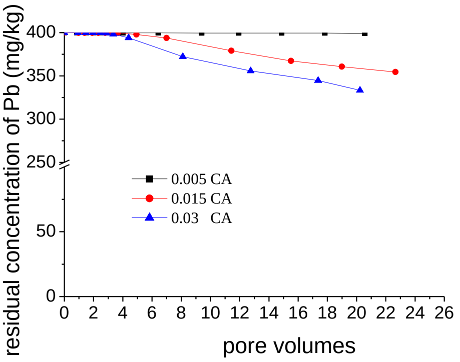

Fig. 12 Residual concentration of Pb(II) in 400 mg/kg Pb(II) contaminated soil after flushed by different concentrations of citric acid

the soil flushed by 0.005, 0.015 and 0.03mol/L citric acid respectively. Analogous to this study (Ash *et al.* 2016) also observed that removal amount of Pb(II) from soils contaminated by very high concentrations of Pb(II) increased with increasing citric acid concentration from 0.0005 to 0.025 mol/L in column leaching test.

In general, as well as the soil contaminated by Cu(II), with the increase of pore volumes, the removal ratio increased and finally became stable under different concentrations of citric acid. The higher concentration of citric acid, the higher final removal ratio of Pb(II) was, which was different from the results of soil contaminated by Cu(II). (Gao *et al.* 2003) found that citric acid had influence on the desorption behavior of Cd(II) and Cu(II) from soil using lab batch test, and when the concentration of citric acid was relatively low, the desorption amount of Cd(II) could decrease, while with increasing the concentration of citric acid, the desorption ratio began to steadily increase. In the presence of the same concentration of citric acid, Cu(II) desorption was enhanced directly and no inhibition was found. (Yang *et al.* 2006) found small concentration of citric acid inhibited Pb(II) desorption from soil in batch experiments. (Perez-Esteban *et al.* 2013) found 0.0005 mol/L citric acid inhibit desorption of Cu(II) and Zn(II), and 0.01 mol/L citric acid was very effective to help enhance Cu(II) desorption from soil but not effective for Zn(II) desorption by column leaching test.

The effect of citric acid on the removal of Pb(II) was just like Cd(II) and Zn(II). When small amount of citric acid entered into the soil pores, they would be mostly adsorbed by soil organic matter and clay minerals or be rapidly biodegraded (Evangelou *et al.* 2008, Liu *et al.* 2008). Then, with continuous enhancing the citric acid concentration, the ratio of organic ligands concentration in pores to the organic ligands adsorbed by soil became larger and desorbed Pb(II) from soil.

As can be seen from Figs. 11 and 12, the concentration of Pb(II) in soil which flushed by different concentrations of citric acid declined with the proliferation of the pore volumes. The concentration of Pb(II) in soil flushed by 0.005mol/L citric acid nearly had no change. The residual concentration of Pb(II) in soil at the end decreased with increasing the concentration of citric acid, namely, the removal efficiency of Pb(II) was higher when using higher concentration of citric acid. The final concentration of Pb(II) in contaminated soil with 200 and 400mg/kg Pb(II) was 146.6 and 333.4 mg/kg respectively after flushed by 0.03mol/L citric acid.

3.3 Different concentrations of heavy metal contaminated silt soils flushed by citric acid

Figs. 13 and 14 show the variation of leachate concentration of contaminated soils with different initial concentration of heavy meals using 0.015mol/L citric acid. To be specific, for three groups of Cu(II) contaminated soils, the concentration of Cu(II) in leachate tended to grow

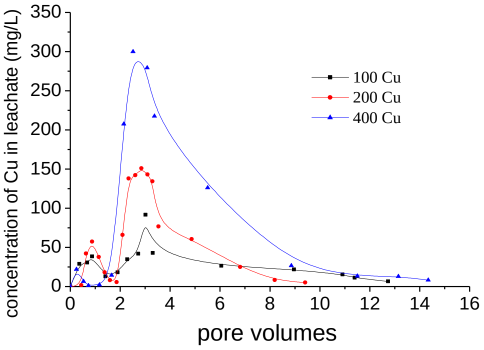

Fig. 13 Variation of Cu(II) concentration in leachate from different concentrations of Cu(II) contaminated silt soil flushed by citric acid

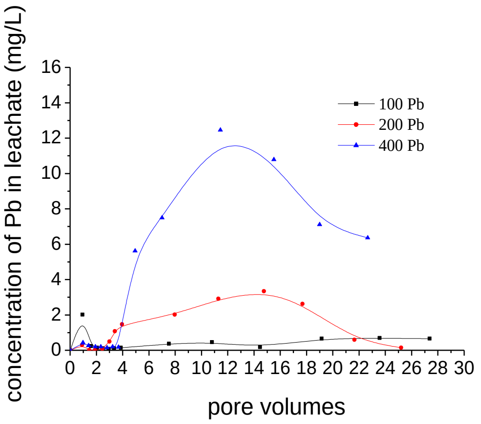

Fig. 14 Variation of Pb(II) concentration in leachate from different concentrations of Pb(II) contaminated silt soil flushed by citric acid

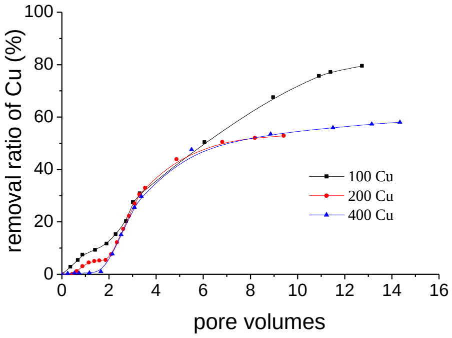

Fig. 15 Removal ratio of Cu(II) for different concentrations of Cu(II) contaminated silt soil flushed by citric acid

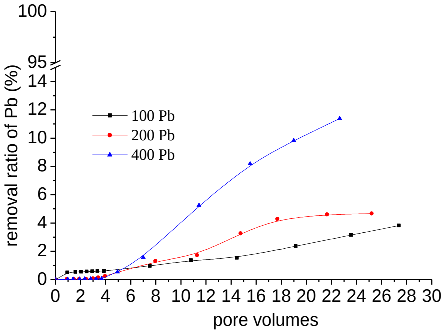

Fig. 16 Removal ratio of Pb(II) for different concentrations of Pb(II) contaminated silt soil flushed by citric acid

at initial stage, and the peak value was found to happen at nearly the same pore volume, 91.6, 151.0 and 300.1 mg/L for soils with Cu(II) concentrations of 100, 200 and 400 mg/kg respectively. When it came to Pb(II) contaminated soils, the concentration of Pb(II) in leachate grew slowly with increasing pore volumes, and there was a small peak concentration of 3.3 and 12.5 mg/L for soils with Pb(II) concentration of 200 and 400mg/kg respectively, while for the soil with 100mg/kg Pb(II) the leachate concentration was very small and there was no obvious change. In a whole, the peak value of the concentration of Pb(II) was found to occur much later than that of Cu(II), and was much smaller than that of Cu(II).

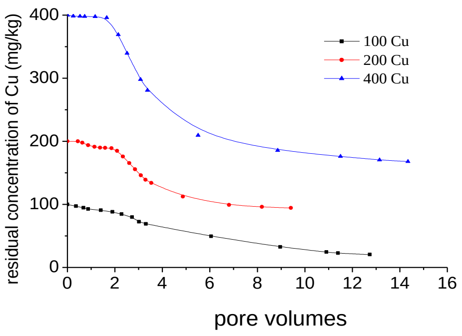

Fig. 17 Residual concentration of Cu(II) in different concentrations of Cu(II) contaminated silt soil flushed by citric acid

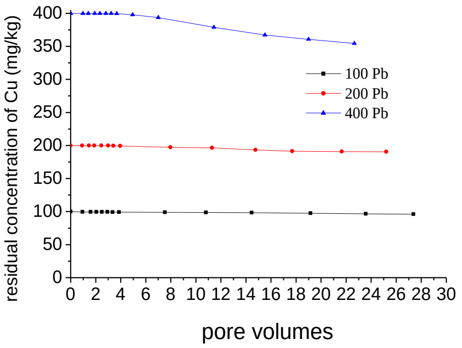

Fig. 18 Residual concentration of Pb(II) in different concentrations of Pb(II) contaminated silt soil flushed by citric acid

As can be seen from Fig. 15, flushed by 0.015mol/L citric acid, the removal ratio of Cu(II) went up mildly and finally became stable with the increase of pore volumes. The final removal ratios were 79.6, 52.9 and 58.0% for contaminated soil with initial Cu(II) concentration of 100, 200 and 400 mg/kg respectively. The removal ratio of Cu(II)was greater when the initial Cu(II)concentration was lower in the contaminated soil. When the concentration of Cu(II) was lower, the copper ions could have more chances to react with the citric acid, therefore, removal ratios could become higher.

As shown in Fig. 16, the removal ratio of Pb(II) got higher with higher initial Pb(II) concentration in the contaminated soil after leached by 0.015mol/L citric acid, and the final removal ratios were 3.8, 4.7 and 11.4% for contaminated soil with initial Pb(II) concentration of 100, 200 and 400 mg/kg respectively. In contrast, the removal ratio of Cu(II) was much higher than that of Pb(II) when using the same concentration of citric acid.

Figs. 17 and 18 show the residual concentration of heavy metals in different contaminated soils after flushed by 0.015mol/L citric acid. As can be seen, the residual concentration of Cu(II) and Pb(II) both declined, however, the residual concentration of Pb(II) was eventually much higher that of Cu(II), indicating the same dosage of citric acid has a better effect on Cu(II) contaminated soil. The final concentration of Cu(II) in contaminated soil with initial Cu(II) of 100, 200 and 400mg/kg was 20.5, 94.2 and 167.9 mg/kg respectively after flushed by 0.015mol/L citric acid. While for contaminated soil with initial Pb(II) concentration of 100, 200 and 400mg/kg, the final Pb(II) concentration was still very high, with 96.2, 190.7 and 354.4 mg/kg respectively.

In comparison, the removal ratios of different concentrations of Cu(II) leached by citric acid were all higher than those of Pb(II). Obviously, the effect of Cu(II) contaminated soil leached by citric acid was better than the Pb(II) contaminated soil, obeying the complexation formation constant of Cu(II) ($\lg K_{Cu} = 6.1$) greater than that of Pb(II) ($\lg K_{Pb} = 4.1$) (Wuana *et al.* 2010, Di Palma and Mecozzi 2007). Copper and Pb(II) could form tridentate, mononuclear complexes with citric acid involving two carboxyl acid groups and the hydroxyl group (Francis *et al.* 1992, Bassi and Prosher 2000). With high doses of citric acid, Cu(II) desorption was governed by the low pH and the formation of complexes. The higher amount of Cu(II) extracted by citric acid could be explained by the greater effectiveness in oxides dissolution and the stronger chelating ability of citric acid with Cu(II) (Perez-Esteban *et al.* 2013). As observed from the results of Cu(II) contaminated soil column flushed by 0.03mol/L citric acid, the test terminated in advance because of low permeability, while this phenomenon does not exist in the tests for Pb(II) contaminated soil column. The complex compounds formed by the desorption of Cu(II) from the soil may cause the clogging in the soil. However, the concentration of citric acid was not enough to obtain significant efficiency for Pb(II) removal. This is in some connection with the complexation formation constant as described above. And the molecular weight of Pb(II) is much larger than Cu(II), so the mole number of Pb(II) is much smaller than that of Cu(II) when using the same element weight, while the same unit of citric acid must consume the same mole number of divalent Cu(II) and Pb(II) to complex. In addition, the first peak of Cu(II)concentration as occurred in Fig. 1 might indicate that Cu(II) was not fully bounded by the soil sample, which was flushed out quickly within a unit pore volume of fluid wash. This behavior has not been found for Pb(II) in Fig. 7, suggesting that Pb(II) is more strongly bounded with the silty soil than Cu(II) could. Some Pb(II) might be adsorbed by soils tightly due to preferential adsorption by the sites of highest binding energy(Wasay *et al.* 1998), and could not be removed easily, thus longer contact time is needed to break the covalent bonding to ensure better removal efficiency. As increasing Pb(II) concentration, the limited amount of citric acid can have more chance to contact with Pb(II), so the removal ratio is a little higher.

4. Conclusions

Citric acid was found to have a profound effect on facilitating removal of heavy metals from contaminated silty soil using soil flushing method.

(1) The removal efficiency of Cu(II) was very remarkable, and the final removal ratio of Cu(II) could reach 73.4% for contaminated soil with initial Cu(II) concentration of 400mg/kg when using 0.005mol/L citric acid, whereas the removal efficiency decreased with increasing the concentration of citric acid because higher concentration of chelating agent could lead to decrease of permeability in soil.

(2) The removal efficiency of Pb(II) was not so good as Cu(II), with the maximum removal ratio only 16.7%, and higher citric acid concentration led to higher removal efficiency, because lower concentration of citric acid would be adsorbed on soil surface and caused inhibitory effect on Pb removing from contaminated soil.

(3) As for soil contaminated with different concentrations of heavy metals, the peak value of the concentration of Pb in leachate was found to occur much later than Cu(II), and was much smaller than Cu(II). The removal ratio of Cu(II) was greater when the initial Cu(II) concentration was lower in the contaminated soil, however, the same concentration of citric acid was not effective to remove Pb(II).

In conclusion, removal behaviors of Cu(II) and Pb(II) from contaminated silty soils were rather different, therefore, appropriate concentration of citric acid should be carefully chosen to flush the heavy metal contaminated site, and this study can provide some theoretical basis for remediating heavy metal contaminated site by soil flushing.

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