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Chemical properties of rare earth elements in typical medical waste incinerator ashes in China

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Abstract

Medical waste (MW) ashes from different types of MW incinerators were examined to detect the characteristics and environmental impact of rare earth elements (REEs). The results showed that total REE contents in the ash samples ranged from 10.2 to 78.9 mg/kg. REEs in bottom ash were apparently higher than those in fly ash. Average REE contents in the ashes followed the sequence of Ce > La > Nd > Y > Gd > Pr > Sm > Dy > Er > Yb > Ho > Eu > Tb > Lu > Tm. Some of the elements, such as Sm, Dy, Ho, Er, Yb in the ash samples were in normal or nearly normal distribution, but Y, La, Ce, Pr, Nd, Eu, Gd, Tb, Tm, Lu were not normally distributed, indicating some of the ash samples were enriched with these elements. Crust-normalized REE patterns indicated that two types of the MW ashes were obviously enriched with Gd and La. Sequential extraction results showed that REEs in the ash mainly presented as residual fraction, while exchangeable and carbonate fractions were relatively low. DTPA- and EDTA-extraction tests indicated that REEs in the MW ashes were generally in low bioavailability. © 2008 Elsevier B.V. All rights reserved.

Keywords: Medical waste; Incinerator ash; Rare earth element; Sequential extraction; Bioavailability

1. Introduction

Since the national-scale outbreak of Severe Acute Respiratory Syndrome (SARS) in 2003, incineration has become the most popular option for the treatment of medical waste (MW) in China [1]. Incineration shows effectiveness in reducing toxic effects of infectious materials, as well as reducing the amount and volume of the waste to be landfilled [2]. However, with the growing trend of using disposal plastic or metallic goods in medical treatment, the amount of MW increased rapidly [3]. Therefore, incinerator residues, such as bottom ash and fly ash are generated in a large volume, which have been of environmental concern, as they may cause secondary pollution to the environment [4].

Identification of the chemical composition of the ash is important to evaluate its environmental impact. Previous studies have focused on the toxic chemicals such as heavy metals [3,5], polycyclic aromatic hydrocarbons (PAHs) exist in the ash [6,7].

0304-3894/\$ - see front matter © 2008 Elsevier B.V. All rights reserved. doi:10.1016/j.jhazmat.2008.01.091 Nevertheless, little information is available on the characteristics and environmental impact of rare earth elements (REEs) contained in the ash thus far. REEs are valuable resources, while high concentrations of REEs may cause significant damage to ecosystem, thus REEs should be given enough attention [8]. There have been some studies on the toxic effects of REEs [9–11].

It is known that the largest proved REEs reserve (4.3 million tons) is located in China, contributing 43% of the world's total reserves [12]. In recent years, REEs are widely applied in medical fields such as medical reagents, laser materials, magnets manufacture, etc. Both the high levels and frequently use in medical treatment may lead to the enrichment of REEs in MW incinerator residues. In addition, Yao et al. [13] found that the concentration of REE in fly and bottom ash is several to 20 times higher than those in their raw coals, indicating REEs are concentrated in fly and bottom ash after coal combustion. Furthermore, chemical speciation of REEs may occur after combustion [12]. Therefore, it is necessary to obtain more information of REEs in this type of waste ashes so as to predict any potential risk to environment in future. In most cases, total concentration of REEs is not a good tool for potential risk assessment. Extraction procedures using a selective chemical extractant, such as strong

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chelating agents, could provide information on the bioavailability of REEs [14].

The current study aimed to investigate the chemical properties of REEs in different types of MW incinerator ashes. The potential mobility and bioavailability of the REEs were also assessed.

2. Materials and methods

2.1. Sample collection and preparation

Four types of MW incinerator ashes including twenty-three ash samples were collected from different MW incineration plants located in four cities in China. MWI-1 was a mediumscale incinerator located in north China, two mixed bottom ash samples was collected every half-month. MWI-2 was a special type of incineration facility, located in south China, which combined more than ten small fixed grate furnaces. Although air pollution control devices (APCDs) are equipped to each furnace, fly ash could hardly be collected in this type of incinerator, thus only bottom ash were obtained. Two samples were collected every half-month from this incinerator. MWI-3 was a typical small incinerator run by a hospital in a county located in middle China, which has no APCDs, thus no fly ash could be collected. Twelve mixed samples of this incinerator were collected once a week. MWI-4 was a large-scale incineration plant located middle China. This incinerator, representing more than 100 similar facilities, is equipped with APCDs. In this type of incinerator, incineration ashes are mainly collected in the bag filters as fly ash, and the residue mainly consists of unburned glasses, plastics and metallic matters, thus only fly ash samples were collected. Seven mixed samples were collected every week from this incinerator. After sampling, the samples were dried at 105 °C for 24 h and ground to <0.15 mm using an agate mortar for analysis.

2.2. Total REEs analysis

The samples were digested using the method described by Zhang et al. [15]. Specifically, 0.5 g of dry sample was weighed into a Teflon beaker, 2.5 ml HNO₃ and 2.5 ml HClO₄ added and heated for 2–3 h, after cooling, 2.5 ml HClO₄ and 5 ml HF was added and heated for 15 min, then 5 ml HF was added until the residue became dry, the residue was dissolved using 5 ml HNO₃ and diluted 1000 times [16]. REEs concentrations in the solution were determined using an Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) (Agilent 7500A, USA). All ash samples were digested and analyzed in replicate. The following elements were determined: Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu.

2.3. Sequential extraction procedure (SEP)

Chemical speciation of REEs in the samples was determined using SEP suggested by Tessier et al. [17]. The procedure classified elements into five fractions: (1) exchangeable fraction, (2) carbonate bound fraction, (3) Fe–Mn oxide bound fraction, (4) organic matter bound fraction, and (5) residual fraction. The

Table 1	
Element analysis of reference material sludge 101 (mg/kg)	

	Certified value	This study	Recovery
Y	2.5 ± 0.084	2.5 ± 0.056	100.05
La	3.9	4.1 ± 0.247	104.85
Ce	7.4 ± 0.084	7.4 ± 0.371	99.63
Pr	0.87 ± 0.005	0.87 ± 0.017	99.80
Nd	3.5 ± 0.048	3.4 ± 0.292	97.10
Sm	0.48 ± 0.017	0.50 ± 0.045	104.81
Eu	0.21 ± 0.008	0.20 ± 0.018	95.22
Gd	0.57 ± 0.014	0.58 ± 0.022	102.43
Tb	0.078 ± 0.007	0.082 ± 0.002	105.26
Dy	0.44 ± 0.014	0.44 ± 0.013	99.76
Но	0.094 ± 0.001	0.100 ± 0.002	106.65
Er	0.27 ± 0.01	0.30 ± 0.018	110.85
Tm	0.04 ± 0.003	0.03 ± 0.002	87.19
Yb	0.26 ± 0.008	0.28 ± 0.003	106.20
Lu	0.04 ± 0.002	0.036 ± 0.003	89.27

extraction method is described in more detail by Tan et al. [18]. Among the five fractions, exchangeable fraction is more mobile thus easy to leach out into the environment. In contrast, residual fraction is considered stable [5].

2.4. Bioavailability of REEs

Diethylenetriamine pentaacetic acid (DTPA) extract test was suggested by Lindsay and Norvell [19], according to the method, 10 g ash was mixed with 20 ml 0.005 mol 1^{-1} DTPA, add 0.01 mol 1^{-1} CaCl₂, pH of the suspension was adjusted to 7.3 in a 50 ml plastic centrifuge tube, and shaken for 2 h. Ethylene diamine tetraacetic acid (EDTA) extract test was conducted by Wear and Evans [20] method: 2.0 g ash sample was mixed with 20 ml 0.05 mol 1^{-1} EDTA and its pH was adjusted with ammonia solution to 7.0 in a 50 ml plastic centrifuge tube, and the suspension was shaken for 1 h. For all extraction procedures, after shaking, the suspensions were centrifuged at 3000 rpm for 20 min and the supernatant was filtered through a 0.45 µm cellulose acetate membrane filter prior to determination. All extractions including blanks were carried out in triplicate.

2.5. Quality control

The quality and precision of sample digestion and analyzing procedures were controlled using a certified standard reference material: sewage sludge no. 101 from the National Institute of Agro-Environmental Science, Ministry of Agriculture, Forestry and Fisheries (Japan). A comparison between certified values and those found in this study were illustrated in Table 1. The recoveries were between 87.19 and 110.85%.

3. Results and discussion

3.1. Total REE concentrations in the MW ash samples

Table 2 shows the REEs content in the four types of MW ashes. As can be seen, the sum of REE (\sum REE) concentrations in the ashes were around 10.15–78.86 mg/kg. Concentrations

Table 2			
REE concentrations in	the MW	ashes (m	g/kg)

	MWIA-1		MWIA-2		MWIA-3		MWIA-4	
	Mean	S.D.	Mean	S.D.	Mean	S.D.	Mean	S.D.
Y	4.39	1.36	6.11	0.92	4.72	0.94	1.28	0.66
La	10.59	3.08	18.13	4.29	8.81	2.28	1.76	1.23
Ce	17.54	4.07	28.41	4.40	15.05	3.39	2.85	2.04
Pr	2.10	0.56	3.23	0.37	1.78	0.41	0.53	0.23
Nd	7.73	2.49	11.98	2.19	6.40	1.60	1.35	1.04
Sm	1.34	0.37	2.09	0.23	1.32	0.26	0.74	0.69
Eu	0.25	0.01	0.44	0.08	0.25	0.05	0.06	0.04
Gd	3.21	0.35	5.03	4.61	1.17	0.19	0.59	0.13
Tb	0.19	0.06	0.25	0.02	0.18	0.02	0.07	0.02
Dy	0.80	0.28	1.17	0.14	0.79	0.16	0.16	0.13
Но	0.30	0.05	0.38	0.04	0.30	0.03	0.18	0.02
Er	0.72	0.17	0.93	0.11	0.74	0.10	0.39	0.07
Tm	ND	_	0.01	0.01	ND	-	ND	-
Yb	0.71	0.33	0.71	0.11	0.55	0.08	0.20	0.06
Lu	ND	_	0.02	0.01	ND	0.01	ND	_
NEE	49.85		78.86		42.04		10.15	

ND: not detected.

of \sum REEs in bottom ashes are 4–8 times of fly ash, which may attribute to the high boiling points of REE elements (1194–3426 °C). In addition, the input composition, combustion temperature may also contribute to the different concentration of REE in ashes between different incinerators.

In case of individual REEs, Ce was the most abundant of the whole REEs, with a range of 1.60-31.52 mg/kg, followed by La (1.02-21.16 mg/kg), whereas, Tm (0-0.01 mg/kg) was the least abundant. Average REE contents in MW ash followed the sequence of Ce>La>Nd>Y>Gd>Pr>Sm> Dy>Er>Yb>Ho>Eu>Tb>Lu>Tm. It is apparent that the content of total light REEs (\sum LREEs, including La, Ce, Pr, Nd, Sm, Eu) is much higher than that of heavy REEs (\sum HREEs, including Y, Gd, Sm, Dy, Er, Yb, Ho, Tb, Lu, Tm). Relatively higher variation was found for Y (S.D.: 0.66–1.36), La (S.D.: 1.23-4.29), Ce (S.D.: 2.04–4.40), Nd (S.D.: 1.04–2.49), Gd (S.D.: 0.13–4.61) compared to other elements (S.D.: below 1).

3.2. Distribution of REEs in the waste ashes

The distribution patterns of the REEs in the MW ash samples were examined using the software SPSS (version 13.0J). The results showed that Sm, Dy, Ho, Er, Yb in the ash samples were in normal or nearly normal distribution, but Y, La, Ce, Pr, Nd, Eu, Gd, Tb, Tm, Lu were not normally distributed. For example, the histograms of Y, Tb, Ce, Sm, La, \sum REE were shown in Fig. 1. As can be seen, curves for Tb and Sm were normally distributed, while that for Ce, La, Y, \sum REE were slanted either to the left or to the right.

Since REEs in natural materials approximated the normal distribution [21,22], the non-normal distribution of Y, La, Ce, Pr, Nd, Eu, Gd, Tb, Tm, Lu in the ash samples revealed that some of samples are enriched with these REEs. REE enrichment in MW ash may be caused by many activities in medical fields. In recent years, REEs are applied for medical instruments, e.g., Ho, Nd, Y are used as additives to laser materials; Ce, La and Yb are

used in special glass; Eu is used in X-ray system to diagnostic illness; Ce, La, Yb, Sm are widely used to make drugs treating coagulant, tumour [15]. The foregoing activities may contribute to the redistribution of some of the REEs in the MW ash.

3.3. Crust-normalized REE patterns for the MW ashes

Crust-normalized REE pattern could be used to evaluate the enrichment of REEs in studied waste ash. If the MW ashes are not enriched with REEs, the normalized pattern should be horizontal regardless of the concentration levels of REEs in material [23]. In the present study, the REE abundance ratios between MW ash and continental crust were calculated and plotted in Fig. 2. As can be seen, two types of MW ashes were obviously enriched with Gd (0.93 and 0.59, respectively) and La (0.60 and 0.35, respectively). Among them, Gd is widely used in medical field as an effective chelate reagent to improve NMR imaging signal, while La is used as additives to glass to make medical instruments and applied for some medical drugs. Comparatively, flat REE patterns for MWI-3 and MWI-4 were found. For MWI-3, LREE abundance ratios (0.22-0.29) were higher than HREE (0.01-0.26), indicating that this type of MW ash are abundant with LREEs.

3.4. Comparison of REEs between the MW ashes and Chinese soils

Average REE abundance in the MW waste ash was compared with those in the average Chinese soils [24] and the results were shown in Fig. 3. Ratios between all the MW ash and soils were well below 1.0, with a range of 0.01–0.93, except for Gd (1.09) in MWI-1. Thus, most of the MW ashes will cause less environment problem of REE when landfill. Nevertheless, it should be noted that MWI-1 were contaminated by Gd (1.09) and MWI-2 were slightly enriched with Gd (0.70), thus Gd may accumulate in the soils when the ash is disposed by landfill.



Fig. 1. Histograms for REEs showing their distribution in the MW ash samples.

3.5. Fractional distribution of REEs in the MW ashes

Fig. 4 illustrates that the distribution patterns of different REEs in the MW ash samples, in which Eu, Tb, Tm and Lu were excluded because the total concentrations of these elements were much lower. The REE distribution patterns of all



Fig. 2. Crust-normalized REE patterns for the MW ashes.

the examined REEs in the ashes are quite similar. In general, residual were the predominant fraction of all the REEs accounting for 47.03-87.67% (0.46–2.62 mg/kg), the organic matter fraction is the second important fraction, in a range of 1.83-31.94% (0.21–4.152 mg/kg). In contrast, the concentration of Fe–Mn oxide associated REEs were relatively low, accounting



Fig. 3. Comparison of REE contents in the MW ashes and Chinese soils.



Fig. 4. Fraction distribution of REEs in the MW ashes. F1: exchangeable fraction; F2: carbonate bound fraction; F3: Fe–Mn oxide bound fraction; F4: organic matter bound fraction and F5: residual fraction.

Table 3 DTPA- and EDTA-extractable REEs in the MW ashes (µg/kg)

for 2.73–22.01%, while the exchangeable and carbonate fraction (0–5.89% and 0.32–4.28%, respectively) were the lowest. Residual fraction is considered to be stable thus difficult to leach out into the environment, while exchangeable and carbonate fractions represent the potential availability and leaching abilities of metals [25]. Accordingly, it is believed that REEs in MW ash is relatively safe after landfill.

Among the four types of MW incinerator ashes, concentrations of exchangeable and carbonate REEs in fly ash (3.55%) are higher than those in bottom ash (2.32%, 2.38% and 2.69%, respectively), indicating that REEs in MW fly ash has more mobility than bottom ash. In case of the individual REEs, Sm, Ho, Er, Dy showed the highest preference to bind to exchangeable (1.31–5.89%) and carbonate (2.45–4.28%) fractions, suggesting these elements have more potential to release compared with other REEs.

3.6. Bioavailability of REEs in the MW ashes

Fang et al. [26] suggested that both DTPA- and EDTAextractable REEs were significantly correlated to REE concentrations in wheat roots and shoots. The extractable REE concentrations in the ash using DTPA and EDTA were shown in Table 3. Both EDTA- and DTPA-extractable REEs were very low, accounting for 0.002–0.144% and 0.012–0.108% of the total concentration, respectively. The results revealed the bioavailability of the REEs in the MW ash is extremely low, which is consistence with that of sequential extraction as mentioned before.

Generally, DTPA-extractable REEs are higher than those of EDTA except for MWI-4. This could be mainly ascribed to the different extractability of the regents. The EDTA-extractable REEs in MWI-4 were the highest compared with other ash samples since this sample has the lowest pH (3.0). Previous study reported that EDTA were able to extract more REEs in the soils with low pH because both DTPA- and EDTA-extractable REEs were significantly negatively correlated with soil pH [24]. According to this report, high concentrations of DTPA-extractable REEs in samples from MWI-4 were sup-

DIIII and	DTTY and EDTY extractione REES in the STY asies (µg/Rg)												
	Y	La	Ce	Pr	Nd	Sm	Eu	Gd	Dy	Но	Er	Yb	∑REE
MWIA-1													
DTPA	ND	18.960	25.890	3.482	13.926	2.060	0.447	5.653	1.037	0.104	0.038	0.298	71.896
EDTA	ND	1.414	0.816	0.868	1.488	1.573	ND	0.830	1.211	0.726	1.340	ND	10.266
MWIA-2													
DTPA	11.652	11.214	9.276	2.265	8.370	1.582	0.262	2.635	0.773	0.075	0.011	0.178	48.292
EDTA	ND	0.916	0.698	0.833	1.383	1.571	ND	0.737	1.161	0.673	1.287	ND	9.259
MWIA-3													
DTPA	3.136	4.018	0.220	0.761	2.368	0.553	0.125	0.152	0.230	ND	ND	ND	11.564
EDTA	ND	0.891	0.674	0.814	1.366	1.481	ND	0.855	1.161	0.658	1.276	ND	9.176
MWIA-4													
DTPA	ND	ND	0.134	0.018	ND	ND	ND	ND	ND	ND	ND	ND	0.153
EDTA	ND	1.130	1.113	0.871	1.629	2.180	ND	0.886	1.183	0.662	1.277	ND	10.931

ND: not detected.

posed, but the results were on the contrary, which should be further investigated.

4. Conclusions

The contents of REEs in the MW ashes examined in the present study were in the range of 10.15–78.86 mg/kg, lower than those in the crust. REEs in the ash samples mainly existed as residual fraction (47.03–87.67%) and organic matter fraction (1.83–31.94%), while small amounts of the REEs were in exchangeable and carbonate fractions (0–5.89% and 0.32–4.28%, respectively). Some of the REEs, such as Y, La, Ce, Pr, Nd, Eu, Gd, Tb, Tm, Lu which are frequently used in medical fields, were enriched in the MW ash samples. Generally, REEs in the bottom ash were much higher than those in the fly ash. Among the REEs, EDTA- and DTPA-extractable species accounted only for 0.002–0.144% and 0.012–0.108%, respectively, implying a relatively low bioavailability and little environmental risk of the REEs in this type of waste ashes.

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