Extraction of silica gel from rice husk ash to stabilize Pb and Zn in municipal solid waste incineration fly ash

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Abstract: The present research has focused on producing low-cost material from agricultural waste (rice husk ash: RHA) as an alternative for stabilizing heavy metals in municipal solid waste incineration (MSWI) fly ash. Silica gel extracted from RHA has 97.47 % SiO₂. The silica gel product was able to stabilize 99 % of both Pb and Zn. The results show that the silica gel can be used as an effective and low-cost stabilizer of heavy metals in MSWI fly ash. *Keyword: silica gel, rice husk ash, MSWI fly ash, heavy metal*

1. Introduction

Rice husk (RH) is a waste material from the rice processing industry. The ingredient of RH depends on the soil condition, rice species, and fertilizer ingredients. The burning or calcination of RH leads to the production of rice husk ash (RHA) composed of approximately 80–90% silica. Typically, silica in RHA is amorphous and has a large surface area (Shelke et al., 2010). RHA is very useful with added value that can be used as a raw silica-rich material due to its chemical and physical properties.

The amount of municipal solid waste (MSW) is generated rapidly as the world's population grows. The proper treatment and disposal of MSW are essential, and MSW incineration technology has rapidly become expanded in many countries for MSW disposal. However, large amounts of heavy metals, soluble salts, and other pollutants accumulate in fly ash (FA) residues after incineration (Bell & Petrlik, 2017). Therefore, fly ash is classified as a hazardous waste. So, proper treatment of FA is the key to control environmental hazards. This study aimed to develop a procedure to produce silica gel from RHA and determine the stabilization of Pb and Zn from MSWI fly ash.

2. Materials and methods

2.1 RHA collection and preparation

RH was collected from a local rice mill in the Itoshima area, Fukuoka, Japan. RH was calcined in a muffle furnace at 450 $^{\circ}$ C for 15 min. After calcination, a black product called RHA was generated.

2.2 Extraction of silica gel from rice husk ash

1 M NaOH was added to RHA at a liquid/solid (L/S) ratio of 10 mL/1 g and stirred at 100 °C for 2 h. Then, the solution was cooled and filtered. 2 M HCl was slowly added to the filtrate to form a gel until pH 10. The gel was aged for 24 h and washed with distilled water several times to remove NaCl. Then, the gel was filtered and oven-dried at 105 °C for 24 h.

2.3 Stabilization experiments

Mixtures of FA and silica gel were prepared by weight at FA to silica gel ratios of 0.9:0.1 and 0.8:0.2 named as J0.9 and J0.8, respectively. FA and silica gel were mixed well with distilled water in a beaker at an L/S ratio of 0.5 mL/1 g. All mixtures were allowed to hydrate at ambient conditions. The hydrated mixtures were then mixed with distilled water at an

L/S ratio of 10:1 by weight, shaking at 200 rpm for 6 h. The mixtures were then filtered using a 0.45 μ m filter paper to obtain solutions for ICP-OES analysis.

2.4 Characterization study

The water content was measured by oven drying the sample at 105 °C for 24 h to a constant mass and measuring the weight. Loss on ignition (LOI) was measured after calcining the oven-dried sample at 440 °C for 2 h. The chemical and mineralogical compositions of the samples were determined using X-ray fluorescence (XRF), X-ray diffraction (XRD), and scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM-EDS). The solutions after filtering were analyzed by inductively coupled plasma-optical emission spectrometry (ICP-OES).

3. Results and discussion

3.1 Chemical compositions

The bulk chemical compositions of RH, RHA, silica gel, and FA were analyzed by XRF. The chemical compositions of RH, RHA, and silica gel are listed in Table 1. The results showed that the major component of these samples was SiO₂. Silica gel has the highest SiO₂, among others. Therefore, silica gel was selected as the material to stabilize heavy metals in FA in this study. SEM-EDS analysis was used to study the material morphology and composition of the elements. Fig. 1 exhibits the SEM images of RH, RHA, and silica gel. Fig. 1a shows the RH surface before the calcination process, displaying the protuberance and outer epidermis. Fig. 1b shows the RHA surface with increased porosity and pore volume after the calcination process of RH. Fig. 1c shows the surface of silica gel obtained from RHA with the highest pore volume and smallest size of porosity compared to RH and RHA. The EDS analysis of the silica gel surface indicates that Si and O account for 77.25 and 20.70 wt.%, respectively, with 2.05 wt.% of Na as an impurity.

In Table 1, Zn and Pb showed essentially higher concentrations compared to others in FA. According to the standard leaching test results (JLT-46), their initial leaching concentrations were the highest among other elements. Hence, these two elements were the targets of this study.

3.2 Phase identification

The broad XRD spectra in the 2θ range of 20– 30° indicate that RH, RHA, and silica gel are essentially amorphous.

Table 1 Chemical compositions of FA, RH, RHA, and silica gel

<u>C1 1</u>	MOM	DII	DILA	C'1'	
Chemical	MSWI	KH	KHA	HA Silica	
composition	fly ash			gel	
Chemical (%)					
SiO ₂	6.20	93.82	93.33	97.47	
Al ₂ O ₃	3.56	0.07	0.07	0.08	
Fe ₂ O ₃	2.55	0.15	0.19	0.06	
CaO	49.16	1.09	0.23	0.20	
MgO	3.03	0.35	1.30	0.01	
Na ₂ O	6.98	0.20	0.38	1.65	
K ₂ O	1.92	2.08	0.23	0.19	
Cl	18.35	0.58	2.70	0.05	
Water content	0.11	2.09	1.67	1.61	
LOI	1.16	66.49	36.60	6.62	
Trace element					
contents (ppm)					
Zn	15216				
Cu	1402				
Pb	2748				
Cr	264				



Fig. 1. SEM images of (a) RH, (b) RHA, and (c) silica gel derived from RHA.

3.3 Effects of silica gel on Pb and Zn stabilization

Fig. 2 shows the initial concentrations of Pb and Zn leached by different FA and silica gel ratios. The leachate concentrations of Pb and Zn rapidly decreased after 6 h and reached equilibrium after that.

The effect of silica gel on the stabilization of Pb and Zn in FA was examined at various times. The effect of silica gel was measured by metal removal efficiency in the leachate. The stabilization efficiency of the target metal was calculated using Eq. 1.

$$R_{e}(\%) = 1 - \frac{c_{f}}{c_{i}} \times 100$$
(1)

where R_e is the target metal removal efficiency (%), C_f is the metal concentration in the test group, and C_i is the initial metal concentration.

The results in Table 2 show that silica gel stabilized Pb and Zn in the FA sample despite high initial leached concentrations of both metals (64.03 mg/L and 27.62 mg/L, respectively). These outstanding results show that silica gel from RHA can stabilize heavy metals in FA.

Table 2 Removal efficiency of Pb and Zn in the leachates of MSWI fly ash in the presence of silica gel

Heavy	Sample	Removal efficiency (%)						
metal	-	6 h	1 d	2 d	3 d	7 d	14 d	
Pb	J0.9	96.32	99.72	98.09	97.58	97.00	98.73	
	J0.8	99.19	98.86	99.11	99.16	98.30	99.16	
Zn	J0.9	92.02	91.04	92.52	89.72	95.50	93.43	
	J0.8	95.33	92.31	90.05	91.85	84.09	92.30	



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Fig. 2. Leaching concentrations of Pb and Zn.

4. Conclusions

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This study found that silica gel had 97.47% of SiO2 which can be produced from RHA using a low-cost method. The silica gel can be used directly as an effective metal stabilizer in the incinerator. Furthermore, this alternative method of using RHA to produce silica gel can help minimize the agricultural waste and chelate that is commonly used in the industry. The results showed that after proper mixing of silica gel (from RHA) with MSWI FA, heavy metals such as Pb and Zn in FA could be efficiently stabilized up to 99%.

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