

HIGH CONCENTRATION OF ARSENIC REMOVAL FROM ACID LEACHING SOLUTION OF ZINC OXIDE DUST BY WATER-QUENCHED SLAG

The present work provides a study on high concentration of arsenic removal from acid leaching solution of zinc oxide dust by water-quenched slag. The water-quenched slag is a waste slag produced from fuming furnace of lead pyrometallurgical process and used as a substitute of ferrous sulfate heptahydrate to precipitate arsenic at purification section. The effects of reaction temperature, reaction time, the addition of H_2O_2 and the addition of water-quenched slag on arsenic removal rate were systematically investigated. The reaction temperature of $70^\circ C$, reaction time of 1 h, H_2O_2 addition of 10.8 mL/L and water-quenched slag addition of 17.8 g/L are identified as the best technical parameters. At the optimum conditions, the arsenic (III) with high concentration (As 4.13 g/L) is efficiently removed (arsenic removal rate > 99%). The filtrate (Fe 1.21 mg/L, As 1.53 mg/L) with low concentrations of arsenic and iron and the stable filter residue are also obtained successfully. The United States EPA Toxicity Characteristic Leaching Procedure (TCLP) test shows that the As leachability never exceeds the regulatory limit of 5 mg/L As. The results suggest that the water-quenched slag shows potential for removing high concentration of arsenic from acid leaching solution of zinc oxide dust.

Keywords: Arsenic removal, Water-quenched slag, Zinc oxide dust, Purification

1. Introduction

Arsenic is toxic to both animals and plants owing to its affinity for lipids, proteins and other cellular components. It is relatively abundant in the earth's crust, especially in minerals. Therefore, some arsenic contained in these minerals inevitably enters into metallurgical process. In lead pyrometallurgical process, such as roasting, smelting and converting, **most arsenic** volatilize as As_2O_3 and transform into $Pb_3(AsO_3)_2$ or $Zn_3(AsO_3)_2$ finally [1], which are collected as zinc oxide dust in cloth screen or electrostatic precipitator. The zinc oxide dust is a hazardous waste and **is harmful to** environment due to the high arsenic content (2 ~ 20%). But it contains lots of valuable components such as Zn, Pb, Cd, Ag and In. So the valuable metals must be recovered from zinc oxide dust by acid leaching and arsenic precipitation. And the arsenic must be purified and separated from acid leaching solution [2].

In industrial process, the arsenic removal technology from acid leaching solution of zinc oxide dust is chemical precipitation. Firstly, the ferrous sulfate heptahydrate (the molar ratio of Fe:As > 2) and hydrogen peroxide are added into acid leaching solution. The Fe^{2+} and As^{3+} are oxidized to Fe^{3+} and As^{5+} by hydrogen peroxide respectively. Secondly, the neutralization reagent lime or limestone [3] is added to control pH = 5.0 ~ 5.2 (iron (III) and arsenic precipitation, and satisfy industrial solu-

tion standard: pH = 5.0 ~ 5.2, As < 2 mg/L, Fe < 20 mg/L). At last, the precipitates of ferric arsenate and ferric hydroxide are obtained and the arsenic is removed from acid leaching solution (As < 2 mg/L). In the above mentioned traditional method, it presents two problems as follows: Firstly, lots of ferrous sulfate heptahydrate are expended owing to high concentration of arsenic (3 ~ 20 g/L) in acid leaching solution of zinc oxide dust. Secondly, many neutralization reagents are expended because it will release acid in the process of arsenic and iron precipitation.

Therefore, to solve these problems, a new method about high concentration of arsenic removal from acid leaching solution using smelting slag and oxidant is proposed. In our preliminary studies [4,5], the high concentration of arsenic could be removed from acid solution by water-quenched slag. But the operation technology and characteristic of filter residue were not investigated systematically.

In this paper, high concentration of arsenic removal from acid leaching solution of zinc oxide dust by water-quenched slag was investigated comprehensively. In the process, the water-quenched slag which is a waste slag produced from fuming furnace of lead pyrometallurgical process is used as substitute of ferrous sulfate heptahydrate. So the worthless water-quenched slag is comprehensively utilized and the cost will be reduced.

At present, arsenic remediation is highly topical, especially high concentration of arsenic. This work can be cited in a wide

* COLLEGE OF SCIENCE, HONGHE UNIVERSITY, MENGZI 661199, YUNNAN, CHINA; LOCAL CHARACTERISTIC RESOURCE UTILIZATION AND NEW MATERIALS KEY LABORATORY OF UNIVERSITIES IN YUNNAN, HONGHE UNIVERSITY, MENGZI 661199, YUNNAN, CHINA

** FACULTY OF METALLURGICAL AND ENERGERY ENGINEERING, KUNMING UNIVERSITY OF SCIENCE AND TECHNOLOGY, KUNMING 650093, YUNNAN, CHINA

Corresponding authors: senrenwei@163.com; alios@126.com

range of metallurgical and environmental engineering journals or industry. This novel arsenic removal method has not been reported at home and abroad. Some researchers [6-8] have reported the sorption of Pb^{2+} [9], Zn^{2+} , Cd^{2+} [10], Cu^{2+} [11], P [12] and Cr^{3+} for wastewater with the blast furnace slag or water-cooled slag. Grubb [13] and Zhang [14] have researched the arsenic (As^{3+} , As^{5+}) removal from waste solution by steel slag or oxide-loaded slag. The removal mechanisms are all attributing to the adsorption properties of slag. Unfortunately, the adsorption capacity of slag is limited. It is just suitable for treating with wastewater containing low concentration of impurities such as arsenic (lower than 100 ~ 200 mg/L), and the addition of slag is very large (approximately 30 ~ 50g/L). For the high concentration of arsenic solution or wastewater (such as 4 g/L), the addition of slag is approximately 600 ~ 1000 g/L (calculated proportionately) and no researcher can assure that the arsenic will be removed efficiently.

The main purpose of this paper is to investigate the high concentration of arsenic removing from acid leaching solution of zinc oxide dust using water-quenched slag with different operation technology. The effects of reaction temperature, reaction time, additions of oxidant hydrogen peroxide and water-quenched slag on arsenic removal rate and the characteristic of filter residue were investigated.

2. Experimental

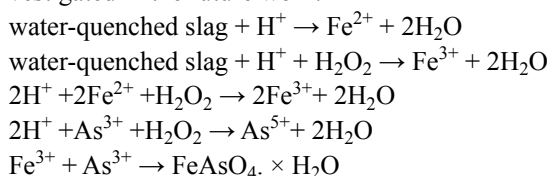
2.1. Materials

An industrial acid leaching solution of zinc oxide dust containing 4.13 g/L As (mainly As^{3+} ; and $\text{Fe} = 1.39$ mg/L; $\text{Zn} = 121$ g/L; $\text{Cu} = 0.04$ mg/L; $\text{Ni} = 1.46$ mg/L; $\text{pH} = 2.1$) was used as the feed solution. The water-quenched slag was milled to 120 mesh by a planetary ball mill (QM-1SP4). And the hydrogen peroxide (industrial grade, 27.5 wt.%) was used as oxidant.

2.2. Experimental procedure

A 2 L beaker was used as the reaction vessel which was indirectly heated by sealing electric furnace (1.5 kW). The solution was stirred with 65 mm in diameter turbine blades driven by a variable-speed motor (0 ~ 3000 rpm) (DJ1C, 100W). And the pH value was measured with pH-meter (pHS-25).

The possible chemical reactions are listed as follows for calculating the ratio of $\text{H}_2\text{O}_2/(2\text{Fe}+\text{As})$ and Fe/As (in this paper, for calculating the ratio expediently, we presume that all the iron in water-quenched slag was reacted into solution). The exact chemical reactions and complicated reaction mechanism will be investigated in the future work.



1 L acid leaching solution of zinc oxide dust containing arsenic (III) was charged into the beaker and added the 1/3 pre-calculation amount ($0.5 \sim 2 \times \text{H}_2\text{O}_2/(2\text{Fe} + \text{As})$ molar ratio) of hydrogen peroxide to oxidize the arsenic (III). The solution was heated to different temperatures (25 ~ 90°C). Then the pre-calculation amount ($0.8 \sim 2.5 \times \text{Fe}/\text{As}$ molar ratio) of water-quenched slag was added into the solution. Following the reaction of acid and water-quenched slag, another 2/3 calculation amount of hydrogen peroxide was added to oxidize the arsenic (III) and iron (II). After some reaction time, a little neutralizer limestone was added to neutralize the remaining acid and control terminal pH 5.0 ~ 5.2 (remaining iron (III) and arsenic precipitation, and satisfy industrial solution standard: $\text{pH} = 5.0 \sim 5.2$, $\text{As} < 2$ mg/L, $\text{Fe} < 20$ mg/L). The total reaction time was performed in the range of 0.5 ~ 3 h. At last, the slurry was vacuum filtered to separate the precipitate, and the filter residue was washed by water to recover zinc. Then it was vacuum filtered and dried in drying cabinet. The filter residue was pulverized and analyzed chemical compositions and phase characteristics. And the filtrate was analyzed for elements content.

It has become customary to determine the stability of compounds by subjecting them to leachate tests, usually the TCLP test [3], which involves mixing the solid with a measured volume of buffered acetic acid solution (pH 5) and determining the dissolution of elements into the aqueous solution after 20 h. So the final drying filter residue was subjected to TCLP test. The concentration of arsenic in the filtrate was determined by ICP-AES.

2.3. Analysis methods

Phase compositions of water-quenched slag and filter residue were observed with X-ray diffraction (XRD, D/MAX-3D) using $\text{Cu K}\alpha$ radiation in the range of 10° - 90° (2θ) with a step of $5^\circ \cdot \text{min}^{-1}$. The morphology of filter residue was characterized by scanning electron microscopy (SEM, Philips-XL30ESEM). The water-quenched slag, filter residue and filtrate were analyzed by X-ray fluorescence (XRF, ZSX100e), inductively coupled plasma-atomic emission spectrophotometry (ICP-AES, ICPS-1000II) and atomic absorption spectrometry (AAS-990) for As, Fe, and, if necessary, for other elements.

3. Results and discussion

3.1. Characteristic of the water-quenched slag

The element contents of water-quenched slag are shown in Table 1. The contents of Fe, Si, Al, Mg and Ca are 28.1%, 10.6%, 1.62%, 1.28% and 7.46%, respectively. According to the lead smelting process and other related researches [15,16], the major constituents of water-quenched slag are $2\text{FeO} \cdot \text{SiO}_2$, $2\text{CaO} \cdot \text{SiO}_2$, $2\text{MgO} \cdot \text{SiO}_2$ and $3\text{CaO} \cdot \text{Al}_2\text{O}_3$.

The XRD patterns (Fig. 1) of water-quenched slag show that it is a non-crystalline and vitreous material with amorphous

TABLE 1

The element contents of water-quenched slag

Element	Fe	Si	Ca	Mg	Al	As	Zn	Pb	Cu	Bi	Ag
Content (wt %)	28.1	10.6	7.46	1.28	1.62	0.27	5.82	0.45	0.05	0.006	0.0012

phase. Meantime, traces of fayalite phase (JCPDS# 34-0178) and calcium aluminum hydrate phase (JCPDS# 02-0083) are also observed, indicating that traces of crystalline phase exist in the water-quenched slag.

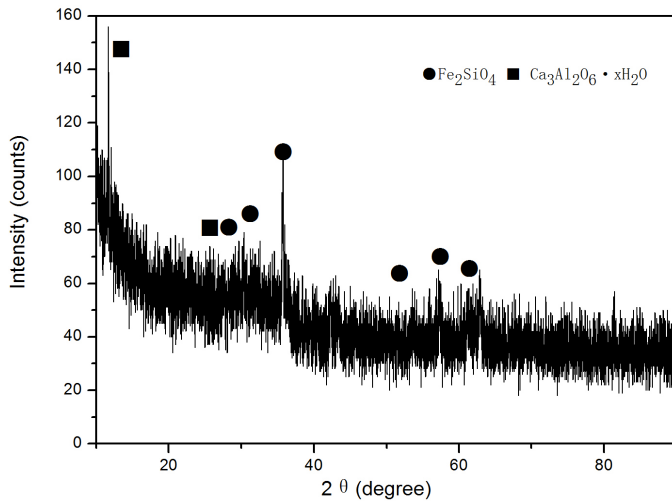
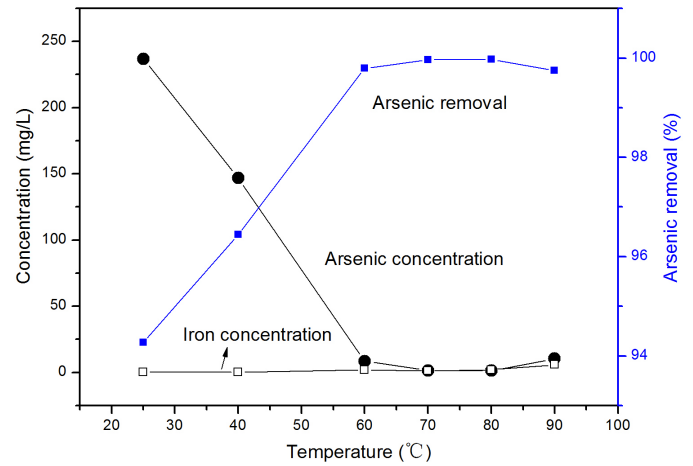


Fig. 1. XRD patterns of water-quenched slag

3.2. Effect of reaction temperature on As removal

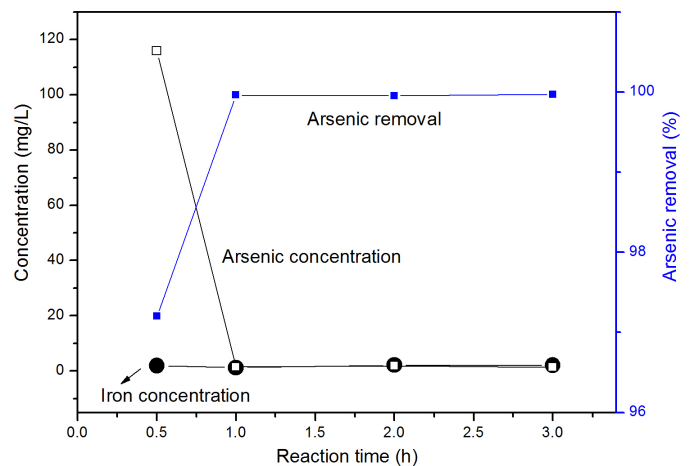
The concentrations of arsenic and iron, and arsenic removal from acid leaching solution of zinc oxide dust were studied in temperature range of 25 ~ 90 °C. Experimental conditions were shown as follows: initial concentration of As was 4.13 g/L, reaction time was 1 h, H₂O₂ addition was 10.8 mL/L and water-quenched slag addition was 17.8 g/L. The arsenic removal as a function of reaction temperature is presented in Fig. 2. As shown, the concentrations of arsenic in filtrate are 237 mg/L and 147 mg/L with arsenic removal rates of 94% and 96% when the temperatures are 25 °C and 40 °C, respectively. It was presumed that the iron (II) is not enough to react with arsenic and H₂O₂ at low temperature. The concentrations of arsenic in filtrate are reduced to 8.63 mg/L, 1.53 mg/L, 1.27 mg/L, 10.4 mg/L and arsenic removal rates are all more than 99% with the temperatures increasing up to 60°C, 70°C, 80°C and 90°C, respectively. When the reaction temperature is 90°C, the concentration of arsenic in filtrate is inversely higher than those of 70 °C and 80 °C. It may be ascribed to fast decomposition of more hydrogen peroxide at higher temperature. Meantime, the concentrations of iron in filtrates are all in the range of 0.37 ~ 5.91 mg/L (lower than impurity standard of 20 mg/L) at different reaction temperatures.

Consequently, the concentrations of arsenic in filtrate are lower than 2 mg/L and the arsenic removal rate is more than 99% when the reaction temperatures are 70°C and 80°C. Considering the cost, the optimal temperature is 70°C.

Fig. 2. As removal as a function of temperature (initial As 4.13 g/L, reaction time 1h, H₂O₂/(2Fe + As) molar ratio = 1, Fe/As molar ratio = 1.5)

3.3. Effect of reaction time on As removal

The concentrations of arsenic and iron, and arsenic removal from acid leaching solution of zinc oxide dust were researched in reaction time range of 0.5 ~ 3 h. Experimental conditions were shown as follows: initial concentration of As 4.13 g/L, temperature of 70°C, H₂O₂ addition of 10.8 mL/L and water-quenched slag addition of 17.8 g/L. Fig. 3 shows the arsenic removal as a function of reaction time. From Fig. 3, it can be seen that the concentration of arsenic in filtrate is 116 mg/L with arsenic removal rate of 97% when the reaction time is 0.5 h. However, the concentrations of arsenic in filtrate are reduced to 1.53 mg/L, 1.86 mg/L and 1.38 mg/L with arsenic removal rates of more than 99% when the reaction times are 1 h, 2 h and 3 h,

Fig. 3. As removal as a function of reaction time (initial As 4.13 g/L, temperature 70°C, H₂O₂/(2Fe + As) molar ratio = 1, Fe/As molar ratio = 1.5)

respectively. Furthermore, the concentrations of iron in filtrate are all in the range of 1.21 ~ 2.26 mg/L (lower than impurity standard of 20 mg/L) at different reaction time.

The arsenic removal rate increases apparently with increasing of the reaction time from 0.5 to 1 h. The concentrations of arsenic in filtrate are lower than 2 mg/L and the removal rate of arsenic are more than 99% when the reaction time in the range of 1 ~ 3 h. Therefore, the optimal reaction time is determined to 1 h.

3.4. Effect of oxidant hydrogen peroxide addition on As removal

The concentrations of arsenic and iron, and arsenic removal from acid leaching solution of zinc oxide dust were investigated when $H_2O_2/(2Fe+As)$ molar ratios were 0.5 (the addition of oxidant H_2O_2 is 5.4 mL), 0.8 (8.7 mL), 1 (10.8 mL), 1.2 (13 mL), 1.5 (16.2 mL) and 2 (21.6 mL), respectively. Experiment conditions were that initial concentration of As of 4.13 g/L, reaction time of 1 h, temperature of 70 °C and the water-quenched slag addition of 17.8 g/L. The arsenic removal as a function of $H_2O_2/(2Fe + As)$ molar ratio was shown in Fig. 4. It can be seen from Fig. 4 that the concentrations of arsenic in filtrate are 706 mg/L and 130 mg/L with arsenic removal rates of 83% and 97% when the $H_2O_2/(2Fe + As)$ molar ratio are 0.5 and 0.8, respectively. Meanwhile the concentrations of iron in filtrate are 632 mg/L and 40.4 mg/L. The results indicate that the oxidant H_2O_2 is insufficient for the oxidization of arsenic (III) and iron (II). However, the concentrations of arsenic in filtrate are reduced to 1.53 mg/L, 1.05 mg/L, 0.62 mg/L, and 0.21 mg/L with arsenic removal rates more than 99% when the $H_2O_2/(2Fe + As)$ molar ratios are up to 1, 1.2, 1.5 and 2, respectively. Meanwhile the concentrations of iron in filtrate are reduced to 1.21 mg/L, 1.02 mg/L, 0.49 mg/L and 0.34 mg/L (lower than impurity standard of 20 mg/L), respectively. Consequently, the concentrations of arsenic in filtrate are lower than 2 mg/L and the arsenic removal rate are more than 99% when the $H_2O_2/(2Fe + As)$ molar ratio in the range

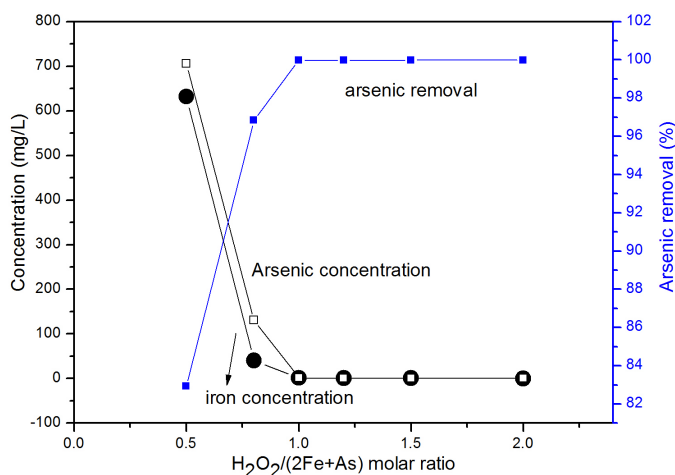


Fig. 4. As removal as a function of $H_2O_2/(2Fe + As)$ molar ratio (initial As 4.13 g/L, temperature 70°C, reaction time 1h, Fe/As molar ratio = 1.5)

of 1 ~ 2. Considering the cost, the optimum H_2O_2 addition is 10.8 mL ($H_2O_2/(2Fe + As)$ molar ratio = 1).

3.5. Effect of water-quenched slag addition on As removal

The concentrations of arsenic and iron, pH value before neutralization, limestone addition and arsenic removal from acid leaching solution of zinc oxide dust were investigated when Fe/As molar ratios were 0.8 (water-quenched slag addition is 9.49 g/L), 1 (11.9 g/L), 1.5 (17.8 g/L), 2 (23.7 g/L) and 2.5 (29.7 g/L). Experimental conditions were shown as follows: initial concentration of As = 4.13 g/L, reaction time = 1 h, temperature = 70 °C and H_2O_2 addition = 10.8 mL/L. Fig. 5 shows the arsenic removal as a function of Fe/As molar ratio. As shown in Fig. 5, the concentrations of arsenic in filtrate are 468 mg/L and 251 mg/L with arsenic removal rates of 89% and 94% when Fe/As molar ratios are 0.8 and 1, respectively. Meanwhile the iron concentrations in filtrate are 0.37 mg/L and 0.74 mg/L. The results illustrate that the water-quenched slag is insufficient for the reaction with arsenic and oxidant H_2O_2 . However, the concentrations of arsenic in filtrate are reduced to 1.53 mg/L, 5.35 mg/L, and 7.38 mg/L with arsenic removal rates more than 99% when Fe/As molar ratios are up to 1.5, 2 and 2.5, respectively. At the same time, the concentrations of iron in filtrate increase to 1.21 mg/L, 50.7 mg/L and 263 mg/L (more than impurity standard of 20 mg/L), respectively. The results illustrate that the water-quenched slag is excess when Fe/As molar ratio is 2 and 2.5. Consequently, the concentrations of arsenic or iron in filtrate will obviously increase when the water-quenched slag is insufficient or excess. The optimum addition of water-quenched slag is 17.8 g (Fe/As molar ratio = 1.5).

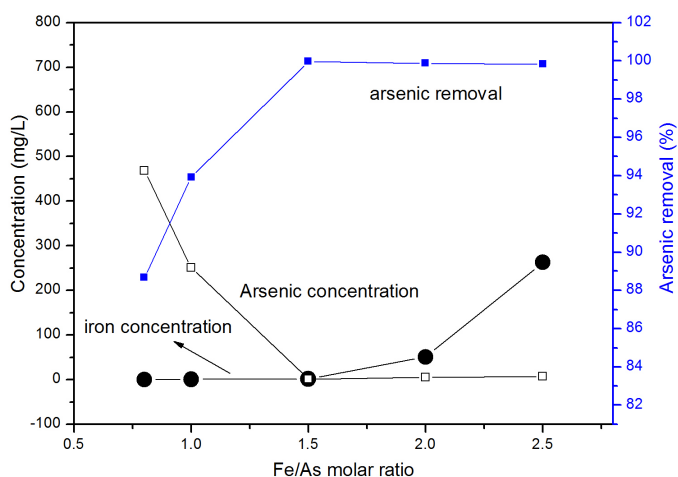


Fig. 5. As removal as a function of Fe/As molar ratio (initial As 4.13 g/L, temperature 70°C, reaction time 1h, $H_2O_2/(2Fe + As)$ molar ratio = 1)

Furthermore, the pH values before neutralization gradually increase from 2.9 to 4.2 (initial pH is 2.1) with the increasing of water-quenched slag addition while the requirement of neutralizer limestone decreases from 10.7 g/L to 3.3 g/L (shown in Fig. 6).

TABLE 2

The mass balances at the optimum condition

Item	Weight/ volume	As content (g)	Zn content (g)	Fe content (g)
Acid leaching solution	1 L	4.13	121	
H ₂ O ₂	0.0108 L			
Water-quenched slag	17.8 g	0.05	1.04	5
Limestone	5.1 g			
Water	0.5 L			
Filtrate	0.84 L	0.0013	100.8	0.001
Filter residue	46.2 g	3.86	1.86	4.67
Washed water	0.5 L	0.31	18.05	0.27
Mass balance		99.79%	98.91 %	98.82%

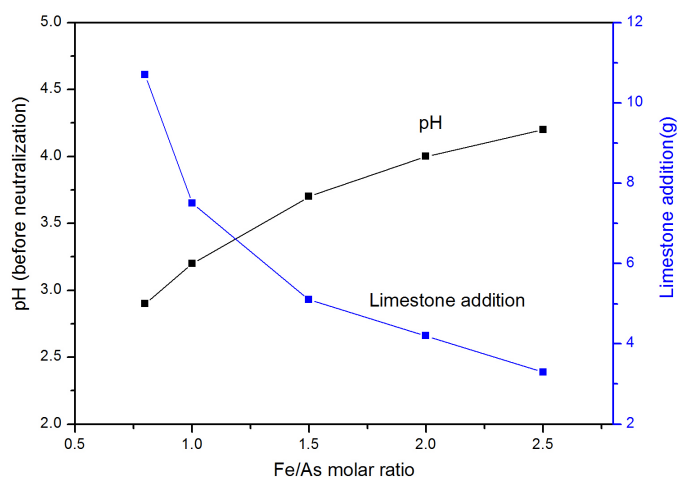


Fig. 6. pH value before neutralization and limestone addition as a function of Fe/As molar ratio (initial As 4.13 g/L, temperature 70°C, reaction time 1 h, H₂O₂/(2Fe + As) molar ratio = 1)

3.6. Characterization of filter residue prepared at the optimum condition

At the optimum condition, the arsenic (III) with high concentration (As 4.13 g/L) is efficiently removed (arsenic removal rate > 99%) from acid leaching solution of zinc oxide dust, and the filtrate (Zn 120 g/L, Fe 1.21 mg/L, As 1.53 mg/L) with low concentration of arsenic and iron is prepared successfully. The filtrate (impurity standard: As < 2 mg/L, Fe < 20 mg/L) can be send to electrolysis system for electrowinning zinc slabs. The mass balances of As, Zn and Fe at the optimum condition are listed in Table 2.

Fig. 7 shows the X-ray diffraction patterns of filter residue prepared at the optimum parameters condition. It can be seen that the main phase compositions of the filter residue are ZnSO₄·6H₂O (JCPDS#32-1478), CaSO₄·2H₂O (JCPDS#33-0311), Ca(Mg,Fe,Al)(Si,Al)₂O₆ (JCPDS#41-1483), FeAsO₄·2H₂O (JCPDS#37-0468) and FeAsO₄ (JCPDS#21-0909).

Moreover, the agglomeration and growth of crystalline grain, multi-aperture and crack are observed in the micrograph of filter residue (shown in Fig. 8). So the filter residue can be easily filtered from slurry. The result is consistent with our experimental phenomenon.

The major elements of the filter residue are listed in Table 3. Zn of 15.6% is mainly existed as ZnSO₄·6H₂O phase in the filter residue (before recovering Zn). As we known, ZnSO₄·6H₂O

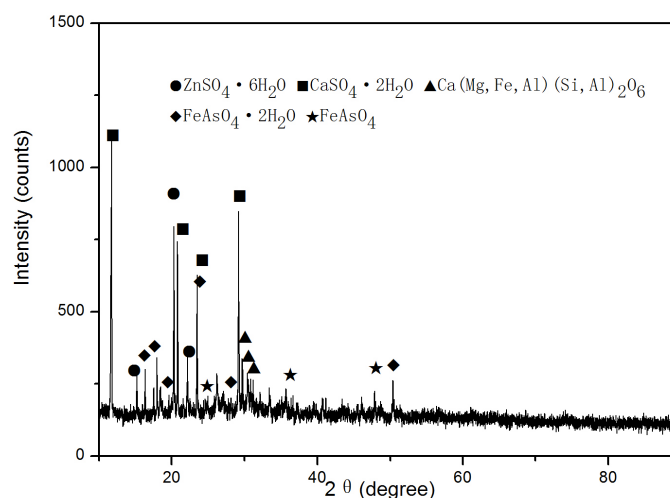


Fig. 7. XRD patterns of filter residue prepared at optimum condition (initial As 4.13 g/L, temperature 70°C, reaction time 1 h, H₂O₂/(2Fe + As) molar ratio = 1, Fe/As molar ratio = 1.5)

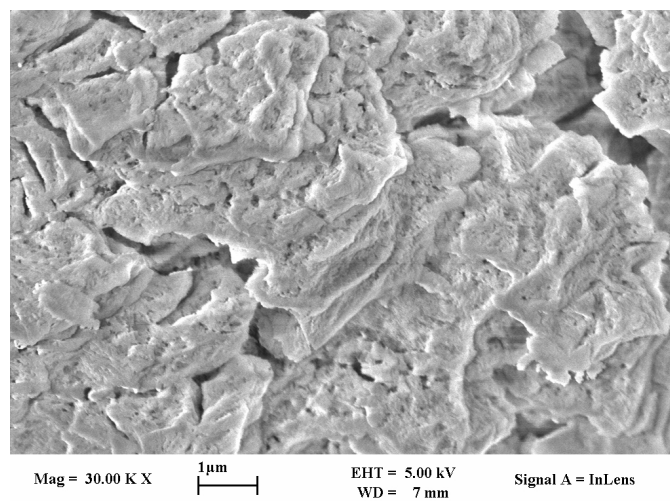


Fig. 8. SEM micrograph of filter residue prepared at optimum condition (initial As 4.13 g/L, temperature 70°C, reaction time 1 h, H₂O₂/(2Fe + As) molar ratio = 1, Fe/As molar ratio = 1.5)

can easily dissolve into water. Therefore, the filter residue was washed by water (0.5 L) to recover zinc. The washed water with Zn of 36.1 g/L, As of 0.62 mg/L and Fe of 0.53 mg/L can be

return to industrial acid leaching solution for recovering zinc. At last, the final filter residue (throw dreg) was obtained with Zn of 4.03%, Fe of 10.1%, As of 8.36%, Ca of 11%, Si of 5.74% (shown in Table 3).

TABLE 3

Major elements contents of filter residue(before recovering Zn) and throw dreg (wt. %)

Element	Zn	Fe	As	Ca	Si
filter residue (before recovering Zn)	15.6	5.35	4.42	5.01	2.71
throw dreg	4.03	10.1	8.36	11.0	5.74

According to above experiment results, the arsenic could be removed when only small amounts of H_2O_2 and water-quenched slag need to be added. Therefore, the iron in water-quenched slag can be partly leached into acid solution and reacted with arsenic (III) in the presence of oxidant H_2O_2 . Finally, ferric arsenate precipitation is formed in residue. And most possibly some adsorptive arsenic removal takes place by means of $Ca(Mg,Fe,Al)(Si,Al)_2O_6$, $Fe(OH)_3$ and the unreacted water quenched slag.

3.7. As leachability of filter residue by TCLP test

For studying the stability of compounds, the throw dregs prepared at optimum condition was subjected to TCLP test. And the As leachability is 2.61 mg/L. Consequently, stable filter residue was obtained because that the TCLP As leachability never exceeds the regulatory limit of 5 mg/L As. Maybe it is attributable to the fact that the $FeAsO_4 \cdot 2H_2O$ and $FeAsO_4$ of lowest solubility in the weak acid (pH range of 3-5) [20].

4. Conclusions

In the present study, a novel high concentration of arsenic removal method from acid leaching solution of zinc oxide dust by water-quenched slag was studied. Owing to the non-crystalline and active material with amorphous phase of the water-quenched slag, it can easily react with acid in solution as a neutralizer. So less neutralizer limestone should be expended.

Basing on the systematically studies, the optimum process parameters were obtained as follows: reaction temperature = 70°C, reaction time = 1 h, H_2O_2 addition = 10.8 ml/L, water-quenched slag addition = 17.8 g/L. At the optimum condition, the arsenic (III) with high concentration (As 4.13 g/L) was efficiently removed (arsenic removal rate more than 99%) from acid leaching solution of zinc oxide dust, and the filtrate (Fe 1.21 mg/L, As 1.53 mg/L) with low concentration of arsenic and iron was prepared successfully.

The filter residue can be easily filtered from slurry due to the existence of agglomeration and growth of crystalline grain,

multi-aperture and crack. At last, stable throw dregs are obtained because that the TCLP As leachability never exceeds the regulatory limit of 5 mg/L As.

Acknowledgments

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