Study on the Effect of Mineral Compounds on the Behavior of Heavy Metals During Oily Sludge Incineration

Yue Han [1,2], Zhiqiang Gong [1,2,*] and Mingxin Xu [3,*]

[1] State Grid Shandong Electric Power Research Institute, Jinan 250003, China; yueaibamadr@sina.com
[2] Shandong Smart Grid Technology Innovation Center, Jinan 250003, China
[3] National Engineering Research Center of New Energy Power Generation, North China Electric Power University, Changping District, Beijing 102206, China
* Correspondence: zhqgong@foxmail.com (Z.G.); mingxin.xu@ncepu.edu.cn (M.X.)

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Table S1. Experimental instruments and equipment.

Instrument and equipment	Specification type	Manufacturer
Horizontal tube furnace	BLMT-1200°C	Bremant Test Electric Furnace
ICP-OES	Agilent ICP OES-720	Agilent Technology Company
Element analyzer	VARIO EL III	Elementar Germany
Microblog disintegrator	MWD-850	Shanghai Yuanhua Instrument
X-ray diffractometer	X-Pert PRO MPD	Panaco (Netherlands)
Scanning electron microscope	S-4800	Hitachi Corporation of Japan
Thermogravimetric analyzer	ZH-1250	Yingnuo Precision Instrument
Constant temperature oscillator	SHZ-B	Wanning Scientific Instrument
A magnetic stirrer	DF-101	Corey Instruments Company
Centrifuge	LSC-50H	Zhengrong Experimental Instrument
Electric heating plate	DB-XAB	Lichen Bangxi Instrument
Electric drying box	DGG-9003	Boxun Industrial Company
Electronic bottom ashlance	FA1204C	Tianmei bottom ashlance Instrument

Table S2. Experimental reagent.

Experimental reagent	Specifications	Manufacturer
CaO	Analytical purity	Chemical Reagent Co., Ltd. of Guoyao Group
Fe_2O_3	Analytical purity	Chemical Reagent Co., Ltd. of Guoyao Group
Al_2O_3	Analytical purity	Chemical Reagent Co., Ltd. of Guoyao Group
MgO	Analytical purity	Chemical Reagent Co., Ltd. of Guoyao Group
$HClO_4$	Analytical purity	Tianjin Zhengcheng Chemicals
HAc	Analytical purity	Tianjin Zhengcheng Chemicals
HCl	Analytical purity	Taicang Shanghai Trial Agent
HF	Analytical purity	Kunshan Jincheng Reagent
H_2O_2	Analytical purity	Shanghai Wokai Biotechnology
HCl	Analytical purity	Shanghai Titan Chemical
HNO_3	Analytical purity	Chemical Reagent Co., Ltd. of Guoyao Group
Deionized water	Analytical purity	China University of Petroleum (East China)

Table S3. Concentration of heavy metal in different forms in incineration bottom ashes.

Samples	Heavy metal	Total	F1	F2	F3	F4	Recovery rate (%)
				mg/L			
OS	Cr	92.25	36.16	28.03	19.89	6.33	98.24
	Cu	65.64	37.31	12.22	7.08	7.72	98.24
	Zn	725.14	241.62	255.83	127.91	85.28	98.24
	Pb	128.25	35.91	37.19	39.75	15.39	98.24
	As	84.24	29.48	21.94	14.32	18.53	98.24
	Cd	24.24	8.73	5.58	6.06	3.88	98.24
OSA	Cr	173.62	27.74	22.85	60.39	52.22	96.21
	Cu	113.55	35.61	27.82	20.03	27.82	96.21
	Zn	488.11	69.56	139.11	69.56	171.57	96.21
	Pb	65.12	9.76	16.28	20.84	18.23	96.21
	As	16.43	3.94	3.12	4.76	4.61	96.21
	Cd	8.25	2.06	2.97	2.06	1.16	96.21
OSA-CaO	Cr	186.22	40.15	12.77	111.32	18.25	94.21
	Cu	145.24	16.73	25.14	51.59	46.01	94.21
	Zn	451.78	52.59	166.53	65.73	153.38	94.21
	Pb	202.14	56.59	30.32	44.47	70.74	94.21
	As	60.11	10.81	15.02	17.44	16.83	94.21
	Cd	15.37	4.92	3.38	3.38	3.69	94.21
OSA-Fe_2O_3	Cr	173.21	32.91	83.14	19.05	38.11	97.41
	Cu	134.37	30.91	26.87	37.62	38.96	97.41
	Zn	434.98	143.54	126.14	73.94	91.34	97.41
	Pb	63.32	29.12	11.39	8.86	13.93	97.41
	As	57.21	20.59	6.29	12.01	18.31	97.41
	Cd	13.73	6.59	1.51	2.88	2.75	97.41
OSA-Al_2O_3	Cr	168.37	62.29	23.57	35.35	47.14	96.12
	Cu	140.14	56.05	15.41	37.83	30.83	96.12
	Zn	496.32	24.81	104.22	173.71	193.56	96.12
	Pb	198.32	95.19	21.81	41.64	39.66	96.12
	As	75.37	11.30	8.29	26.37	29.39	96.12

	Cd	11.12	5.67	1.33	2.22	1.89	96.12
	Cr	170.26	34.05	18.72	56.18	61.29	94.51
	Cu	115.21	71.43	17.28	12.67	13.82	94.51
OSA-MgO	Zn	505.31	106.11	181.91	106.11	111.16	94.51
	Pb	64.32	30.87	9.64	12.86	10.91	94.51
	As	16.21	6.97	2.26	3.44	3.56	94.51
	Cd	12.37	7.05	1.73	1.86	1.73	94.51

Step S1. The total amount of heavy metal experimental steps.

The amounts of heavy metals in the samples were tested by microwave digestion. First, a 0.050g sample was added to the HNO_3-HF-H_2O_2 (8 mL; 2 mL; 1 mL) digestion system and digested using a microwave digestion apparatus. In order to reduce the influence of the acid on the instrument and the measurement accuracy, the acid absorption device was set at 120°C to remove the residual acid. Finally, we collected and stored the clear solution in a volume of 100 mL and measured the concentration of heavy metals by ICP-OES. In order to ensure the authenticity of the results, all experiments were set up with two parallel samples and one blank.

Step S2. Toxicity Characteristic Leaching Procedure (TCLP) experimental steps.

As the latest legal heavy metal pollution assessment method in the United States, the TCLP is the most widely used ecological risk assessment method in the world. It is mainly used to detect the dissolution and mobility of heavy metals in solid waste.

First, the incinerated bottom ash was ground and sieved with a 20-mesh sieve to prepare a sample with a particle diameter of less than 0.95 mm. Then, a 5.000 g sample was weighed and transferred to a beaker, mixing with 96.5 mL of deionized water and covering the watch glass, vigorously stirring for 5 min with a magnetic stirrer, and extracting the supernatant into a beaker of 50 mL to test the pH. If pH < 5.00, leaching agent A (CH_3COOH-NaOH-HNO_3 with pH = 4.93) was used, and, if pH > 5.00, 3.5 mL of a 1 mol/L HAc solution was added to the beaker and heated to 50°C in a water bath, preserving the temperature for 10 min and then cooled to room temperature, taking the supernatant to test the pH. If the pH was still less than 5, then the leaching agent A was selected; otherwise, the leaching agent B (CH_3COOH with pH=2.88) was selected. A total of 100 mL of the selected leaching agent was put in the beaker and shaken at room temperature for 18 hours with oscillation at 30±2 r/min. The filter was drained with a 0.6-0.8 μm filter without standing, and, then, the ICP-OES was used to test the clarified filtrate.

Step S3. BCR experimental steps.

The Bureau Community of Reference (BCR) proposed by the European Community Bureau of Reference can effectively distinguish the activity of heavy metals in the samples from their toxicity to the environment. The speciation fractions of heavy metals include the exchangeable and acid-soluble fraction（F1）, reducible fraction（F2）, oxidizable fraction (F3), and residual fraction (F4).

a. Exchangeable and acid-soluble fraction

The 1.000 g sample was mixed with 40 mL of HAc extract (0.11 mol/L) in a 50 mL polypropylene centrifuge tube and shaken at room temperature for 16 h with an oscillation speed of 250 r/min. Then, it was centrifuged for 20 min with a centrifuge speed at 4000 r/min, and the supernatant was decanted and tested with ICP-OES. A total of 20 mL of deionized water was added to the centrifuge tube, and the cleaning solution was shaken, centrifuged, and discarded to retain the solid residue.

b. Reducible fraction

Add a 40 mL NH_2OH/HCl extract (0.5 mol/L) to the residue left after step 1, and repeat the rest operation as the previous step.

c. Oxidizable fraction

A total concentration of 10 mL of H_2O_2 was slowly added to the residue left after the step 2 and digested at room temperature for 1 h. The mixture was shaken intermittently for the first half hour then heated to 85°C in a water bottom ash and digested for 1 h. Then, the lid was removed, and the solution was heated until it reached near dryness. Another 10 mL of H_2O_2 was added, and the above digestion and temperature evaporation process were repeated, until 1-2 mL of turbid liquid was obtained. Add 50 mL of NH_4OAc (1 mol/L) extract, and repeat the rest of the procedure as the previous step.

d. Residual fraction

The residue extracted from the step 3 was transferred to a Teflon beaker then mixed with 10 mL of HNO_3, 10 mL of HF and 3 mL of $HClO_4$, covered and heated at low temperature for 1 h and then medium temperature for 1 h on a hot plate. After that, open

the cover to remove silicon. Continuously heat until white smoke generated and capped to break down black organic carbides. After the black organic matter on the wall disappeared, the lid was opened to drive off the $HClO_4$ white smoke and evaporated until the contents become viscous. Add another 3 mL of HNO_3, 3 mL of HF, and 1 mL of $HClO_4$ depending on the digestion, and repeat the above digestion process. When the white smoke again disappeared and the contents of the crucible were viscous, cool down the crucible, rinse the lid and inner wall with water, and, then, add 1 mL of V (HNO_3) / V (H_2O) = 1 solution to dissolve the residue at low temperature. After the digestion solution was cooled, it was transferred to a 25 mL volumetric flask and tested with ICP-OES after constant volume.