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# Distribution and speciation of heavy metals in two different sludge composite conditioning and deep dewatering processes

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Pilot-scale sewage sludge dewatering experiments were conducted using two composite conditioners: $FeCl_3$ + lime (Fe-lime) and Fenton's reagents + red mud (Fenton-RM). Mass balance analysis was performed on Cu, Zn, Pb, Cd, and Cr during the conditioning and dewatering processes to investigate their transformation and distribution. Speciation of heavy metals was also investigated by the Tessier sequential extraction method. Results show that (1) most of the heavy metals were retained in the solid cake during the dewatering process, especially Cu and Cr; (2) in the sludge cakes, more than 87 wt% of Cu and Cr existed in organic bound and residual forms, and the contents of the bioavailable fractions (exchangeable, carbonate bound and Fe–Mn oxides bound form) for Zn, Pb, and Cd were larger in the Fenton-RM system than those in the Fe-lime system; (3) the main factors affecting the distributions of these heavy metals are pH and chemical speciation in two conditioning processes. Generally, heavy metal concentrations in filtrate and dewatered sludge cakes in both systems were below the corresponding standards, and heavy metals in the dewatered sludge cake could be more effectively immobilized in the Fe-lime system than in that of the Fenton-RM system.

## 1. Introduction

Wastewater treatment plants produce a large amount of sewage sludge worldwide. Its treatment and disposal options include incineration, landfilling and agricultural applications as soil conditioners or fertilizers. However, its high moisture content often limits the applicability of these options. In addition, heavy metals retained in the sludge may have adverse impacts on humans and the environment.[1] In conventional sewage sludge dewatering processes, the water content of a dewatered sludge cake is typically about 75–85 wt% after chemical conditioning with polymers, for example polyacrylamide, followed by mechanical dewatering.

Various attempts have been made to improve sludge dewatering by applying pretreatments. The pretreatments can be physical (such as electro-dewatering),[2] chemical (including salt addition, pH modification, and advanced oxidation),[3–5] thermal,[6] or biological.[7]

Recently deep dewatering of sewage sludge after chemical pretreatment has been extensively studied. In a deep dewatering process, water content might be reduced to less than 60 wt%, which is beneficial for subsequent reuses or disposal. The combined ferric chloride and lime system (referred to as Fe-lime) is a common composite conditioner recommended by U.S. EPA[8] applied in pretreatment of sewage sludge before deep dewatering.[9,10] In this composite conditioner, ferric chloride (Fe) serves as an inorganic coagulant to provide polyvalent cation ($Fe^{3+}$), while lime functions as a skeleton builder to promote a rigid and permeable structure of sludge flocs that leads to an improvement in sludge compressibility. However, high pH of the produced filtrate and huge dosage of chlorides limits its applications.

An innovative composite conditioner (referred to as Fenton-RM) has been developed by our research group, in which Fenton's reagent (Fenton) provides an advanced oxidation process, while red mud (RM) works as a skeleton builder. The experimental results showed that the Fenton-RM system achieved better dewatering with less chemical doses and a neutral filtrate.[11] Furthermore, red mud is a kind of industrial residue, so the cost will be reduced by replacing lime with red mud, and the emission of $CO_2$ will also decrease. However, the distribution and speciation of heavy metals (*e.g.*, Cu, Zn, Pb, Cd, and Cr) during sewage sludge conditioning and dewatering procedures have seldom been investigated.

The total contents of heavy metals determined after acid digestion of sludge samples may serve as a gross index, while it provides little insight for their potential mobility.[12,13] Various

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extraction methods, simple or sequential, have been developed. Sequential extraction is considered as the preferred method for obtaining information on bioavailability, mobilization and transportation of heavy metals in sludge.[14,15] Therefore, sequential extraction method has commonly been used to evaluate the speciation of heavy metals in the field of sewage sludge. In previous studies, numerous studies have been investigated on speciation of heavy metals in the reuse and disposal of sewage sludge. Speciation of heavy metals was studied during sewage sludge solidification,[16] anaerobic digestion,[17] incineration[18] or co-composting for agricultural use.[19–21] Results showed that cement kiln dust could modify the chemical speciation of metals into less available forms. High-solid anaerobic digestion could increase the bioavailability of Cu, Zn, Ni and Cr while decrease the bioavailability of Pb. Lime is a suitable material to co-compost with sewage sludge to reduce the availability of heavy metals. And the evolution of heavy metal speciation in incineration was complicated and decided by the heavy metal itself. However, less study on distribution and speciation of heavy metals has been investigated during sewage sludge deep dewatering process.

The overall objective of this study (as depicted in Fig. 1) was to investigate the distribution and speciation of heavy metals in the filtrate and the dewatered cake using two different composite conditioners (Fe-lime and Fenton-RM). The mobility and bioavailability of heavy metals in the cakes produced from these two conditioning systems were also compared to evaluate the immobilization effect. This study on distribution and speciation of heavy metals could provide supporting information for selecting dewatering process and the following disposal approach for the dewatered sludge cake.

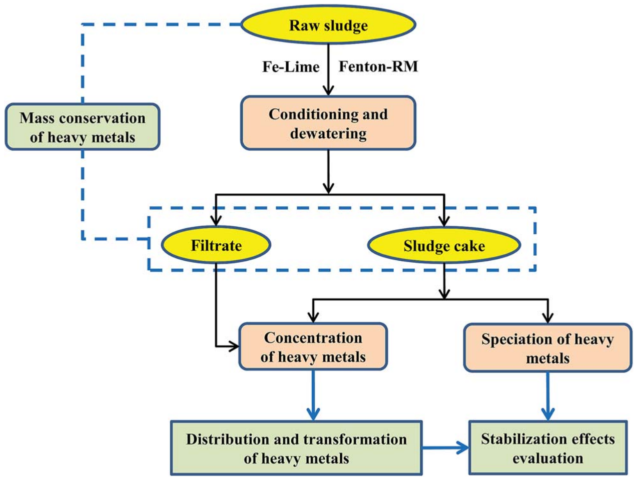

Fig. 1 Schematic of the study.

## 2. Materials and methods

### 2.1. Materials

The raw sludge (RS) used in this study were the excessive waste activated sludge from Tangxunhu Wastewater Treatment Plant (WWTP) in Wuhan City of China and was collected in Oct. 2013. Oxidation ditches are applied for biological treatment of the waste water in this plant. RS samples of the thickened sludge were obtained from the gravity thickener in the pilot-scale deep dewatering facilities in order to make sure that RS samples were the same batch as used in the following conditioning and deep dewatering process. RS Samples were transported to the laboratory in polypropylene containers and stored at 4 °C before use. Some basic characteristics and heavy metal concentrations of RS samples (three parallel samples) are tabulated in Table 1. The moisture content of sludge was measured by oven drying the sludge at 105 °C for 24 h, and the pH was measured by a pH meter. It can be seen from Table 1 that the RS samples had a pH close to neutral, and the heavy metals concentration are in the order of Zn > Cu > Cr > Pb > Cd.

$FeCl_3$ (industrial grade), $FeSO_4\cdot7H_2O$ (industrial grade) and $H_2O_2$ (27.5 wt%, industrial grade) were obtained from Sinopharm Chemical Reagent Company, China. $H_2SO_4$ (analytical grade, Xinyang Chemical Company, China) was used to adjust the initial pH of the RS samples to the optimal value of 5.0 before the addition of Fenton's reagent.[22] All the acids used for sludge sample digestion (including $HNO_3$, HF and $HClO_4$ from Sinopharm Chemical Reagent Company, China) were of analytical grade.

Lime and red mud were used as skeleton builders. They were dried, milled and sieved to less than 1 mm in particle size before used. The red mud was supplied by an alumina plant using the Bayer process for the production of alumina from the bauxite leaching with sodium caustic solution in Zhengzhou City of China, while the lime was obtained from a local factory. Their chemical compositions were presented in Table 2. In Table 2, LOI (Loss of ignition)[23] is the mass loss of organic matters and some thermal decomposition of inorganic matters in 1200 °C for 24 h.

### 2.2. Methods

**2.2.1. The dewatering methods.** A pilot-scale sludge dewatering facility was built in the Tangxunhu WWTP. A sketch of the sludge conditioning and dewatering process is presented in Fig. 2. First, raw sludge with a moisture content of about 99 wt% was concentrated to approximately 96 wt% by gravity thickening. Then 500–800 kg of the thickened sludge was pumped into a tank and conditioned according to the procedure presented in Table 3. In this study, the dosages of the reagents and

Table 1 Some basic characteristics and heavy metal concentrations of the raw sludge samples

| pH | Moisture (wt%) | VSS/TSS (wt%) | Cu (mg kg$^{-1}$) | Zn (mg kg$^{-1}$) | Pb (mg kg$^{-1}$) | Cd (mg kg$^{-1}$) | Cr (mg kg$^{-1}$) |
|---|---|---|---|---|---|---|---|
| 7.2 | 96.0 ± 0.1 | 41.3 ± 0.3 | 132.1 ± 2.9 | 279.6 ± 12.9 | 39.8 ± 0.5 | 4.5 ± 0.2 | 73.3 ± 0.9 |

Table 2 Chemical compositions of the skeleton builders (wt%)

| Skeleton builders | $SiO_2$ | CaO | $Al_2O_3$ | MgO | $Na_2O + K_2O$ | $Fe_2O_3$ | $TiO_2$ | $SO_3$ | $Cl^-$ | LOI[a] |
|---|---|---|---|---|---|---|---|---|---|---|
| Lime | 7.2 | 62.0 | — | 1.7 | — | — | — | — | — | 24.1 |
| Red mud | 20.4 | 12.9 | 24.5 | 1.0 | 12.3 | 9.5 | — | 0.7 | 0.1 | 4.0 |

[a] LOI = loss of ignition at 1200 °C.

optimal conditions were used according to our previous studies.[11,24]

After the conditioning process, the sludge was pumped into a diaphragm filter press by a screw pump for dewatering, comprising a 60 min feeding pressing phase to a pressure of 0.8 MPa through four steps and a 30 min diaphragm pressing phase with a pressure of 1.5 MPa. The whole process was controlled by a programmable logic controller control system.

**2.2.2. Determination of heavy metals.** To get insights on the migration and distribution of heavy metals during conditioning and dewatering, mass balances on heavy metals (Cu, Zn, Pb, Cd and Cr) before conditioning and at the end of conditioning and dewatering were conducted. Mass balances during conditioning and dewatering were evaluated by using eqn (1) below:

$$m_{rs} + m_c = m_{cs} + m_g = m_{sc} + m_f + m_r \tag{1}$$

where $m_{rs}$ = mass of RS samples (kg); $m_c$ = mass of the conditioner (kg); $m_{cs}$ = mass of the conditioned sludge (kg); $m_g$ = mass of gases released to the air (negligible); $m_{sc}$ = mass of the dewatered sludge cake (kg); $m_f$ = mass of the filtrate (kg); and $m_r$ = mass of the residual sludge (kg), which refers to the conditioned sludge that could not enter into the filter press chamber. Mass balances during conditioning and dewatering process provide the basis of mass balances of heavy metals during conditioning and dewatering process.

500 kg and 800 kg of RS samples were treated by the Fe-lime system and Fenton-RM system, respectively. For analysis, three parallel samples were firstly oven dried at 105 °C, and then all the dried sludge samples were first digested by three acids ($HNO_3$–HF–$HClO_4$) following the procedure in Modern Analysis Method of Soil Elements.[25] Thus, some of the retained heavy metals in the liquid portion would be kept in the solid sludge cake. Heavy metals in the digested solutions were then analyzed by an atomic absorption spectrometer (Analytik Jena AG NovAA 400, Germany), and the certified reference material came from National Testing Center of Nonferrous Metals and Electronic Materials Analysis, China. All the results were the mean values of triplicate samples.

Mass balances on heavy metals were calculated using eqn (2) below:

$$m_{cs} \times C_{cs} = m_{sc} \times C_{sc} + m_f \times C_f + m_r \times C_r \tag{2}$$

where $C_{cs}$ = heavy metal concentration in the conditioned sludge (mg kg$^{-1}$ for solid); $C_{sc}$ = heavy metal concentration in the dewatered sludge cake (mg kg$^{-1}$ for solid); $C_f$ = heavy metal concentration in the filtrate (mg L$^{-1}$), assuming that the specific gravity of the filtrate is equal to 1; $C_r$ = heavy metal

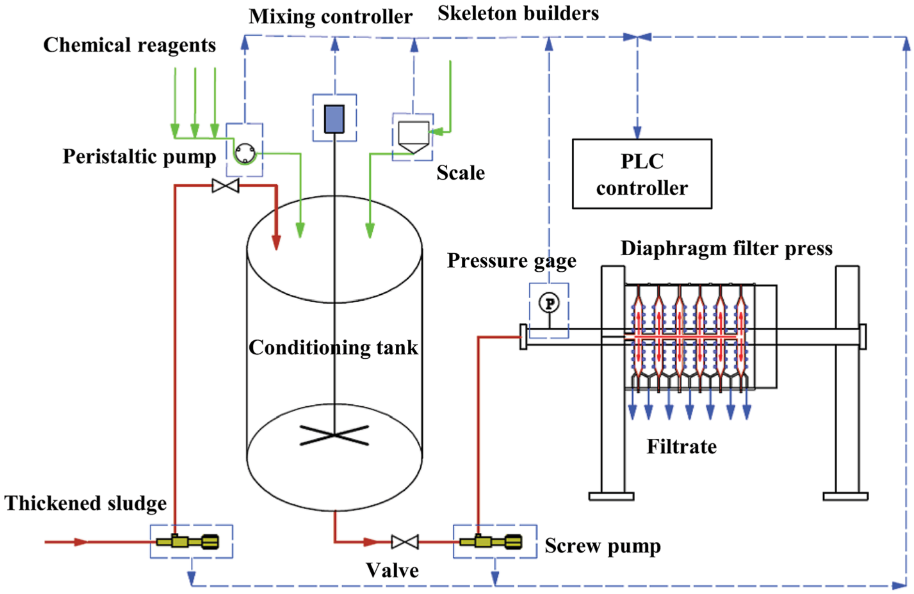

Fig. 2 Sketch of the experimental setup for sludge conditioning and dewatering process.

Table 3 Main parameters of two sludge conditioning systems

| | Reagent dosage | | |
|---|---|---|---|
| Conditioning system | Chemical agents | Skeleton builders | Conditioning method |
| Fe-lime | $FeCl_3$ (50 mg $g^{-1}$ DS[a]) | Lime (500 mg $g^{-1}$ DS) | Lime (rapid mixing/15 min) → $FeCl_3$ (slow mixing/5 min) |
| Fenton-RM | $H_2SO_4$ (adjust pH to 5)<br>$Fe^{2+}$ (32 mg $g^{-1}$ DS)<br>$H_2O_2$ (34 mg $g^{-1}$ DS) | Red mud (275 mg $g^{-1}$ DS) | $H_2SO_4$ (rapid mixing/3 min) → $Fe^{2+}$ solutions (rapid mixing/3 min) → $H_2O_2$ (slow mixing/30 min) → red mud (slow mixing/10 min) |

[a] DS = dry solid.

Table 4 Detailed information of the modified five-step sequential extraction method

| Step | Target phase | Extraction agents and conditions |
|---|---|---|
| F1 | Exchangeable | 16 mL 1 M $MgCl_2$ (pH = 7.0), 2 h |
| F2 | Bound to carbonates | 16 mL 1 M NaOAc adjusted to pH 5 with HOAc, 5 h |
| F3 | Bound to Fe–Mn oxides | 40 mL 0.04 M $HONH_3Cl$ (pH = 2.0), 4 h |
| F4 | Bound to organic matter | 6 mL 0.02 M $HNO_3$ and 10 mL $H_2O_2$ (30%), 2 h at 85 °C; 1 h at ambient temperature; 6 mL $H_2O_2$ (30%), water bath for 1.5 h at 85 °C; 10 mL 3.2 M $NH_4OAc$ and 8 mL distilled water, 0.5 h at ambient temperature |
| F5 | Residual | 15 mL $HNO_3$, 10 mL HF, 5 mL $HClO_4$, until totally digested |

concentration in the residual sludge (mg $kg^{-1}$ for solid) and $C_r$ is assumed to be the same as $C_{cs}$. Mass balances on heavy metals are important to study the distribution of heavy metals between the dewatered cake and the filtrate.

**2.2.3. Speciation of heavy metals.** Speciation of the heavy metals were determined by using a five-step method as described in Tessier *et al.*[26] (see in Table 4). The five fractions are exchangeable (noted as F1), bound to carbonates (noted as F2), bound to Fe–Mn oxides (noted as F3), bound to organic matters (noted as F4), and residual (noted as F5).

At the end of each extraction, separation was achieved by centrifugation at 3000 rpm for 30 min, and the supernatant was then filtered through a 0.45 μm membrane. The solid residual was then used in the subsequent extraction step.

## 3. Results and discussion

### 3.1. Mass balance of heavy metals in conditioning and dewatering processes

Results of mass balance on solids are tabulated in Table 5. The mass loss in the dewatering process can be calculated from eqn (3) below:

$$\text{Mass loss} = m_{cs} - m_{sc} - m_f - m_r \tag{3}$$

Consequently, the mass loss in percentage in dewatering can be calculated by using eqn (4):

$$\text{Mass loss in percentage} = (m_{cs} - m_{sc} - m_f - m_r)/m_{cs} \times 100\% \tag{4}$$

As shown in Table 5, both the mass loss in percentage from both Fe-lime and Fenton-RM systems were less than 2 wt%.

Heavy metal concentrations of conditioned sludge, dewatered sludge cake, and filtrate samples are shown in Table 6. Although the filtrate would be returned to the front end of the plant for further treatment, all the heavy metal concentrations in the filtrate were below the China's Grade A Standard of the Water Quality Standard of Sewage Discharged into Urban Sewer (CJ 343-2010) (Cu < 2 mg $L^{-1}$, Zn < 5 mg $L^{-1}$, Pb < 1 mg $L^{-1}$, Cd < 0.1 mg $L^{-1}$, Cr < 1.5 mg $L^{-1}$). Moreover, the heavy metal concentrations in the filtrate can also meet the requirements in Integrated Wastewater Discharge Standard (GB 8978-1996). The pH values of the filtrate in the Fe-lime and the Fenton-RM

Table 5 Mass balance in the conditioning and dewatering processes[a]

| Conditioning system | $m_{rs}$ (kg) | $m_c$ (kg) | $m_{cs}$ (kg) | $m_r$ (kg) | $m_f$ (kg) | $m_{sc}$ (kg) | Moisture content (wt%) | Mass loss (kg) | Mass loss in percentage (wt%) |
|---|---|---|---|---|---|---|---|---|---|
| Fe-lime | 500 | 16.0 | 516.0 | 38.7 | 417.2 | 70.2 | 52.8 | −10.1 | −1.96 |
| Fenton-RM | 800 | 27.6 | 827.6 | 29.0 | 727.0 | 70.5 | 47.7 | 1.1 | 0.13 |

[a] $m_{rs}$ = Mass of RS samples (kg); $m_c$ = mass of the conditioner (kg); $m_{cs}$ = mass of the conditioned sludge (kg); $m_r$ = mass of the residual sludge; $m_f$ = mass of the filtrate (kg); $m_{sc}$ = mass of the dewatered sludge cake (kg); mass loss = $m_{cs} - m_{sc} - m_f - m_r$, and mass loss in percentage = $(m_{cs} - m_{sc} - m_f - m_r)/m_{cs} \times 100\%$.

Table 6 Heavy metal concentrations of the conditioned sludge, sludge cake and filtrate samples

| Conditioning system | Sample | Cu | Zn | Pb | Cd | Cr |
|---|---|---|---|---|---|---|
| Fe-lime | Conditioned sludge (mg kg$^{-1}$) | 90.9 ± 0.9 | 247.5 ± 6.6 | 31.4 ± 0.7 | 2.6 ± 0.2 | 57.7 ± 3.9 |
| | Sludge cake (mg kg$^{-1}$) | 91.7 ± 0.9 | 251.3 ± 7.0 | 30.3 ± 1.3 | 2.4 ± 0.4 | 59.9 ± 3.4 |
| | Filtrate (mg L$^{-1}$) | 0.02 | 0.01 | 0.42 | 0.03 | 0.06 |
| Fenton-RM | Conditioned sludge (mg kg$^{-1}$) | 115.5 ± 2.0 | 230.9 ± 6.5 | 36.3 ± 1.1 | 4.7 ± 0.1 | 117.3 ± 5.0 |
| | Sludge cake (mg kg$^{-1}$) | 101.6 ± 1.6 | 253.6 ± 5.6 | 36.0 ± 0.0 | 4.8 ± 0.1 | 117.4 ± 0.9 |
| | Filtrate (mg L$^{-1}$) | 0.01 | 1.99 | 0.57 | 0.03 | 0.16 |

Table 7 Mass balance of heavy metals between the conditioning and dewatering processes

| Conditioning system | Item | Cu | Zn | Pb | Cd | Cr |
|---|---|---|---|---|---|---|
| Fe-lime | Mass difference (mg) | −182.1 | −532.4 | −187.3 | −10.5 | −193 |
| | Mass difference in percentage (%) | −5.86 | −6.29 | −17.42 | −11.94 | −9.79 |
| Fenton-RM | Mass difference (mg) | 398.4 | −2500.3 | −437.3 | −28.7 | −230.5 |
| | Mass difference in percentage (%) | 9.26 | −6.97 | −11.87 | −16.37 | −5.28 |

systems were measured to be 12.3 and 6.7, respectively. With a pH of 6.7, the filtrate from the Fenton-RM system can be recycled to the plant or discharged without further treatment, while the filtrate from the Fe-lime system might need to go through neutralization because of the high pH of 12.3.

Results of mass balance calculations on heavy metals are shown in Table 7. Mass difference for each heavy metal before and after dewatering was calculated by eqn (5) below:

$$\text{Mass difference} = m_{\text{cs}} \times C_{\text{cs}} - m_{\text{sc}} \times C_{\text{sc}} - m_{\text{f}} \times C_{\text{f}} - m_{\text{r}} \times C_{\text{r}} \tag{5}$$

Consequently, mass difference in percentage for each heavy metal was calculated by eqn (6):

$$\text{Mass difference in percentage} = (m_{\text{cs}} \times C_{\text{cs}} - m_{\text{sc}} \times C_{\text{sc}} - m_{\text{f}} \times C_{\text{f}} - m_{\text{r}} \times C_{\text{r}})/(m_{\text{cs}} \times C_{\text{cs}}) \times 100\% \tag{6}$$

As shown in Table 7, mass difference in percentage for all the heavy metals were less than 18 wt%, which indicated that the mass balance in the conditioning and dewatering processes (Table 5) has influence on mass balance of heavy metals, especially in the case of Pb and Cd with the lower concentrations.

## 3.2. Distribution of heavy metals in the solid cake and filtrate

The distributions of the heavy metals in the sludge cake and the filtrate are shown in Fig. 3. As shown in Fig. 3, for the Fe-lime system, most of heavy metals were retained in the sludge cake after dewatering; >99 wt% for Cu and Zn, >95 wt% for Pb and Cr, while only 88.09 wt% for Cd. The percentages of heavy metal retention by the sludge cake were in the order of (Cu, Zn) > (Pb, Cr) > Cd for the Fe-lime system. In other words, the fractions of heavy metals in the filtrate were 0.23 wt% (Cu), 0.03 wt% (Zn), 4.91 wt% (Pb), 1.33 wt% (Cr) and 11.91 wt% (Cd).

It should be noted that the water content of the dewatered sludge cake was about 60 wt%. Consequently, some of the retained heavy metals would be present in the liquid portion within the sludge cake. Assuming that the heavy metal concentrations in the liquid portion were the same or similar to these of the filtrate, the mass in that fraction should be small and was ignored here since the concentrations in the filtrate were low.

For the Fenton-RM system, most of heavy metals were also retained in the sludge cake after dewatering. The percentages of heavy metal retention by the sludge cake were in the order of Cu > Cr > Cd > (Zn, Pb) for the Fenton-RM system. In other words, the fractions of heavy metals in the filtrate were 0.24 wt% (Cu), 2.59 wt% (Cr), 10.65 wt% (Cd), 13.42 wt% (Zn), and 13.72 wt% (Pb).

Similar to the case of the Fe-lime system, Cu fraction in the filtrate of the Fenton-RM system was also negligibly low. One plausible reason is that the oxidizable fraction was the dominant Cu species, so Cu was associated with organic matters to form stable chelated substances in the sludge.[27,28] This speciation of Cu in dewatered cake will be discussed in the next section.

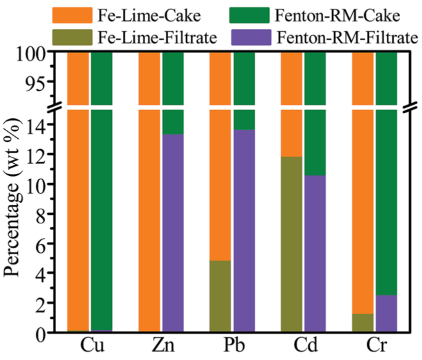

Fig. 3 Distributions of heavy metals between the dewatered cake and the filtrate in the Fe-lime and Fenton-RM systems.

On the other hand, the Zn fraction in the filtrate of the Fenton-RM system was much higher than that of the Fe-lime system (13.4 wt% *vs.* 0.1 wt%). The difference is possibly due to the difference in pH, which is a significant parameter on the mobility and fate of heavy metals in soils.[29–31] The difference of the mobility of Zn between the Fenton-RM system and Fe-lime system was attributed to higher pH (12.3) in the Fe-lime system compared with neutral pH in the Fenton-RM system. Furthermore, CaO in lime could have some stabilization effect on Zn. The fractions of Pb and Cr in the filtrate of the Fenton-RM system were also larger than those of the Fe-lime system.

The fraction of Cd in the filtrate of the Fenton-RM system was essentially the same as that in the Fe-lime system (10.7 wt% *vs.* 11.9 wt%). The possible reason is that the concentration of Cd in raw sludge is extremely low (4.5 mg kg$^{-1}$), as shown in Table 1. The variation was not obvious.

In general, the retentions of the metals by the sludge cakes were in the order of (Cu, Zn) > (Pb, Cr) > Cd for the Fe-lime system and Cu > Cr > Cd > (Zn, Pb) for the Fenton-RM system. Differences in distributions of the heavy metals in these two composite conditioning systems might come from differences in pH and fractions of heavy metals. Further studies are needed to explore the actual causes for the differences.

### 3.3. Speciation of heavy metals in the sludge cake

The biotoxicity of a heavy metal depends not only on its concentration, but also on its bioavailability. Generally, heavy metals in F1, F2, and F3 are considered mobile and bioavailable; while those in F4 and F5 are considered relatively stable and non-bioavailable.[32]

Speciation of heavy metals in the raw sludge and the sludge cakes from the Fe-lime and the Fenton-RM systems are shown in Fig. 4. As shown in Fig. 4, the fractions of Cu and Cr have a similar trend among the three samples; a high proportion (>87 wt%) in F4 and F5. This result appears to be in accordance with the distribution of the Cu and Cr in the sludge cake and the filtrate (Fig. 3). In both the Fe-lime and the Fenton-RM systems, Cu and Cr were significantly retained in the sludge cakes.

In the case of Zn, Pb and Cd, the fractions in the mobile state increased in the following order: raw sludge > Fenton-RM > Fe-

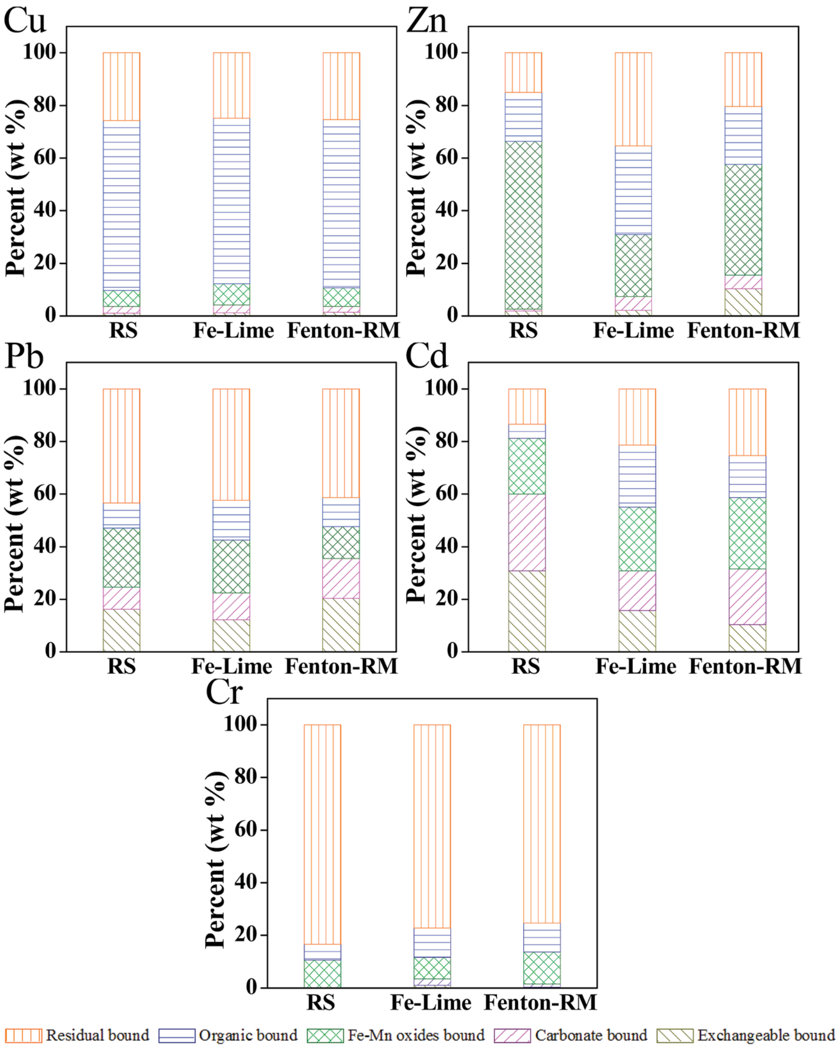

Fig. 4 Speciation of heavy metals in raw sludge and the dewatered sludge cakes from two different composite conditioner systems.

lime. Due to the high pH of the Fe-lime system, significant proportions of Zn and Pb existed in F4 and F5, when compared to those of the sludge cake of the Fenton-RM system and the raw sludge. Correspondingly, the fractions of Zn and Pb in the filtrate of the Fe-lime system are larger than those of the Fenton-RM system.

The results indicate that heavy metals can be more effectively stabilized by the dewatered cake of the Fe-lime system than that of the Fenton-RM system. However, the Fe-lime system was restricted by the higher pH of its filtrate, and the filtrate in the Fe-lime system should be further treated with acid. Some chlorides unavoidably retain in dewatered sludge cake in the Fe-lime system. If incineration process or other thermal conversion technologies were used to these sludge from the Fe-lime system, chlorides in the sludge cake easily cause the corrosion of equipment, and lead to the risk of emission of dioxins in combustion process. In contrast, it is not a question for the Fenton-RM system because no chlorides were added. Thus, landfill disposal is the only approach for the dewatered sludge cake in the Fe-lime system. For the Fenton-RM system, both combustion process and landfill disposal could be approaches for treatment of the dewatered sludge cake. Since the stabilization of heavy metals in sludge is a primary factor to evaluate a process, we have done a series of solidification experiments to the sludge cakes to solve the problem of high mobility of heavy metals in Fenton-RM system. The result will be discussed in the next paper.

## 4. Conclusions

The distribution and speciation of five heavy metals in two different composite conditioning and dewatering processes were investigated. By performing a mass balance analysis on heavy metals during the dewatering process, for the Fe-lime system, the retentions of heavy metals in the sludge cake were in the order of (Cu, Zn) > (Pb, Cr) > Cd; while for the Fenton-RM system, they were Cu > Cr > Cd > (Pb, Zn). Differences in distributions of heavy metals in these composites conditioning systems might come from differences in pH and speciation of heavy metals.

The experimental results show that more than 87 wt% of Cu and Cr existed in the F4 and F5 in both the Fe-lime and the Fenton-RM systems. For Zn, Pb and Cd, their fractions in the bioavailable state (F1–F3) were larger than those of Cu and Cr; and they were also higher in the Fenton-RM system than those in the Fe-lime system.

Generally, heavy metals in dewatered sludge cake could be more effectively immobilized in the Fe-lime system than in that of the Fenton-RM system. Landfill disposal is a more appropriate approach for the dewatered sludge cake in the Fe-lime system, while both combustion process and landfill disposal could be approaches for treatment of the dewatered sludge cake in the Fenton-RM system.

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