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Temporary stabilization of air pollution control residues using carbonation

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Abstract

Carbonation presents a good prospect for stabilizing alkaline waste materials. The risk of metal leaching from carbonated waste was investigated in the present study; in particular, the effect of the carbonation process and leachate pH on the leaching toxicity of the alkaline air pollution control (APC) residues from municipal solid waste incinerator was evaluated. The pH varying test was conducted to characterize the leaching characteristics of the raw and carbonated residue over a broad range of pH. Partial least square modeling and thermodynamic modeling using Visual MINTEQ were applied to highlight the significant process parameters that controlled metal leaching from the carbonated residue. By lowering the pH to 8–11, the carbonation process reduced markedly the leaching toxicity of the alkaline APC residue; however, the treated APC residue showed similar potential risk of heavy metal release as the raw ash when subjected to an acid shock. The carbonated waste could, thereby, not be disposed of safely. Nonetheless, carbonation could be applied as a temporary stabilization process for heavy metals in APC residues in order to reduce the leaching risk during its transportation and storage before final disposal.

1. Introduction

Carbonation has been recognized to be an important process affecting alkaline materials (waste) such as bottom ash from municipal solid waste incinerators (MSWI) and cement-stabilized waste (Chimenos et al., 2000; Garrabrants et al., 2004; Gervais et al., 2004; Meima et al., 2002; Van Gerven et al., 2005; Walton et al., 1997). It has also been proposed as a means to stabilize Pb and Zn in air pollution control (APC) residues from MSWI (Bone et al., 2003; Ecke et al., 2003a; Kim et al., 2003). The effect of carbonation on the leaching toxicity of these samples was intensively investigated (Freyssinet et al., 2002; Meima and Comans, 1998; Polettini and Pomi, 2004; Van Gerven et al., 2004, 2005; Yu et al., 2005), which could be summarized as: (1) lowering pH of the leachate; (2) changing the metal solubility due to precipitation of metal carbonates or formation of oxyanions (Cr and Mo); (3) reducing release of certain metals (Cu and Mo) by sorption to the neoformed minerals (in 1.5-year weathered bottom ash); and (4) decreasing the matrix porosity because of the formation of calcite.

However, certain leaching results have been interpreted based on single batch tests, which may obtain discrepant results (Astrup et al., 2006; Li et al., 2001). Detailed leaching behavior of uncarbonated and carbonated ash has been investigated using pH varying test or semi-dynamic leaching tests (Garrabrants et al., 2004; Meima et al., 2002; Van Gerven et al., 2005, 2006); most of these tested samples were bottom ash or cement-stabilized ash. Leaching behavior of carbonated APC residues without cement stabilization is rarely discussed. Furthermore, it is inconclusive whether the change in metals retention properties are due to the precipitation of metal-carbonates as some researchers have suggested (Bone et al., 2003; Freyssinet et al., 2002) or to a shift in pore water pH as a result of

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carbonation which changed the solubility of metals (Sanchez et al., 2002; Van Gerven et al., 2004). Therefore, the effects of environmental change on the leaching potential of carbonated APC residues have not been satisfactorily explored.

To address these problems, a previous study (He et al., 2006a) examined the leaching behaviors of the APC residue during the natural aging process evaluated by three kinds of regulatory leaching tests. It was demonstrated that both leaching tests and carbonation substantially affected the leaching results. Model calculations based on the geochemical thermodynamic equilibrium model MINTEQA2 suggested that the formation of metal carbonates did not correspond to the noted change in the leaching behaviors. Rather, the partial neutralization of alkaline ash by dissolved CO_2, lowering the final pH of the leachate, dominated the leaching characteristics. Metals immobilized by carbonation will be released again under an acidic environment. However, the experimental proof to demonstrate this proposal remained preliminary.

This work is different from He et al. (2006a) in the following three aspects. (1) The pH varying test was conducted over pH 0.4–12.9 to demonstrate the leaching characteristics of raw and carbonated samples. (2) Effects of carbonation were analyzed using partial least square (PLS) modeling (Umetrics AB, 2005) of experimental data and using thermodynamic modeling, Visual MINTEQ (Allison et al., 1991; Gustafsson, 2005), considering redox reactions for illustrating the impact of carbonation on leaching characteristics. (3) Based on the experimental findings, the use of waste carbonation as a temporary pretreatment process for heavy metal stabilization was proposed.

2. Methods and materials

2.1. Residues

The APC residue samples were collected from the flue gas treatment units of a MSW incineration plant (mass burn) with a treatment capacity of approximately 1200 t/d in Shanghai City, China, equipped with a semi-dry reactor with lime slurry injection, activated carbon adsorption, and bag filters to remove acid gas, heavy metals, dioxins, and particulate matter, respectively. Table 1 lists the chemical compositions of the raw residue. Due to the presence of a semi-dry process unit, calcium was the most abundant element in the APC residue. Moreover, the residue was composed of Cl^-, K, Na, Fe, Mg, SO_4^{2-}, and CO_3^{2-}. The concentrations of trace elements followed the order: Zn > Pb > Cu > Cr > Ni > Cd > As > Hg.

Table 1
Raw APC residue composition

Major components	Content (wt.%)	Trace elements	Content (mg/kg)
Ca	30 ± 1.4	As	71 ± 1.5
K	3.2 ± 0.04	Cd	57 ± 0.2
Na	2.9 ± 0.04	Cr	450 ± 21
Fe	3.0 ± 0.07	Cu	980 ± 19
Mg	1.4 ± 0.02	Hg	39 ± 4.2
Cl^-	9.4 ± 0.10	Ni	130 ± 2.0
SO_4^{2-}	1.5 ± 0.03	Pb	2620 ± 60
CO_3^{2-}	1.4 ± 0.14	Zn	5400 ± 140
PO_4^{3-}	<0.001	Mn	1000 ± 5

The raw APC residue was mixed with distilled water to make a sample of 20 wt.% moisture content, which was fed into two columns (Φ 5 cm × 25 cm), with a 500-g residue sample in each. In column 1, the residue was carbonated at accelerated rates by flowing through pure CO_2 at a rate of 0.4 m^3/h for 10 h (rapidly carbonated). In column 2, air (0.03% v/v CO_2) was flowing through at 0.4 m^3/h for 7 days to simulate a normal aging process (slowly carbonated).

The amounts of carbonate (CO_3^{2-}) in raw, rapidly and slowly carbonated residues were measured by mixing 1 g of residue with 10 ml of 1 mol/l HNO_3, and then analyzing the CO_2 gas release using a gas chromatograph (GC102, Shanghai Analytical Instrument Overall Factory, China).

2.2. Leaching test

Leaching tests were conducted based on a regulatory leaching test (State Environmental Protection Administration of China, 1997); 100 g of each residue sample (raw, rapidly and slowly carbonated) was mixed with 1 l of distilled water (liquid to solid ratio, L/S = 10 l/kg) and tumbled at 30 ± 2 rpm for 18 h. The mixture was vacuum-filtered through a 0.45-μm membrane. The leachate pH and ORP were recorded and the concentrations of Cd, Cr, Cu, Ni, Pb and Zn in the filtrate were then analyzed using an atomic absorption spectrophotometer (AAS), while those of As and Hg were analyzed using an atomic fluorescence spectrometer (AFS).

The pH varying test was carried out to determine the pH-dependent leaching behavior of the residues (van der Sloot et al., 1997). The raw residue sample was equally divided into 15 parts, each of 30-g by weight, which were extracted individually for 48 h by using 300-ml of NaOH solutions of 0.5, 0.2, 0.1, 0.05 mol/l, distilled water, and HNO_3 solutions of concentrations of 0.1, 0.2, 0.4, 0.6, 0.8, 1.0, 1.2, 1.5, 1.8, and 2.0 mol/l, respectively. A similar extraction procedure was adopted to the rapidly and slowly carbonated residues; however, they were each divided into only 10 parts, which were individually extracted by using NaOH solutions of concentrations of 0.5, 0.2, 0.05 mol/l, distilled water, and HNO_3 solutions of 0.2, 0.4, 0.5, 0.6, 0.8, 1.0 mol/l, respectively. The final pH and the metal concentrations in the filtrates were measured.

The variability of Hg and As concentration in duplicates was within 10%, and other heavy metals concentration within 3%.

2.3. Leaching modeling

Solubility of carbonates is greatly dependent on leachate pH. Therefore, Visual MINTEQ was applied herein to determine the chemical stability of pure metal carbonates as a function of pH, by solely inputting the metal carbonates as finite solid.

The equilibrium species of metals and As leached from the real carbonated samples were also modeled, by inputting in Visual MINTEQ the aqueous ion concentrations, and the pH and Eh values (−18 mV for slowly carbonated ash and 70 mV for rapidly carbonated ash) measured in the regulatory leaching test, allowing precipitation/dissolution, redox, aqueous complexation reactions. The input molar concentrations for each component (Al^{3+}, Ca^{2+}, Cd^{2+}, $Cr(OH)_2^+$, Cu^{2+}, Fe^{3+}, H_3AsO_4, Hg_2^{2+}, H_4SiO_4, Mg^{2+}, Mn^{2+}, Ni^{2+}, Pb^{2+}, Zn^{2+}, Cl^-, CO_3^{2-}, SO_4^{2-}, PO_4^{3-}) were based on the determined total amount (Table 1) divided by the L/S ratio 10 and the molar mass of the component. The input redox couples included H_3AsO_3/AsO_4^{3-}, $Cr(OH)_2^+/CrO_4^{2-}$, $Cr^{2+}/Cr(OH)_2^+$, Cu^+/Cu^{2+}, Fe^{2+}/Fe^{3+}, $Hg_2^{2+}/Hg(OH)_2$, HS^-/SO_4^{2-}. The log K value of $Pb(OH)_2$ concentration was taken as −10.15, from van der Bruggen et al. (1998). The other log K values referred to the default values in Visual MINTEQ. This work disregards the role of gas phase and surface complexation/precipitation (Eighmy et al., 1995; van der Bruggen et al., 1998; van Herck et al., 2000). The temperature in calculations was set at 25 °C.

2.4. Multivariate data analysis

PLS modeling was used to highlight the inter-relationships between three process parameters (factors), being extractant concentrations (negative for alkaline and positive for acid solution), extent of carbonation (rapid or slow, denoted by carbonate content), and leachate pH, on the leaching behavior (response variables).

3. Results

3.1. Stabilization of heavy metals and As in the APC residue using carbonation

During carbonation, CO_2 was absorbed and reacted with alkaline compounds in the residue. After 10 h of rapid carbonation, the CO_3^{2-} content in the residue increased from 1.43 wt.% to 8.85 wt.%, comparable to the result of 7.2 wt.% in Van Gerven et al. (2005), and the leachate pH (extracted by distilled water at $L/S = 10$ l/kg) decreased from 12.0 to 8.7 (Fig. 1), which was comparable to the carbonation level of natural aging for more than 1 month (He et al., 2006a). Slow carbonation for 7 days, however, obtained a lower carbonation level, revealed by the low CO_3^{2-} content of 4.08 wt.% and relatively high leachate pH of 10.1.

After carbonation, the acid neutralization capacity of the ash decreased from 4.2 mmol-H^+/g-ash to 3.9 and 3.3 mmol-H^+/g-ash for slowly carbonated and rapidly carbonated ash, respectively (data not shown), taken pH 7 as endpoint.

Fig. 1 presents the leached concentrations of metals and As from the raw and carbonated APC residues. The concentrations of Hg and Pb leached from the raw APC residue, particularly the latter, were higher than the limit values (0.05 and 3 mg/l, respectively) set by the Chinese identification standard for hazardous waste (State Environmental Protection Administration of China, 1996). Hence, the raw ash was classified as hazardous and might pose risk to environmental safety and public health following disposal (Chandler et al., 1997).

The leaching behavior of metals and As following carbonation could be categorized into three types: (raw residue ≈ carbonated residue) As (around the limit value), Cd, and Cu; (raw residue > carbonated residue) Hg, Ni, Pb and Zn; and (raw residue < carbonated residue) Cr only. After carbonation, the leachability of Hg and Pb had become much lower than the corresponding limit values, and Zn was not even detected in the leachate of the carbonated residues. The carbonated residues would be then classified as non-hazardous for disposal. Based on similar observations, Ecke (2003b), Kim et al. (2003), and Meima and Comans (1999) proposed that carbonation could be used as a stabilization technique for Pb and Zn in APC residues or bottom ash. As reported in the following section, using Visual MINTEQ modeling, we examined whether the heavy metals in the APC residue were transformed into more stable carbonate or oxide forms following carbonation, or simply some hydroxides after pH changes.

3.2. Change of metal speciation during the carbonation process

Based on X-ray diffraction (XRD) analysis, He et al. (2006b) observed the increase of calcite and gypsum in the carbonated residues; however, heavy metal carbonates were not detected by XRD because of their trace amount. As such, it is also difficult to determine directly the changes of heavy metal species in the carbonated ash by other nondestructive analytical methods. Visual MINTEQ is an equilibrium speciation model used to calculate the equilibrium composition of aqueous solutions. Thus, it can be used to simulate aqueous speciation of the carbonated product subjected to a leaching system, although it is incapable of indicating reactions occurred in the carbonation process.

Fig. 2 illustrates the modeled pH-dependent leaching behavior of pure metal carbonates. Calculations for As and Cr are lacking since equilibrium data for their carbonates were not available. In a fully carbonated system, leachate pH of the APC residue could reach a value as low as 8.3 (Bone et al., 2003). Within this pH range 8–13, carbonates of all heavy metals are relatively stable except $CdCO_3$, which becomes significantly soluble at $pH < 10$. When pH further decreases to less than 6, carbonates turn into soluble species following the order $CdCO_3 > NiCO_3 > ZnCO_3 > PbCO_3 > HgCO_3 > CuCO_3$, suggesting carbonated

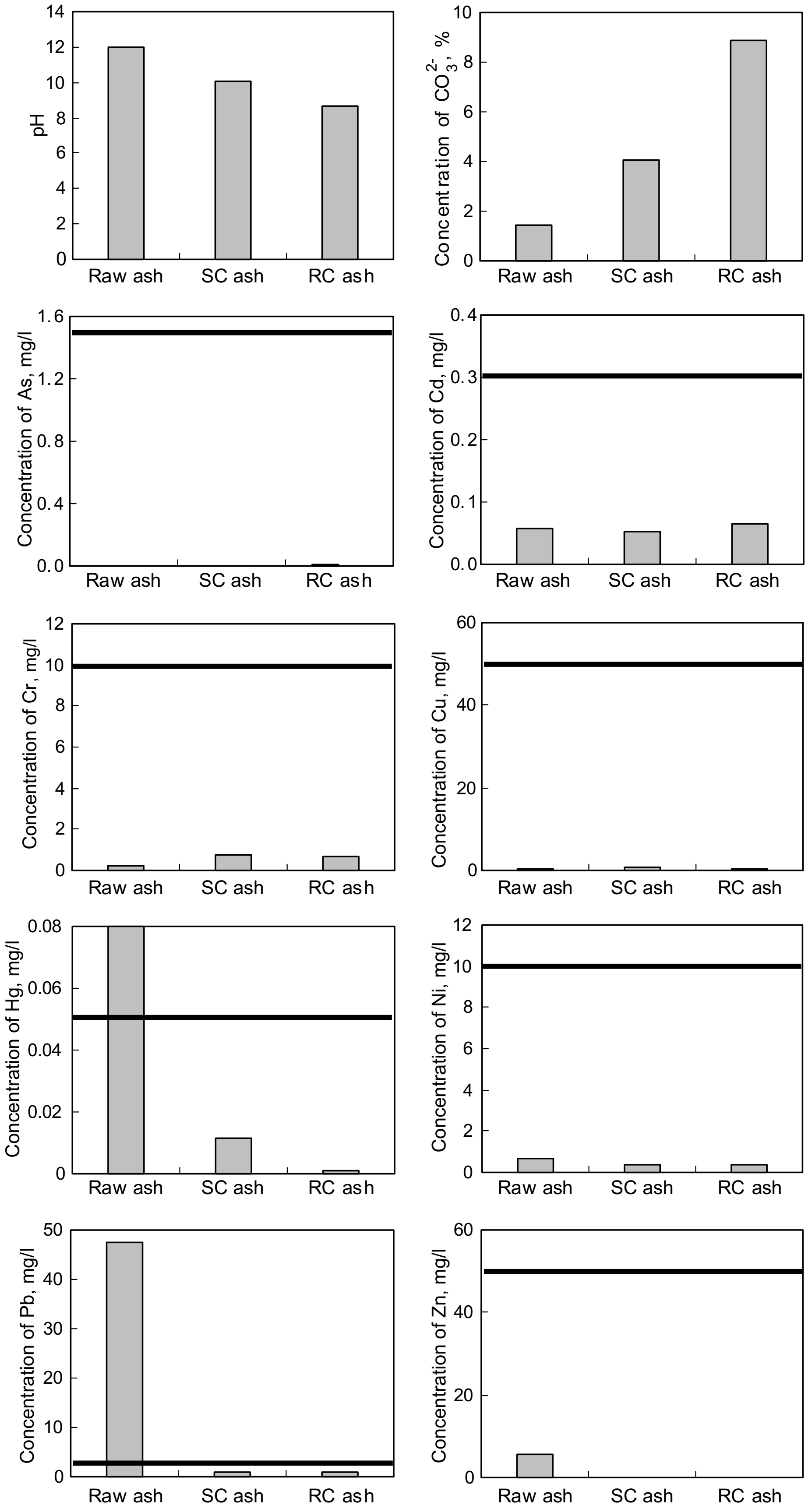

Fig. 1. Leaching characteristics of the raw, slowly carbonated (SC) and rapidly carbonated (RC) ash. The broad-brush solid line is the limit value set by the identification standard for hazardous waste (State Environmental Protection Administration of China, 1996).

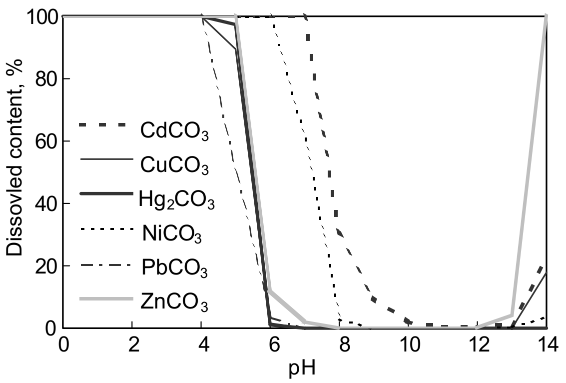

Fig. 2. Modeled pH-dependent leaching behavior of pure metal carbonates.

products will dissolve to a large extent when they encounter an acid shock.

In the leaching system of APC residue, much more complex results, as the combined effect of oxidation/reduction, hydration, competitive precipitation/dissolution and complexation, would be observed. Tables 2 and 3 summarize the modeling results of the carbonated residues under the extraction conditions of the regulatory leaching test. The release ratios of metals and As obtained by direct measurement and modeling were compared.

The major precipitates in the raw APC residue included CaO, $CaCO_3$, SiO_2, $CaSO_4 \cdot 2H_2O$, $Mg_3Si_2O_7 \cdot 2H_2O$, and Fe_2O_3, among which the former three species and anhydrite ($CaSO_4$) had been detected by XRD (He et al., 2004). After carbonation, the content of $CaCO_3$ and gypsum ($CaSO_4 \cdot 2H_2O$) significantly increased, consistent with the XRD analytical results (He et al., 2006b).

Most of the modeling results, except for Cd, were in agreement with the experimental results that the carbonated ash was of less leaching toxicity. Predicted Cd concentration was significantly discrepant with the observed value. According to Visual MINTEQ, Cd could completely release from the rapidly carbonated ash at pH 8.7; however, only 1.14% of Cd was leached from the ash in the real extraction system. Apul et al. (2005) and Meima and Comans (1998) had also found that the predicted Cd concentrations could be one order of magnitude higher than the measured concentrations even when a surface complexation model by hydrous ferric oxide (HFO) and aluminum (hydr)oxides was included. The use of a surface precipitation model did not significantly improve the match between observed and predicted concentrations either. The modeling results for Cd are still inconclusive.

As the model predicted, heavy metals of the carbonated residues precipitated in the leaching system mainly as hydroxide or oxide, $Cd_4(OH)_6SO_4$, Cr_2O_3, $Ni(OH)_2$, $Pb_2(OH)_3Cl$, ZnO, respectively. As, Cu, Hg formed insoluble $Ca_3(AsO_4)_2 \cdot 4H_2O$ (slowly carbonated ash) or $Mn_3(AsO_4)_2 \cdot 8H_2O$ (rapidly carbonated ash), $CuFeO_2$ and Hg in the elute. No heavy metal carbonates precipitated, since CO_3^{2-} preferentially reacted with Ca^{2+} and Mg^{2+} to form $CaCO_3$ and $MgCO_3$ and precipitated from the leachate.

Table 2
Species of major precipitates in the raw and carbonated APC residues based on modeling results

Raw ash (pH = 12.0)		Slowly carbonated ash (pH = 10.1)		Rapidly carbonated ash (pH = 8.7)	
Major precipitates	Concentration (mol/l)	Major precipitates	Concentration (mol/l)	Major precipitates	Concentration (mol/l)
CaO	4.91E − 01				
Fe_2O_3	2.60E − 02				
SiO_2	2.41E − 01	SiO_2	2.62E − 01	SiO_2	2.63E − 01
$Mg_3Si_2O_7 \cdot 2H_2O$	1.86E − 02	$Mg_3Si_2O_7 \cdot 2H_2O$	1.86E − 02	$Mg_3Si_2O_7 \cdot 2H_2O$	1.85E − 02
$CaSO_4 \cdot 2H_2O$	8.70E − 03	$CaSO_4 \cdot 2H_2O$	1.10E − 02	$CaSO_4 \cdot 2H_2O$	1.07E − 02
$CaCO_3$	2.38E − 02	$CaCO_3$	6.79E − 02	$CaCO_3$	1.48E − 01
		$MgCO_3$	1.73E − 02	$MgCO_3$	1.73E − 02

Table 3
Heavy metals and As released from the carbonated APC residues based on experimental and modeling results

	Slowly carbonated ash			Rapidly carbonated ash		
	Measured release (%)	Modeled release[a] (%)	Precipitates	Measured release (%)	Modeled release[a] (%)	Precipitates
As	0.00	0.29	$Ca_3(AsO_4)_2 \cdot 4H_2O$	0.14	1.01	$Mn_3(AsO_4)_2 \cdot 8H_2O$
Cd	0.91	4.45	$Cd_4(OH)_6SO_4$	1.14	100	–
Cr	1.60	0.01	Cr_2O_3	1.43	0.01	Cr_2O_3
Cu	0.58	0.00	$CuFeO_2$	0.54	0.00	$CuFeO_2$
Hg	0.30	0.25	Hg	0.03	0.26	Hg
Ni	2.98	0.00	$Ni(OH)_2$	2.92	0.28	$Ni(OH)_2$
Pb	0.31	0.01	$Pb_2(OH)_3Cl$	0.31	0.01	$Pb_2(OH)_3Cl$
Zn	0.00	0.02	ZnO	0.00	0.04	ZnO

[a] As predicted in Visual MINTEQ.

Future experiments to define the effect of surface complexation and precipitation on the release rates of heavy metals are desirable to improve the match between observed and predicted concentrations.

3.3. Effect of carbonation and pH on the leaching behavior of the APC residues

Fig. 3 demonstrates the pH varying test results of the raw, slowly carbonated and rapidly carbonated residue samples. Regardless of carbonation, all residue samples possessed similar pH-dependent leaching behavior of individual metals and As.

Although complex reactions might occur in carbonation and leaching processes, the final pH of leachate, a result of combined effects of all reactions/processes, presents the major index influencing leaching equilibrium. As shown in Fig. 3, the heavy metals and As release curves can be divided into two main groups in the range pH 0–13: V shaped (Cd, Cr, Hg, Ni, Pb and Zn) and L shaped (As and Cu). With final suspension pH decreasing from 12.0 to 8.7, the concentration ranges of As and Cu are located

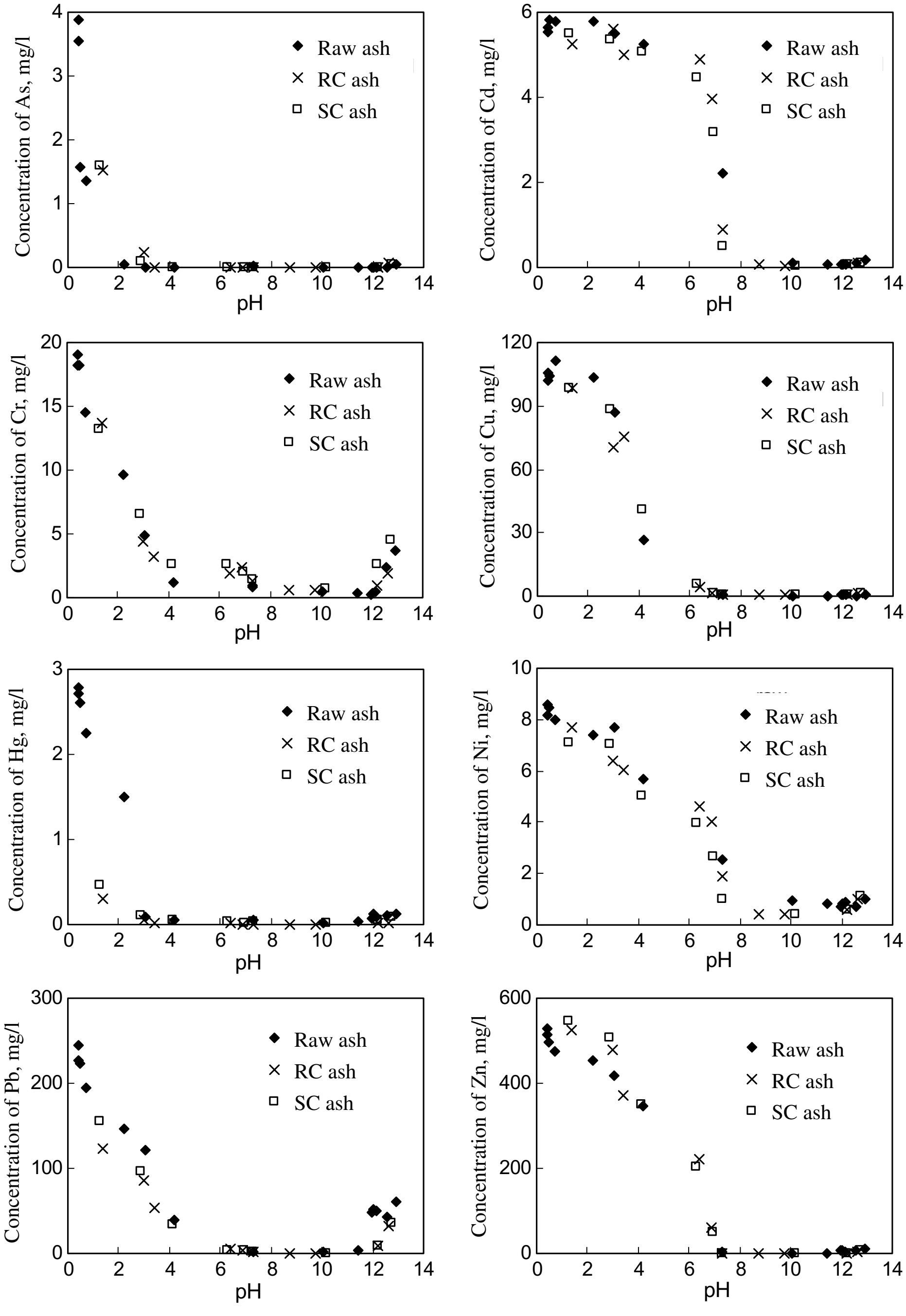

Fig. 3. pH-dependent leaching behavior of the raw, slowly carbonated (SC) and rapidly carbonated (RC) ash.

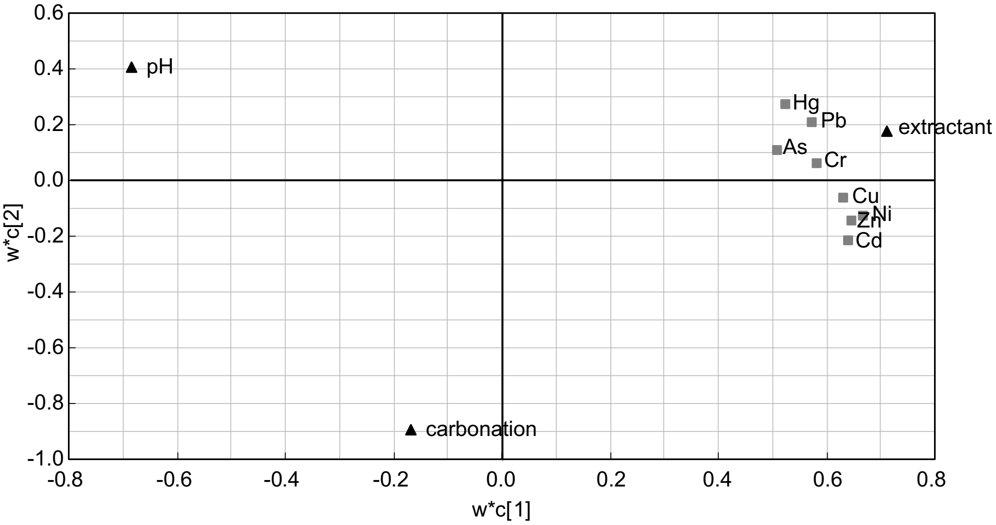

Fig. 4. PLS loading plot of the first and second principal components using data from pH varying tests of the raw and carbonated ashes.

at the alkaline side of the L curves, showing only weak fluctuation during carbonation. In the pH range of 8–13, Cd, Ni, Hg, Pb, and Zn leaching is situated on the right leg of the V curves, so their leaching concentrations decreased as pH lowers (or with carbonation). On the contrary, the Cr leaching is located in the left part of the V curve, where leaching concentration increases when pH approaches 8.7 when coming from more alkaline conditions.

Similar to the modeled pH-dependent result of pure metal carbonates (Fig. 2), within the pH range 8–11, As and heavy metals were relatively stable. When pH decreased to less than 7, however, a significant increase of Cd, Ni, and Zn leaching was observed. At pH 6, Cu and Pb became more leachable. As, Cr, and Hg were relatively stable under weak acidic conditions. From the regulatory viewpoint, the carbonated APC residue was not really "stabilized" and could not be disposed of directly, with regard to its release potential under acid shock ($pH < 6–7$). On the other hand, rapid carbonation (10 h) or slow carbonation (7 d) might be favorably used as a simple pretreatment process to control pollution before the final treatment and disposal of ash, by decreasing pH of the carbonated residue to 8–11.

3.4. PLS modeling

The PLS analysis is a regression algorithm relating input and output samples (x_i, y_i) by a linear multivariate model (Nadler and Coifman, 2005). It obtains PLS-weights for the variables. The weights for the X-variables indicate the importance of these variables, how much they "in a relative sense" participate in the modeling of Y, while the weights for the Y-variables indicate which Y-variables are modeled in the respective PLS model dimension. When these weights are plotted in a PLS loading plot, we obtain a picture showing the relationships between X and Y-variables, which X-variables are important, and which Y-variables are related to which X, etc. (Umetrics AB, 2005).

In this study, the PLS modeling taking into account extractant concentration, carbonation level, and leachate pH, resulted in two principal components comprising 71% and 2.5% of the data variation. The first principal component dominantly explained data variation.

The loading plot (Fig. 4) illustrates the impact of various factors (extractant concentration, leachate pH, and carbonation level) on the response variables (concentration of metals and As). The metals and As release depended significantly on the extractant (acid or alkaline) concentration and leachate pH. Compared to the positive correlation between extractant concentration and heavy metals release, leachate pH (which is the combining result of the extractant concentration, carbonation level, and leaching process), was negatively correlated to metals leaching concentration. The carbonation level dominating the second principal component, which covered only 2.5% of the data variation, was inconclusive.

All metals and As showed similar variation. If further subdivided according to their X-weight, Cu, Cd, Ni, and Zn were more significantly influenced by extractant and leachate pH. This observation corresponds to the results shown in Fig. 3, that these metals were more acid-leachable, with rising release when pH fell to less than 7. As, Hg, Cr, and Pb were relatively resistant to pH decrease, whose leachability increased at pH less than 4–6.

4. Conclusions

After rapid and slow carbonation, the CO_3^{2-} content in the APC residue increased from 1.43 wt.% to 8.85 wt.% and 4.08 wt.%, respectively, and the leachate pH of the rapidly and slowly carbonated ash decreased to 8.7 and 10.1, respectively. The leaching toxicity test showed that carbonation could significantly immobilize heavy metals (Hg, Pb and Zn) and turned the hazardous materials into "non-hazardous" waste. A slight decrease of Ni leaching and increase of Cr was found, while Cd and Cu leaching concentration remained stable, all of them were far below the limit values. However, the so-called stabilization was mainly due to the decrease of leachate pH controlled by the carbonation in the short-term, which finally influenced the solubility of metals, according to the results of pH varying test, PLS analysis, and Visual MINTEQ modeling. Although the long-term leaching potential of the carbonated APC residue was of concern when the residue was under an acid shock, carbonation in air via water spraying could be applied as a temporary stabilization stage during transportation and storage before final disposal.

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