

Leaching Behavior of Heavy Metals in Coal Gasification Residuals

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Abstract

The system of dynamic leaching was designed for samples of a coal gasification residual from a coal-to-methanol plant in northwestern China in order to investigate the leaching behavior of four potentially hazardous elements – Hg, Cr, As, and Pb – in the coal gasification residual and investigate the impacts of the leaching time, size of the samples, and pH of the leaching solution on the leaching behavior of elements. The concentrations of elements in the leachate samples were all determined by inductively coupled plasma-mass atomic spectrometry (ICP-MS). The results showed that the leaching time, size of the samples, and pH of the leaching solution played an important role in affecting the leaching behavior of the selected elements from the coal residual samples. All trends were in decreasing order of significance as the leaching time went on and most leached concentrations reached the maximum at the initial stage of leaching experiments (from hours 0 to 4) accounting for 94.44%. The 80-mesh sample had the different leaching curve from the 10-mesh and 40-mesh samples for the same element because different forms of the heavy metals existed on the different-size samples.

Keywords: leaching behavior, influential factors, coal gasification residual, dynamic leaching system, heavy metal

Introduction

Coal is China's main source of energy, accounting for 47% of the world's total annual coal consumption [1]. And the coal industry will remain a significant part of manufacturing in the next decades, which has shown us we should focus more on rational exploitation and comprehensive use of coal resources. With the rapid development of the Chinese economy and the coal industry, an increasing number of industrial solid wastes

including fly ash, cinder, and coal gangue could lead to a series of environmental issues and human health problems during coal mining and processing [2-3].

Coal industry residue consists of fly ash and coal gasification residual. The disposal technology of fly ash has been quite mature. But there is no disposal method for coal gasification residue because of its low calorific value, large amount, large particle diameter, and high contents of heavy metals and PAHs [4-7]. At present, the main way of dealing with coal gasification residual is still, open dumping, which not only damages land resources but also destroys area scenery, even contaminating surrounding areas. Leaching has proven to be the most

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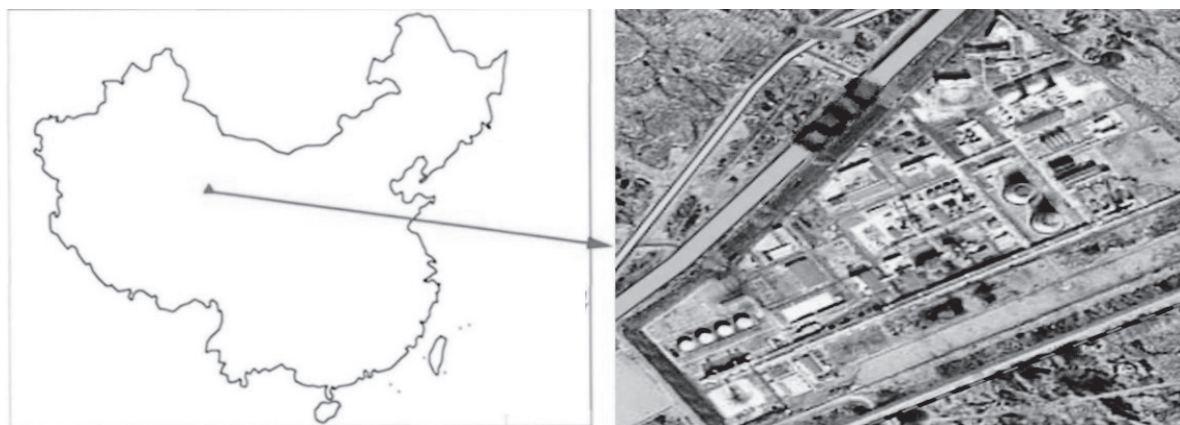


Fig. 1. Geographical location of a coal-to-methanol plant in northwestern China.

important of the primary pathways for toxic substances entering soil and water (even groundwater). Environmental pollution problems and the potential risks for human health are caused as a result of the release and migration of harmful elements contained in coal gasification residual. According to Liu Yang et al. [8], the leaching time and sample mass have a major impact on leaching behavior, and leaching has a high potential risk to groundwater.

Combining the research situation over the world, very little research about the comprehensive utilization of coal gasification residual has been done. Meanwhile, many studies on the disposal methods of fly ash and coal gangue, which may be easier to handle, have been carried out by other authors. These applications include concrete and cement products, structural fill, cover materials [9-11], and adsorption materials for removing pigment, organic compounds, and heavy metals [12-17]. However, studies focused on the coal gasification residual are relatively rare – especially on the leaching behavior and environmental hazards. Therefore, this study was conducted to investigate the leaching behavior of four heavy metals from coal gasification residual. And the impact factors – including leaching time, pH of the leaching solution, and the size of the coal gasification residual sample – were simultaneously analyzed. The research can provide an important scientific basis for site assessment, monitoring, and remediation of the polluted area by the coal industry.

Materials and Methods

Sample Collection

Coal gasification residual samples were taken in a coal-to-methanol plant in northwestern China

(N38°34'41.9", E109°55'50.4") (Fig. 1). The plant is located in the northernmost part of Shanxi Province, which includes the juncture of the Loess Plateau and Maowusu Desert. The study area has a semi-arid continental monsoon climate that features being rich in light energy, where the rainy season meets the hot season, and large temperature differences. As a result of being landlocked, the annual average temperature is 7.9-11.3°C, and this is the area of lightest rainfall in Shanxi, with mean annual rainfall of approximately 316-513 mm.

The great potential economic value of mineral resources reached 43 trillion yuan, accounting for 95 percent of Shanxi. The area is one of the major bases of the coal industry in China, as 54% of the land contains coal, accounting for as much as one-fifth of the total. There are three gasifiers (two operating and one spare) in the gasification workshop of the methanol plant. The amount of coal consumed by gasifiers per day is 3,000 tons with 900 tons of coal residual (include fly ash of 150 tons and coarse residual of 750 tons).

The samples were collected from the coal-to-methanol plant and were immediately stored in individual specially sealed plastic bags to avoid any oxidation and contamination. The appearance of the coal gasification residual samples was similar to the fly ash. Then the chemical compositions of the coal gasification residual samples were analyzed by x-ray fluorescence spectrometry (XRF) (Table 1). A partial proximate analysis and mineralogy analysis of the samples were performed following the Chinese standard method GB212-2001 and powder x-ray diffraction (XRD), respectively (Table 2). The concentrations of elements in the leachate samples were determined by inductively coupled plasma-mass atomic spectrometry (ICP-MS) for Hg, Cr, As, and Pb, following the method described by Dai et al. (Table 3) [1].

Table 1. XRF analysis of coal gasification residual samples.

	CaO	SiO ₂	Fe ₂ O ₃	C	Al ₂ O ₃	Na ₂ O	MgO
Chemical Compositions (%)	30.08	20.12	18.17	16.12	8.23	2.14	1.93

Table 2. Proximate analysis of fly ash and gasification residuals.

Proximate analyses	M _t (%)	M _{ad} (%)	V _{ad} (%)	A _{ad} (%)	FC _{ad} (%)	Q _{net,ar} (kcal/kg)	Q _{gr,ad} (kcal/kg)
Fly ash	64.65	4.94	3.78	44.27	47.01	970	2,465
Gasification residual	28.94	0.42	2.18	81.27	16.12	549	757

M_t, total moisture; M_{ad}, moisture content, air dry basis; V_{ad}, volatile matter, air dry basis; A_{ad}, ash yield, air dry basis; FC_{ad}, fixed carbon, air dry basis; Q_{net,ar}, net calorific value, received basis; Q_{gr,ad}, calorific value, air dry basis

Table 3. Heavy metals of coal gasification residuals.

Trace element	Hg	Cr	As	Pb
Average Concentration (mg/kg)	3.797	920.821	84.237	7.436
World Coal Gangue [20]	0.2-0.6	50-160	10-80	10-40

Leaching Experiments

To investigate the influences of different pH values and sizes of coal gasification residual samples on the leaching behavior of the four heavy metals in the residual samples, we designed column leaching experimental groups for this study. The average pH of the rainfall in the study region is with no acid rain. According to the characteristics of the rainfall, distilled water (pH = 7.0), acidic solution (pH = 6.5), and alkaline solution (pH = 7.5) were prepared as leaching media. And through screening and grinding, the coal gasification residual sample could be divided into three categories: 10 mesh, 40 mesh, and 80 mesh. Each coal gasification residual sample was transferred into a plastics column that was 40 mm internal diameter and 350 mm in length (Fig. 2). The flumes (with a capacity of two liters) were installed over the columns. There are six glass rotor flowmeters (LZB-2WB) between the infusion bottle and column to control the flow from 0.4 mL/min to 4 mL/min.

In nature, leaching behavior of heavy metals in the soil is a long process. Hassett [18] thought that the experimental results of element concentration dynamics during leaching need to conduct a long-term leaching experiment. But it is difficult to simulate the leaching process in the laboratory under natural conditions for a long time. These experiments were designed to last for 96 h because the leaching reaction processes mainly happened in the initial leaching phase [19].

After screening, the samples of coal gasification residual were transferred into a fixed glass column 250 mm high. A small amount of absorbent cotton and much quartz sand to prevent fine particle loss was packed into the bottom of the column during the leaching. According to the rainfall of the study area, the outflow velocity of leaching solutions was controlled at 0.5 mL/min by the tachometers. The resulting leachates were collected after 4 h, 8 h, 16 h, 24 h, 36 h, 48 h, 72 h, and 96 h, collected 10 mL in a sealed container, and refrigerated at 4°C for analysis. The concentrations of elements in the resulting leachates were determined by

inductively coupled plasma-mass atomic spectrometry (ICP-MS) for Hg, Cr, As, and Pb.

Results and Discussion

Chemistry and Mineralogy in the Residual Samples

The coal gasification residual samples were analyzed by x-ray fluorescence spectrometry (XRF) (Table 1). The samples were dominated by CaO (30.08%) followed by SiO₂ (20.12%), Fe₂O₃ (18.17%), and unburned carbon (16.12%), along with trace percentages of Al₂O₃ (8.23%), Na₂O (2.14%), and MgO (1.93%).

The XRD patterns showed that the main minerals present in the coal gasification residual were calcite and quartz, accounting for 72% and 21%, respectively. The coal gasification residuals had low calorific value (757 kcal/kg) and the calorific value of gasification residual was much lower than that of fly ash (2,465 kcal/kg) (Table 2). Because of low calorific value, there is no utilization and disposal methods for coal gasification residual.

The concentration of element Pb (the concentration equaled 7.436 mg/kg in the study area) was close to world Clarke Values and world Coal Gangue (Table 3). The remains of the selected trace elements in this study were enriched in comparison to the Clarke Values and Coal Gangue.

Leaching Time

Time is one of the most important factors affecting the leaching behavior of heavy metals from residuals [21]. Under different conditions, four different trends could be identified from Table 4, and all trends were in decreasing order of significance. The major trend was a sharp drop followed by decreased gradually or a steady curve as the leaching time went on. This trend was observed for elements Cr, As, and Pb, accounting for 52.78% of all trends. Element Hg exhibited a slight and steady

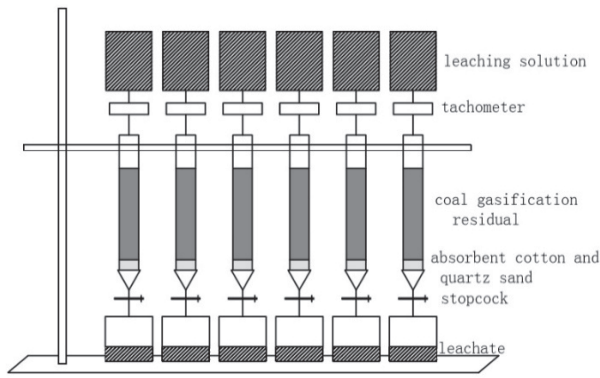


Fig. 2. System of dynamic leaching.

decrease curve that contributed to 22.22%. The slow-sharp-slow drop and sharp-slow-sharp drop trend accounted for 16.67% and 8.33% of the trends, respectively.

The concentrations of the selected elements in the leachates reached the maximum at the initial leaching time periods [22]. Most leached concentrations reached the maximum at 0-4 h, accounting for 94.44% of the leached concentrations. In the other periods of 4-8 h and 8-16 h, the resulting leachates reached the maximum concentration only twice: for Hg (40 mesh, pH = 7.5) and As (80 mesh, pH = 7.5). The possible explanations of a higher extractable concentration at initial stages were as follows. The partial fraction of elements absorbed on

Table 4. Trace element concentrations in leachate (µg/L).

Trace elements	Leaching time/h	pH = 6.5			pH = 7.0			pH = 7.5		
		Sample size			Sample size			Sample size		
		10 mesh	40 mesh	80 mesh	10 mesh	40 mesh	80 mesh	10 mesh	40 mesh	80 mesh
Hg	4	1.448	1.539	2.225	1.471	1.523	1.975	1.583	1.45	1.753
	8	1.433	1.487	1.824	1.437	1.477	1.794	1.551	1.453	1.701
	16	1.432	1.484	1.756	1.436	1.428	1.69	1.526	1.447	1.694
	24	1.423	1.463	1.678	1.439	1.444	1.64	1.521	1.418	1.648
	36	1.42	1.443	1.654	1.435	1.446	1.632	1.522	1.417	1.623
	48	1.42	1.415	1.54	1.427	1.445	1.623	1.472	1.411	1.605
	72	1.409	1.398	1.42	1.427	1.442	1.57	1.467	1.409	1.594
	96	1.407	1.378	1.41	1.42	1.432	1.572	1.461	1.407	1.59
Cr	4	10.67	12.4	12.08	11.42	13.35	13.54	10.98	13.47	15.75
	8	9.869	11.01	11.97	10.94	12	13.15	9.95	10.02	15.75
	16	9.591	10.7	11.61	10.24	11.6	12.12	9.999	9.863	15.6
	24	9.58	10.25	10.16	10.29	11.08	10.45	9.862	9.619	15.26
	36	9.674	10.33	10.11	9.533	10.64	10.65	9.787	9.656	14.4
	48	9.699	10.41	10.28	9.603	10.18	10.71	9.577	9.7	13.71
	72	9.607	10.05	10.03	9.791	10.12	10.09	9.587	9.63	12.36
	96	9.64	10.07	10.06	9.704	9.885	10.02	9.55	9.619	10.08
As	4	4.41	5.665	3.009	3.291	5.342	3.876	6.88	4.992	4.695
	8	3.307	3.022	2.935	2.465	2.787	3.807	5.916	2.345	4.735
	16	2.76	2.853	2.7	2.657	2.337	3.711	5.688	1.997	4.776
	24	2.624	2.366	2.618	2.632	2.39	3.264	5.67	1.952	2.326
	36	2.488	2.247	2.24	2.539	2.297	3.059	5.348	1.875	2.28
	48	2.369	2.137	1.885	2.48	2.218	2.733	2.956	2.021	2.683
	72	1.614	1.948	1.701	2.394	1.955	2.916	2.572	2.074	2.193
	96	1.069	1.945	1.611	2.158	1.688	2.936	2.599	1.9	2.441
Pb	4	20.75	8.414	2.174	43.48	23.4	9.979	7.44	8.381	10.08
	8	14.08	2.859	2.151	12.49	8.083	7.707	6.996	7.42	10
	16	9.317	2.298	2.093	12.26	4.98	6.014	6.536	5.634	8.97
	24	9.703	1.44	2.084	9.616	4.938	4.362	6.123	3.027	7.332
	36	9.35	1.803	1.651	7.675	4.382	3.616	5.803	3.116	7.344
	48	9.365	1.897	1.695	6.64	3.253	1.648	3.551	3.299	7.191
	72	3.398	1.759	1.694	5.375	2.462	0.848	3.08	3.755	2.08
	96	3.539	1.339	1.678	3.966	0.504	0.532	2.265	3.72	1.19

the surfaces or in the form of water-solution entered the solution easily. Then, as the leaching time went on, the concentrations of these elements on the surface and in the form of water-solution decreased gradually [21].

Size of Coal Gasification Residual

For element Hg, a slight and steady descending trend was a major curve type, except that the 80-mesh sample showed a sharp drop followed by a steady curve. And a sharp drop followed by a steady curve were observed for elements Cr, As, and Pb, except the all 80-mesh and some 10-mesh sample showed some other curve types (Table 4). Based on these trends, the 80-mesh sample had a different curve from the 10- and 40-mesh samples for the same element. The size of coal gasification residual was one of the most important factors affecting the leaching of heavy metals from residual samples. The reason this phenomenon happened was possibly that the different forms of the heavy metals existed on the different-sized samples, and the 80-mesh sample particles may have a specific surface.

The relationship between the rate of change of the leached concentrations and the sample size of coal gasification residual is shown in Fig. 3. For elements Hg and Cr, the rate of change of concentrations were found to be reversely dependent on sample size, i.e., their change increased with the decreasing of the sample size as a whole. This was mainly because the mode of occurrence of elements Hg and Cr in the 80-mesh residual sample were easily dissolved into leaching solution.

When pH = 6.5 and 7.5, the rate of change of concentrations of elements As were found to have their change decreased with the decreasing of the sample size as a whole. And when pH was 6.5, 7.0, and 7.5, the rate of change of the 80-mesh sample reached the minimum because the different mode of elements As occurred on small particles. For element Pb, the rate of change could be kept to a higher level, reaching 50% (except the 80-mesh sample at pH = 6.5).

The different form of the heavy metals existed on the different-sized samples [23-26]. And the element Pb was dissolved easily to reach a lower concentration in a short time. When pH = 7.0, the rate of change of the 40-mesh residual sample could reach 94.85%.

pH of the Leaching Solution

Previous studies have shown that the leaching behaviors of these elements are sensitive to pH of the leaching solution [27-29]. Considering the conditions of natural rainfall, three pH values – 6.5, 7.0, and 7.5 – were examined in this study. OriginPro software was used to calculate and draw the curves of the concentrations of the selected elements in the leachates to the leaching time. Then the areas under the curves were calculated as the leachability amount of the selected elements. The changes of leachability amount of the selected elements after 96 h are shown in Fig. 4.

For the 80-mesh coal gasification residual, all selected elements showed minimum leachability at pH = 6.5, although between pH = 7.0 and 7.5 it did not follow any obvious regular patterns. A possible explanation was that

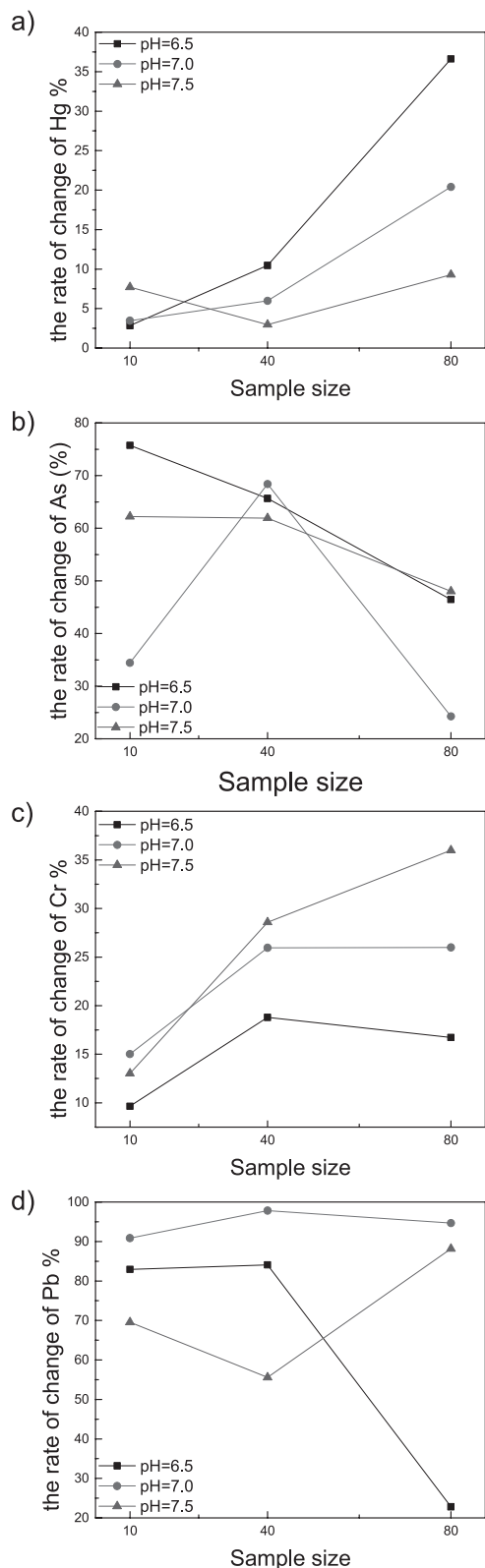


Fig. 3. Rate of change (%) of the leached concentrations with the sample size.

the form of heavy metals could not be easily dissolved in acid solution.

For the 40-mesh coal gasification residual, the leachability amount of elements Hg and Cr showed a

maximum leachability at pH = 7.0. The leachability of As decreased with increasing pH. Pb showed a minimum leachability at pH = 6.5, and there was no significant change of leachability amount at pH = 7.0 and 7.5.

For the 10-mesh coal gasification residual, the leachabilities of Hg and As increased with the increasing pH and attached to a maximum leachability at pH = 7.5. The minimum leachability of element Pb was shown at pH = 7.5. And the leachability of Cr had no significant changes under different pH values.

In general, there was no significant variation rules for the pH values and the leachabilities of the elements from the coal gasification residual. But considerable changes of the amount of leachabilities were experienced with the change of pH of leaching solution. The results indicated that the pH values could influence leaching behavior.

Conclusions

- As the leaching time went on, all trends were in decreasing order of significance. Of them, the major trend was a sharp drop followed by decreased gradually or a steady curve accounting for 52.78% of all trends. And most leached concentrations reached the maximum at the initial stage of leaching experiments (from hours 0 to 4).
- The size of the residual sample was one of the impact factors for the leaching behavior of the elements. The different form of the heavy metals existed on the different-size sample. When pH = 7.0, the rate of change of the 40-mesh residual sample could reach 94.85%.
- In general, there were no significant variation rules for the pH values and the leachabilities of the elements from the coal gasification residual, but pH played an important role in leaching behavior.

Acknowledgements

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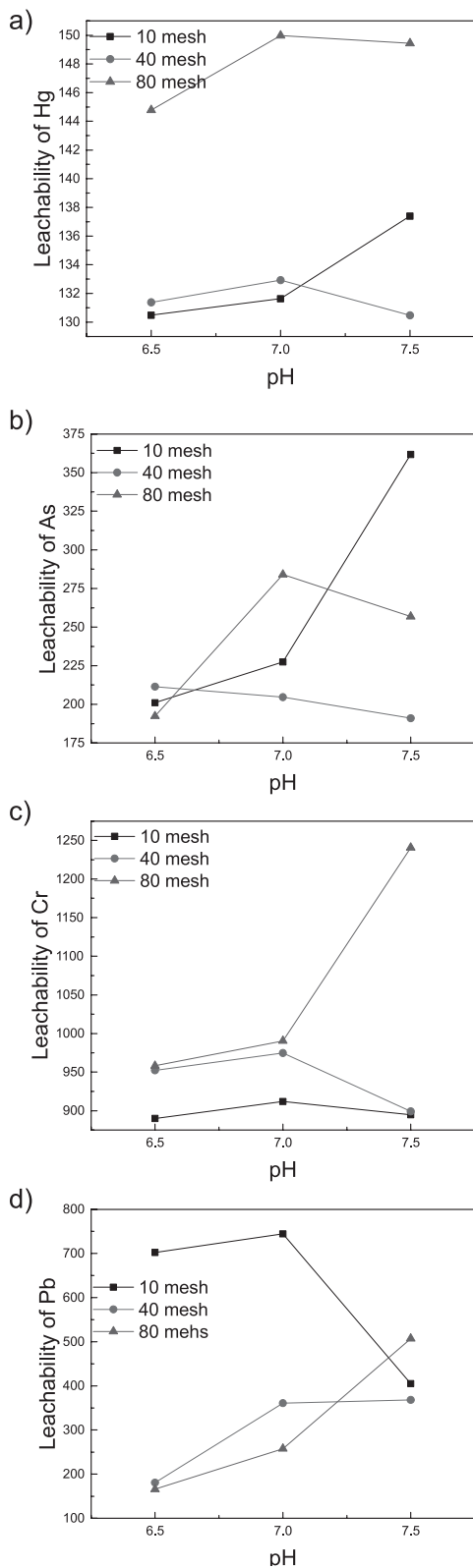


Fig. 4. Leachabilities amount of the four elements from coal gasification residual with pH after 96 h.

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