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RESEARCH ARTICLE

# Removal of heavy metal species from industrial sludge with the aid of biodegradable iminodisuccinic acid as the chelating ligand

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**Abstract** High level of heavy metals in industrial sludge was the obstacle of sludge disposal and resource recycling. In this study, iminodisuccinic acid (IDS), a biodegradable chelating ligand, was used to remove heavy metals from industrial sludge generated from battery industry. The extraction of cadmium, copper, nickel, and zinc from battery sludge with aqueous solution of IDS was studied under various conditions. It was found that removal efficiency greatly depends on pH, chelating agent's concentration, as well as species distribution of metals. The results showed that mildly acidic and neutral systems were not beneficial to remove cadmium. About 68 % of cadmium in the sample was extracted at the molar ratio of IDS to heavy metals 7:1 without pH adjustment (pH 11.5). Copper of 91.3 % and nickel of 90.7 % could be removed by IDS (molar ratio, IDS: metals=1:1) with 1.2 % phosphoric acid effectively. Removal efficiency of zinc was very low throughout the experiment. Based on the experimental results, IDS could be a potentially useful chelant for heavy metal removal from battery industry sludge.

**Keywords** Iminodisuccinic acid · Industrial sludge · Biodegradable chelating ligand · Heavy metal · Extraction

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## Introduction

The currently employed techniques for wastewater treatment result in the generation of large quantities of sludge that must be disposed off (Naoum et al. 2001). One of the most potential routes for the disposal (especially sewage sludge) is land application by virtue of the recycling usage of the valuable components (nitrogen, phosphorus, and organic matters) within the sludge (Zhu et al. 2009). However, industrial sludge contains a lot of harmful substances, such as battery industry, metallurgical industry generates great hazardous heavy metal sludge. Heavy metals are non-biodegradable and could remain almost indefinitely in the soil environment (Wuana et al. 2010). In most of the developing countries, many groups dispose sludge on nearby unused lands. Such practices lead to heavy metal contamination of the biosphere (Chaudri et al. 2000; Pueyo et al. 2001). Therefore, removal of heavy metal and even recovery of heavy metal from industrial sludge is an important problem to be solved.

Various methods for heavy metal removal from sewage sludge have been investigated in order to minimize the prospective health risks of sludge during its disposal or land application, including chemical extraction (Lo and Chen 1990), thermal treatment (Zorpas et al. 2001), bioleaching treatment (Chen et al. 2005), cementation, and ion exchange (Dabrowski et al. 2004). Thereinto, chemical extraction of heavy metals has received extensive attention due to its simple operation and short extraction time. Various inorganic acids ($HNO_3$, HCl, and $H_2SO_4$) (Naoum et al. 2001), organic acids (oxalic and citric acid) (Veeken and Hamelers 1999), and strongly complexing agents (EDTA and NTA) (Sun et al. 2001) have been proposed as effective extracting agents. The removal of heavy metals from sludge using sulfuric acid was reported in 1975 (Scott and Horlings 1975). This process was a hot-acid treatment (Yoshizaki and Tomida 2000) and not used in practice. Heavy metal removal by hydrochloric acid was also carried out (Oliver and Carey 1976), and the extraction rates of heavy metals were around 50 %. In the case of using mixture solution of hydrochloric acid and hydrogen peroxide, over 80 % of heavy metals was removed from the sludge (Yoshizaki and Tomida 2000). However, the use of strong acid to treat heavy metals needs high equipment requirements and neutralization of acid from leachate needs a large amount of lime. Organic acids could be an attractive extracting agent due to them being biologically degradable and performing at mildly acidic conditions (Veeken and Hamelers 1999). But the extraction is time consuming and low efficiency. Moreover, removing heavy metals with chelating agents is a particularly promising technique. Ethylenediaminetetraacetic acid (EDTA) and diethylenetriaminepentaacetic acid (DTPA) are common chelating agents in the removal of heavy metals. EDTA is very effective in mobilizing metals, but unfortunately, due to its low biodegradability (Bucheli-Witschel and Egli 2001), it is also very persistent in the environment. This can cause a rather high risk of metal leaching to the groundwater (Nowack 2002). During the past few years, the non-biodegradability of these ligands and their accumulation in the environment has been a great deal of concern. Thus, replacement of these ligands by more environmental friendly chelating agents would be highly desirable. From the late 1990s, many investigations have examined the suitability of EDDS as a substitute for EDTA. Ethylenediaminedisuccinate (EDDS), a biodegradable chelant, is a structural isomer of EDTA (Kołodyńska 2011; Schowanek et al. 1997). EDDS has been shown to be easily biodegradable, to form strong complexes with transition metals and radionuclides (Meers et al. 2008), and to cause a much smaller leaching of metal down the soil profile than EDTA (Grčman et al. 2003). However, iminodisuccinic acid (IDS), a new biodegradable chelant, has the same aspartic acid derivative as EDDS but better biodegradability than EDDS. After 7 days, 80 % of IDS underwent biodegradation was found (Kołodyńska 2013). IDS is also characterized by stability over a wide pH range, excellent complexation of heavy metal ions, and low environmental impact due to low toxicity (Cokesa et al. 2004a, b), so it may be another environmentally friendly alternative after EDDS for persistent EDTA. Previous works about IDS has focused in its synthesis, properties, as well as application for detergent, etc. In recent years, studies on the biodegradability of IDS, extraction heavy metals from waste water, application of anion exchangers in sorption of heavy metal complexes, and washing heavy metal-contaminated soil with IDS have been carried out in succession. However, using IDS to remove a large quantity of heavy metals in industry sludge is seldom reported.

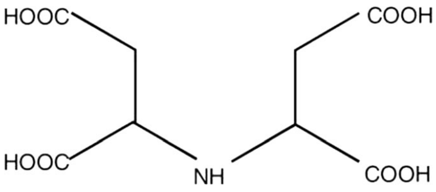
The aim of the present paper was to examine the possibility of using IDS as a kind of environmentally friendly chelating reagent to extract heavy metals from industrial sludge. The extraction performance was evaluated by the removal of cadmium, copper, nickel, and zinc from battery sludge, the effect of pH, contact time, molar ratio of IDS:metal, and addition of phosphoric acid on the extraction efficiency of cadmium, copper, nickel, and zinc from battery sludge in the presence of IDS was studied. Additionally, the effect of species distribution of heavy metals in sludge on extraction by IDS was also investigated. Extraction experiments were carried out under controlled conditions requiring treatment according to environmental regulations of China.

## Materials and methods

### Industrial sludge and chelating agents

The sludge sample used in this study was collected from a battery Co., Ltd. wastewater treatment plant in Xinxiang (Henan province, China). The sludge was air-dried in thin layers and then ground and sieved to the size less than 150 μm. The final material was stored in desiccators until analysis.

Iminodisuccinic acid was obtained from Bayer, Germany. Iminodisuccinic acid, (N-1, 2-dicarboxyethyl)-D,L-aspartic acid, is readily biodegradable and has a relative molecular mass of 249.17, solid content >34 %, density=1.676 g $cm^{-3}$, and the proportion of each element is as follows: C 38.56 %, H 4.45 %, N 5.62 %, and O 51.37 %. The structure of IDS can be described as follows:



All the other chemicals used in the study were analytical-grade reagents.

### Sequential extraction experiments

In order to know speciation of heavy metals in sludge, the samples were analyzed using modified BCR (the Community Bureau of Reference, now the European Union “Measurement and Testing Programme”) sequential extraction method. This method has been widely accepted and applied to metal fractionation in different environmental samples (Arain et al. 2008). Compared with other methods, BCR method procedures possessed better reproducibility, precision, and comparable performance (Quevauviller et al. 1997). Table 1 summarized the steps and operationally defined metal fractions. Sequential extractions were conducted using 0.5 g of sludge

Table 1 Chemical reagents and analytical conditions for the modified BCR sequential extraction

| | Fraction | Agents and procedures | Extraction time (h) |
|---|---|---|---|
| Step 1 | Water soluble | 20 mL deionized water | 16 |
| Step 2 | Acid soluble | 20 mL HOAc (0.11 M, pH 2.8) | 16 |
| Step 3 | Reducible | 20 mL $NH_2OH \cdot HCl$ (0.5 M, pH 2) | 16 |
| Step 4 | Oxidizable | 5 mL $H_2O_2$ (30 %, pH 2) heat to 85 °C for 1 h, add 5 mL $H_2O_2$ (30 %) heat to 85 °C for 1 h, and then add 25 mL $NH_4OAc$ (1 M, pH 2) | 1, 1, 16 |
| Step 5 | Residual | HC1–$HNO_3$–HF–$HClO_4$ (graphite digestion system) | 7 |

sample. Following each extraction step, all suspensions were centrifuged and supernatants were filtered through a 0.45-μm membrane filter. After centrifugation and filtration, the residue was washed with deionized water and the filtrate was transferred to a volumetric flask. Then, the next extraction agent was added into the residue and started the next extraction process. The filtrate was acidified to pH<2 before analysis.

### Extraction experiments

Heavy metal extraction experiments were carried out in conical flasks in an aqueous suspension. The tests were performed under different pH (1~10), different molar ratio of IDS to the total metals in sludge range from 1:1 to 10:1, different contact times (2, 4, 6, 8, 10, 12, 24, 48 h), and in the presence of different concentrations of phosphoric acid in order to determine the optimum combination in terms of metal removal efficiency.

Extraction experiments were conducted in batch experiments at a solid (mass) to solution (volume) ratio of 1:50. For each batch, 0.5 g of sludge was suspended in 25 mL of solution, and the reaction time was fixed at 24 h except for the effect of contact time experiments. The pH of the suspension system was adjusted by $HNO_3$ or NaOH. The suspensions were shaken continuously at 200 rpm at room temperature, and centrifuged at 3,000 rpm for 20 min and filtered through 0.45-μm membrane filters before the analysis. The solutions were timely analyzed or stored at 4 °C prior to analysis.

### Analytical methods

The pH of the sludge was conducted with PHS-3C Professional pH Meter (Shanghai, China). The sludge sample was digested by SISP DS-360 graphite digestion apparatus (Guangzhou, China). The dissolved heavy metal concentrations in the digestion solutions and extraction solutions were measured using a Z-5000 flame atomic absorption spectrometry (Hitachi, Japan). The distribution of heavy metal fractions in the sludge samples were studied by the modified BCR sequential extraction method.

Triplicate extractions and analyses were performed for all samples.

## Results and discussion

### Characterization of sludge

Water content of sludge was 98.2 % and the pH of sludge was 11.5. Main metal elements contained in sludge were displayed in Table 2. The molar sum of the heavy metals Cd, Ni, Zn, Cu, and Cr in sludge was 1,395 mmol $kg^{-1}$. The content of Cd and Ni was 112,500 and 18,135 mg $kg^{-1}$, respectively, which outdistanced the Environment Quality Standard for soils (1 and 200 mg $kg^{-1}$ for Cd and Ni, respectively) and discharge standard of pollutants for municipal wastewater treatment plant in China (5 and 100 mg $kg^{-1}$ for acid soil of pH<6.5, and 20 and 200 mg $kg^{-1}$ for alkali soil of pH>6.5 for Cd and Ni, respectively). It is well-known that Cd is an environmental priority pollutant (Pueyo et al. 2004).

Species distribution of heavy metals in sludge, as determined by the sequential chemical extraction, was shown in Fig. 1. It can be observed that Cu and Zn in the sludge sample are mainly in both oxidizable and residual fractions. Cd mainly exists in oxidizable, acid-soluble, and residual fractions. For Ni, reducible fraction takes the largest proportion.

The metal relative content for the different fractions was found to be the following:

Water-soluble: Cu > Zn > Cd > Ni
Acid-soluble: Cd > Cu > Ni > Zn
Reducible: Ni > Cu > Zn > Cd
Oxidizable: Cu > Cd > Zn > Ni
Residual: Zn > Cu > Cd > Ni

The results showed that the reducible fraction represents the dominant part for Ni; this fraction is unstable and sensitive to the environmental conditions. On the contrary, Zn should mobilize much harder than the other metals, due to residual fraction that takes the highest proportion (about 56 %). For Cd

**Table 2** Heavy metal content of sludge sample and compared to legal standard

| Heavy metal | Cd | Cr | Pb | Ni | Zn | Cu |
|---|---|---|---|---|---|---|
| Value (mg $kg^{-1}$) | 112,500 | 31.5 | 0 | 18,135 | 1,700 | 237 |
| Acid soil ($pH<6.5$)[a] | 5 | 600 | 300 | 100 | 2,000 | 800 |
| Alkali soil ($pH\geq6.5$)[a] | 20 | 1,000 | 1,000 | 200 | 3,000 | 1,500 |
| Environmental soil[b] | 1 | 300–400 | 500 | 200 | 500 | 400 |

[a] National Standard of the People's Republic of China GB 18918–2002: discharge standard of pollutants for municipal wastewater treatment plant (mg $kg^{-1}$)

[b] National Standard of the People's Republic of China GB 15618–1995: environmental quality standard for soils (mg $kg^{-1}$)

and Cu, main fraction is oxidizable, which can be extracted partly. The results indicated that removal of these metals could be achieved under appropriate conditions.

## Effect of contact time on the extraction

In order to get optimal extraction efficiency of heavy metals, contact time versus removal efficiency was studied. The results indicated that the efficiency of extraction increased as contact time increased. After 24 h of treatment, no further removal of metals was observed (Fig. 2). Therefore, 24 h was chosen as the optimum reaction time in the following experiments, which is consistent with other reports (Tandy et al. 2004). This can be explained that when the chelating process reaches equilibrium, the removal efficiency is no change though the contact time increased.

## Effect of pH on the extraction

As a crucial factor that affects extraction, the effect of pH on extraction was studied. The comparison of the removal efficiency of heavy metals at different pH in the presence and absence of IDS were investigated. As shown in Fig. 3a, the removal efficiency of heavy metals decreased rapidly as pH increased in the absence of IDS. Same as shown in Fig. 3b, although the extraction efficiency was improved by addition of IDS over a wide pH range from 2 to 10, the removal efficiency of heavy metals also decreased as pH increased in the presence of IDS (molar ratio of IDS to heavy metals was 1:1). The extraction efficiencies at $pH=2.5$ in the absence of IDS were 42, 21, 67, and 56 % for Cd, Ni, Cu, and Zn, respectively. While at the same pH, the extraction efficiencies in the presence of IDS were 46, 60, 91, and 49 % for Cd, Ni, Cu, and Zn, respectively. The results indicated that IDS could extract heavy metals (exception of Zn) from sludge well. It should be noted that strong acid condition was suited for metal removal. Interestingly, the extraction efficiency of Cd was dramatically affected by IDS in alkaline conditions.

## Effect of IDS concentration on the extraction

In order to examine IDS concentration on heavy metals extraction, different molar ratios of IDS to the total metals in sludge range from 1:1 to 10:1 under different pH were investigated. Generally, increasing IDS concentration was beneficial to heavy metal extraction. As

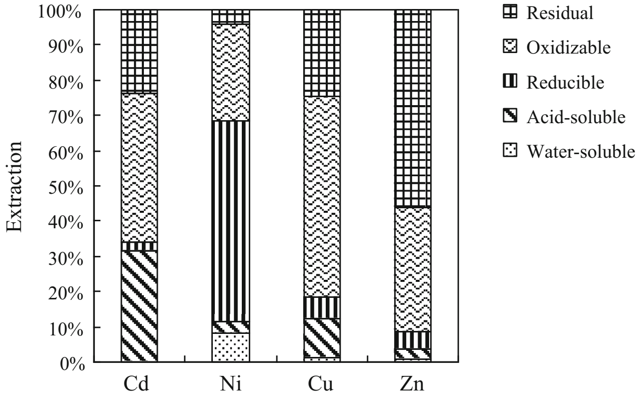


**Fig. 1** Species distribution of heavy metals in sludge sample

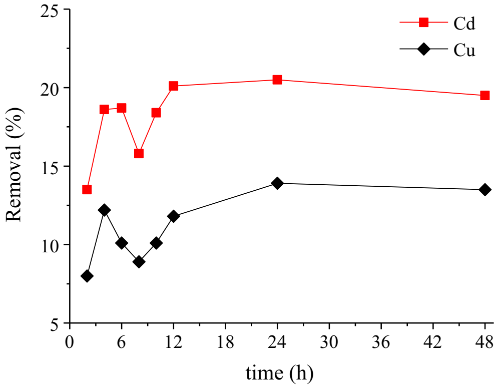


**Fig. 2** Effect of contact time on the extraction

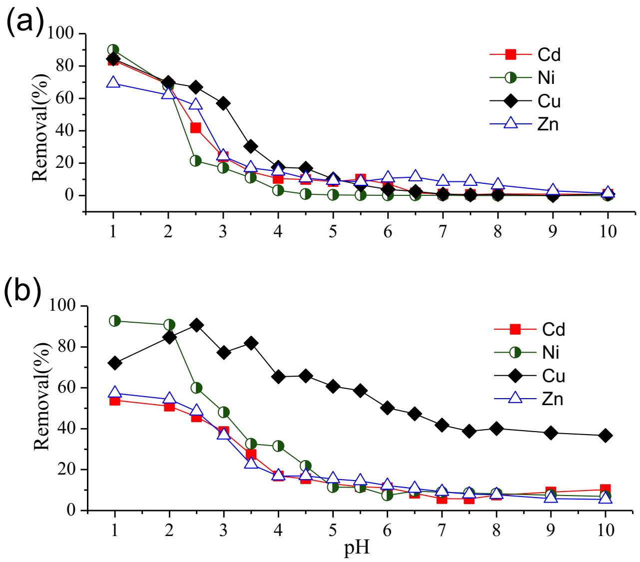


**Fig. 3** Effect of pH on the extraction: **a** without IDS and **b** molar ratio of IDS:metal=1:1

shown in Fig. 4, extraction trend was similar at pH 11.5 and 7, both followed the sequence Cd > Cu > Zn > Ni (Fig. 4a, b). Interestingly, great improvements occurred before the molar ratio reached 7 for Cd in alkaline medium, and about 68 % of Cd was extracted at the molar ratio of 7:1 (Fig. 4a). At acidic medium (pH 4), metal extraction changed greatly; the order of efficiency was Cu > Ni > Cd > Zn. For Ni and Cu, the extraction efficiency increased as IDS concentration increased molar ratio from 1:1 to 3:1. Cd and Zn maintained a lower extraction rates throughout the experiment (Fig. 4c).

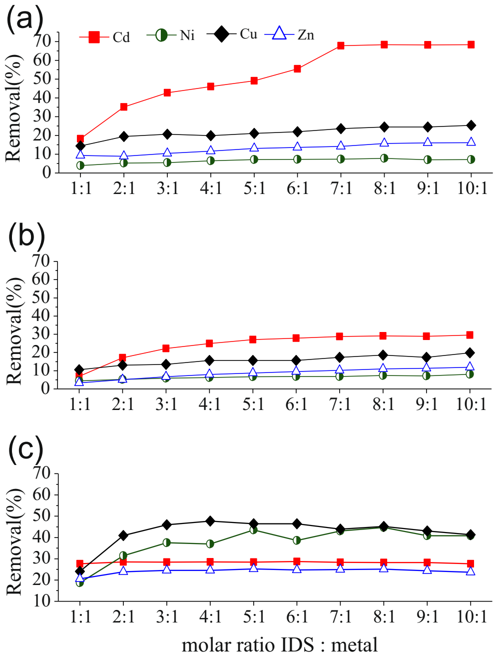


**Fig. 4** Effect of IDS concentration on the extraction under different pH: **a** natural pH, **b** pH=7, and **c** pH=4

IDS is a pentadentate N,O donor ligand forming chelates of octahedral structure with many metal ions. The reaction between the metal ion and the anion of IDS acid is reversible and occurs with the ratio 1:1 (Kołodyńska et al. 2009).

$$M^m + \mathrm{ids}^n \rightleftarrows [\mathrm{M(ids)}]^{(n-m)-} \tag{1}$$

The theoretical ratio of chelant to metal was 1:1, but actually, the ratio achieved 7:1, which could be explained by the fact that there were many substances in the sludge, not only with the heavy metals that we studied but also with other non-target metals. In the process of complexing target heavy metals, those non-target metals compete with the target heavy metals; thus, the consumption of chelating agent increased greatly. Therefore, higher extraction efficiency could be reached at molar ratio more than 7:1 for Cd at the natural pH.

For the four possible equilibrium states:

$$\mathrm{H_3ids^-} + \mathrm{H^+} \rightleftarrows \mathrm{H_4ids} \tag{2}$$

$$\mathrm{H_2ids^{2-}} + \mathrm{H^+} \rightleftarrows \mathrm{H_3ids^-} \tag{3}$$

$$\mathrm{Hids^{3-}} + \mathrm{H^+} \rightleftarrows \mathrm{H_2ids^{2-}} \tag{4}$$

$$\mathrm{ids^{4-}} + \mathrm{H^+} \rightleftarrows \mathrm{Hids^{3-}} \tag{5}$$

IDS is in the form of $\mathrm{Hids^{3-}}$ at around pH 7, protonated or deprotonated with decreasing or increasing pH (Kołodyńska 2009). Kołodyńska's research mentioned that stability

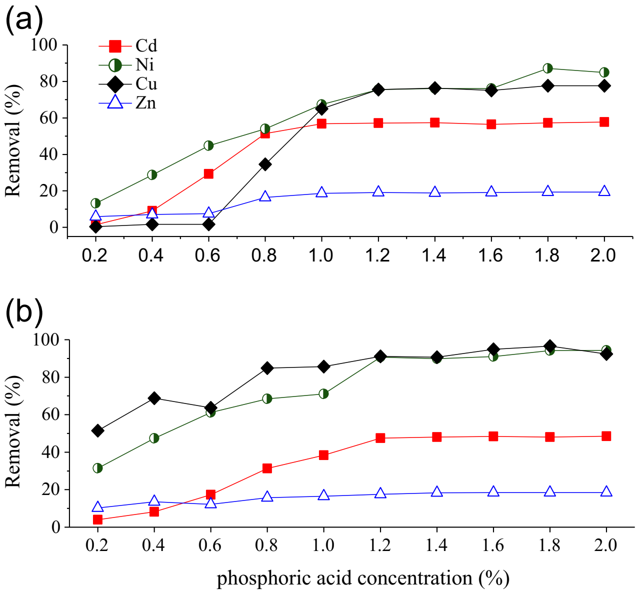


**Fig. 5** Effect of phosphoric acid on the extraction: **a** $H_3PO_4$ and **b** $H_3PO_4$–IDS

constants (log K) of metal complexes with IDS (L) were: log $K_{CuL}$(13.1)>log $K_{NiL}$(12.2)>log $K_{ZnL}$(10.8)>log $K_{CdL}$(8.4) (Kołodyńska 2013). IDS has a strong capability to chelate Cu, Cd, Zn, and Ni and chelating ability follow the sequence as follows: Cu > Ni > Zn > Cd. However, the results indicated that actual chelation affected pH of system, chelating agent's concentration, and speciation distribution of metals.

### Effect of phosphoric acid on the extraction

Phosphoric acid is a component in the human body, and it is safe for human beings who are engaged in sewage treatment work (Yoshizaki and Tomida 2000). The effects of phosphoric acid on the removal of heavy metals from a sludge sample in the presence of IDS were studied. The extraction efficiency increased quickly before phosphoric acid concentration reached 1.2 % (Fig. 5a). IDS–$H_3PO_4$ system greatly improved Ni and Cu extraction. It could be observed that removal efficiency of IDS (molar ratio 1:1) with 1.2 % phosphoric acid was much higher than phosphoric acid and IDS individually; about 90.7 and 91.3 % of Ni and Cu were extracted, respectively (Fig. 5b). On the contrary, IDS–$H_3PO_4$ system is not conducive to Cd extraction. For Zn, extraction in phosphoric acid was slightly better than IDS–$H_3PO_4$ system, but both were low; residual fraction takes dominant part (nearly 60 %) result in low removal efficiency.

## Conclusions

IDS, as an environmental benign biodegradable chelant, was applicable to remove heavy metals from industrial sludge effectively. Factors (contact time, IDS concentration, pH, and speciation of heavy metals) would affect the extraction of heavy metals were studied. Although stability constant of Cd is inferior to Zn, Cd was easier to be extracted than Zn actually. In the whole experimental process, low extraction efficiency of Zn result from nearly 60 % residual fraction of Zn exists in sludge. Therefore, chelating properties is related not only to stability constant but also to species distribution of metals. About 68 % of Cd in the sample was extracted at a molar ratio of 7:1 (IDS:metal) in natural pH. Ni and Cu could be extracted well by IDS (molar ratio, IDS:metal=1:1) with low concentration of phosphoric acid (1.2 %), and the removal efficiencies were 90.7 and 91.3 %, respectively, one reason attribute to low proportion of residual fraction of these two metals; the other one is that Ni and Cu have strong chelating ability with IDS and phosphoric acid facilitation function. The results suggested that IDS can be a potentially useful chelant for Cu, Ni, and Cd removal from battery industry sludge with minimal reagent consumption and under mild conditions; this is consistent with green chemistry and sustainable development idea.

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