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# Simulation Study and Industrial Application of Enhanced Arsenic Removal by Regulating the Proportion of Concentrates in the SKS Copper Smelting Process

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**Abstract:** Arsenic removal is a crucial issue in all copper smelters. Based on the Fangyuan 1<sup>#</sup> smelter, the effects of major elements (Cu, Fe and S) in sulfide concentrates on arsenic removal in the SKS copper smelting process were studied in this paper. The results show that Cu, Fe and S in concentrates have a significant influence on the oxygen/sulfur potential of smelting systems, and also affect the efficiency of arsenic removal. By regulating the proportion of the major elements in sulfide concentrates, the concentrate composition was changed from its original proportions (Cu 24.4%, Fe 26.8%, S 28.7%, and other 20%) to optimized proportions (Cu 19%, Fe 32%, S 29%, and other 20%). The distribution of arsenic among three phases in the original production process (gas 82.01%, slag 12.08%, matte 5.91%) was improved to obtain an optimal result (gas 94.37%, slag 3.45%, matte 2.18%). More arsenic was removed into the gas phase, and the mass fraction of arsenic in matte was reduced from 0.07% to 0.02%. The findings were applied to actual production processes in several other copper smelters, such as the Hengbang copper smelter, Yuguang smelter and Fangyuan 2<sup>#</sup> smelter. Therefore, the optimized result obtained in this work could provide direct guidance for actual production.

Keywords: arsenic removal; copper smelting; SKS; Shuikoushan process; oxygen bottom blown

# 1. Introduction

Arsenic is an element that is toxic for the environment and people's health [1–4], especially As<sub>2</sub>O<sub>3</sub>. In the smelting process for copper sulfide ores, arsenic is dispersed into the fly ash, smelting slag and copper matte [5–7]. Due to the increasing complexity of copper concentrate, arsenic control has been listed as an important issue in copper smelters [8]. The SKS (Shuikoushan) copper smelting process is very adaptable to complex concentrates and it has high arsenic removal efficiency [9–13]. In recent years, the SKS copper smelting process has become a popular research topic [14–20]. Arsenic removal in the SKS smelting process is affected by several factors, such as the composition of concentrate, matte grade, oxygen concentration in air blown into the furnace, oxygen/ore ratio, smelting temperature, the Fe/SiO<sub>2</sub> ratio in slag, and so on. In our previous study, the effects of matte grade, oxygen/ore ratio, oxygen concentration in air blown into the furnace, smelting temperature and ratio of Fe/SiO<sub>2</sub> in slag were investigated [21].

However, the composition of concentrate, especially the major elements (Cu, Fe and S), could affect the oxygen/sulfur potential of the smelting system, and further affect the removal of arsenic. Therefore, in this work, the content of the major elements (Cu, Fe and S) in sulfide concentrate was adjusted,



and the removal of arsenic from the matte to gas phase in the SKS copper smelting process was investigated through the commercial simulation software SKSSIM [22]. This work will help us to better understand the SKS copper smelting process.

#### 2. Research Methodology

The study was carried out by SKSSIM simulation software, combined with actual production in the Fangyuan 1# smelter in Dongying, China.

SKSSIM is an efficient simulation software for the SKS process, and it is based on the SKS smelting mechanism model [22] and the theory of Gibbs free energy minimization [23]. In the mechanism model, the SKS furnace is divided into seven functional layers from top to bottom and three functional regions along the length direction [21–24]. The particle swarm optimization algorithm, C# computer programming language, and Microsoft Visual Studio were used to develop the SKSSIM software. The development process (including activity coefficient, Gibbs free energy including activity coefficient, Gibbs free energy, phase entrainment coefficient, model verification and modification) has been presented in detail in our previous work [23]. SKSSIM has been successfully validated by the actual production process in Fangyuan 1# smelter [21,23,24]. Therefore, SKSSIM is a convenient method to carry out this study.

#### 3. Results and Discussion

## 3.1. Arsenic Distribution in the Actual Production Process and by SKSSIM

The initial conditions and operation parameters of the SKS process are given in our previous work [21]. The calculated results are compared with the actual industrial production data in Tables 1 and 2.

Composition (wt %)		As	Cu	Fe	S	SiO <sub>2</sub>	Others
Actual production	matte	0.07	70.77	5.52	20.22	0.51	2.91
	slag	0.08	3.16	42.58	0.86	25.24	28.08
By SKSSIM	matte	0.07	70.31	4.80	20.38	0.82	3.62
	slag	0.07	2.93	42.07	0.73	25.18	29.02

Table 1. Chemical composition of matte and slag.

Distribution (wt %)	Gas	Slag	Matte
Actual production	82.01	12.08	5.91
By SKSSIM	82.71	11.06	6.23

## 3.2. Effect of Cu Content in Concentrate on Arsenic Distribution

Figure 1 shows the effect of Cu content in concentrate on the distribution of arsenic among the gas, slag and matte phases. With an increase in Cu content, there is an increase in the proportion of arsenic in both the matte and slag phases, and the fractional distribution in the gas phase decreases. At a low Cu content (such as 18%), around 95 pct of the arsenic reports to the gas phase. As the Cu content increases to 27%, about 20% of the arsenic reports to the matte phase, and only 55% reports to the gas phase. Under the fixed total volume of oxygen blown into the SKS furnace and ratio of oxygen/ore (total volume of oxygen/total mass of dry mixed concentrates), the matte grade increases as the Cu content increases. The activity coefficient of As in matte decreases with an increase in matte grade, and higher-grade matte has a higher affinity for arsenic; hence it reduces the activity and vapor pressure of arsenic [7,25].



**Figure 1.** Comparison between the actual production and simulation results of the effect of Cu content in concentrate on the distribution of arsenic among the gas, slag and matte phases.

Therefore, a higher initial Cu content in the concentrate results in more arsenic in the matte phase, and the removal of arsenic from the smelting system to the gas phase is not very efficient.

After arsenic enters the gas phase, it then goes through the waste heat boiler, electrostatic precipitator and flue gas scrubber with the  $SO_2$  off-gas. Most arsenic compounds in off-gas condense into the solid phase and are collected in dust by the waste heat boiler and electrostatic precipitator. In the flue gas scrubber, some of the arsenic enters the waste acid. Arsenic can be detoxified or recovered from the aforementioned dust and waste acid.

#### 3.3. Effect of Fe Content in Concentrate on Arsenic Distribution

In Figure 2, the effect of the initial Fe content in the concentrate on the calculated distribution of arsenic among the gas, slag and matte phases in the SKS process are presented. With the increase in Fe content, the proportion of arsenic reporting to both the matte and slag phases decreases, and the fractional distribution in the gas phase increases. However, the change trend is not as obvious as that caused by the initial Cu content in the concentrate. When the Fe content in the concentrate is varied from 17% to 37%, the distribution of arsenic among the gas, slag and matte phases varies from 75% to 85%, from 15% to 10%, and from 10% to 5%, respectively. Therefore, the change in Fe content in the initial concentrate has a relatively tiny influence on the distribution of arsenic.



**Figure 2.** Comparison between the actual production and simulation results of the effect of Fe content in concentrate on the distribution of arsenic among the gas, slag and matte.

#### 3.4. Effect of S Content in Concentrate on Arsenic Distribution

In Figure 3, as the S content in the initial concentrate increases, the proportion of arsenic reporting to the matte and slag phases decreases sharply, and the arsenic reporting to the gas phase increases. Therefore, the change in S content has a relatively significant influence on the distribution of arsenic. As the S content in the feed increases, the oxygen potential in the smelting system decreases and the sulfur potential increases, causing higher partial pressure of  $S_2$  and hence, a higher degree of volatilization of arsenic as AsS [7,24]. Therefore, high S content in the initial concentrate is beneficial for removing arsenic to the gas phase from the matte phase.



**Figure 3.** Comparison between the actual production and simulation results of the effect of S content in concentrate on the distribution of arsenic among the gas, slag and matte.

#### 3.5. Effect of Arsenic Content in Concentrate on Arsenic Distribution

Figure 4 shows the effect of the initial content of arsenic in copper concentrate on the deportment of arsenic to the phases. In China, the maximum allowed content of arsenic in copper concentrates imported from abroad is 0.5%. In general, the arsenic content in feed is 0.2%–0.7% in the copper smelting process. As shown in Figure 4, in the range studied, the proportion of arsenic reporting to both the gas and slag phases increases slightly with an increase in the arsenic content in copper concentrate. The volatilization of arsenic increases as a result of the arsenic being transferred to the gas phase as AsS, AsO, and As<sub>2</sub>.



**Figure 4.** Comparison between the actual production and simulation results of the effect of arsenic content in copper concentrate on the distribution of arsenic among the gas, slag and matte.

## 3.6. Interactive Effect of Cu and S Content in Concentrate on Arsenic Distribution

As shown in Figures 1–3, it is obvious that the Cu and S elements in concentrates have a more significant influence than Fe on the distribution of arsenic among the gas, slag and matte phases. Therefore, it is necessary to discuss the interactive effect of Cu and S content on arsenic distribution.

#### 3.6.1. Interactive Effect on Arsenic Distribution

In this work, the symbol % represents the proportion of arsenic distribution among the gas, slag, and matte phases, and the symbol wt % represents the mass fraction of arsenic in different phases.

The fractional distribution of arsenic in the gas phase with the change in the initial content of Cu and S in concentrate is given in Figure 5a. The mass fraction of arsenic in gas is also shown in Figure 5b. The variation trends in the two figures are basically consistent. As both the Cu and S content in concentrate increases, both the fractional distribution and mass fraction of arsenic in the gas phase decrease. With Cu 18% and S 28%, around 95% of the arsenic enters the gas phase, and the mass fraction of arsenic in the gas phase is nearly 0.6%. However, as the Cu and S content in the concentrate increase to 24% and 34%, respectively, only 74% of the arsenic reports to the gas phase, and the mass fraction of arsenic in the gas phase is around 0.41%. Therefore, low initial content of Cu and S in concentrate helps to eliminate arsenic from the smelting system to the gas phase. However, these results contradict the result of Figure 3, that is, as the S content in the initial concentrate increases, the proportion of arsenic reporting to the gas phase increases. This phenomenon may be explained because Cu in concentrate has a stronger impact on arsenic distribution than S. Cu, S and Fe exist in concentrate in the form of CuFeS<sub>2</sub>, CuS, Cu<sub>2</sub>S, FeS<sub>2</sub>, etc., the content of the elements have close relevance, and the interactive relationship to arsenic distribution is complex.



Figure 5. Arsenic distribution in the gas phase: (a) fractional distribution; (b) mass fraction.

The fractional distribution and mass fraction of arsenic in the slag phase with the change in the initial content of Cu and S in the concentrate are presented in Figure 6a,b. The variation trend in Figure 6 is contrary to Figure 5. As the Cu and S content in concentrate increases, the fractional distribution and mass fraction of arsenic in the slag phase also increase. With Cu of 18% and S of 28%, only around 2.1% arsenic enters the slag phase, and the mass fraction of arsenic in the slag phase is about 0.012%. However, as the Cu and S content in concentrate increases to 24% and 34%, respectively, about 24% arsenic reports to the slag phase, and the mass fraction of arsenic in the slag phase is nearly 0.18%. Therefore, a high initial content of Cu and S in concentrate helps to eliminate arsenic from the smelting system to the slag phase.



Figure 6. Arsenic distribution in the slag phase: (a) fractional distribution; (b) mass fraction.

In the matte phase, the fractional distribution and mass fraction of arsenic in relation to the change in the initial content of Cu and S in concentrate are presented in Figure 7a,b. The variation trend in the matte phase is different from the gas and slag phases. As the S content in concentrate increases and Cu decreases, both the fractional distribution and mass fraction of arsenic decrease in the matte phase. With Cu 24% and S 28% in concentrate, about 6.3% arsenic reports to the matte phase, and mass fraction of arsenic in the matte phase is nearly 0.073%. However, with Cu 18% and S 34% in concentrate, about 1.1% of the arsenic enters the matte phase, and the mass fraction of arsenic in the matte phase is nearly 0.012%.



Figure 7. Arsenic distribution in the matte phase: (a) fractional distribution; (b) mass fraction.

In the copper smelting process, it is necessary to eliminate arsenic from the matte to gas or slag phase as much as possible. However, arsenic removal is just one parameter; if the concentrate composition is changed, many other important parameters need to be considered, such as smelting temperature, matte grade, copper loss to slag, slag type (Fe/SiO<sub>2</sub>), Fe<sub>3</sub>O<sub>4</sub> content in slag, and so on.

# 3.6.2. Interactive Effects on Other Parameters of the SKS Process

In order to ensure normal production and avoid big changes in other major process parameters (smelting temperature, copper loss to slag, matte grade, etc.), the impact of major elements (Cu, Fe and S) in sulfide concentrate on other parameters (matte grade, slag type  $Fe/SiO_2$ , smelting temperature, copper loss to slag, Fe<sub>3</sub>O<sub>4</sub> content in slag, S content in slag) was evaluated.

Matte grade, smelting temperature and slag type are the main factors considered in the copper smelting process. Matte grade with the change in the initial content of Cu and S in concentrate is given in Figure 8a. As the S content in concentrate decreases and Cu increases, matte grade increases. With Cu 24% and S 28% in concentrate, the matte grade increases to about 70%. With Cu 18% and S 34% in concentrate, the matte grade drops to around 50%. Therefore, as the total oxygen blown into the SKS furnace is fixed, to produce high grade matte, the initial content of Cu in concentrate should be increased, and the initial content of S in concentrate should be reduced. However, in the Hengbang copper smelter (Yantai, China), low grade matte (around 50% [13]) was chosen, and more arsenic was removed to the gas phase.



(e)

Cu in concentrate (wt%)

(**f**)

Cu in concentrate (wt%)

**Figure 8.** Interactive effects on other parameters of the SKS process: (**a**) matte grade; (**b**) slag type Fe/SiO<sub>2</sub>; (**c**) smelting temperature; (**d**) copper loss to slag; (**e**) Fe<sub>3</sub>O<sub>4</sub> content in slag; (**f**) S content in slag.

Slag type could affect the copper loss to slag and the capacity to remove impurities. Slag type Fe/SiO<sub>2</sub> with the change in the initial content of Cu and S in concentrate is given in Figure 8b. As the Cu and S content in concentrate both increase, slag type Fe/SiO<sub>2</sub> decreases. With Cu 18% and S 28%, Fe/SiO<sub>2</sub> is about 1.9. However, as Cu and S content in concentrate increase to 24% and 34%, respectively, Fe/SiO<sub>2</sub> is nearly 1.2. In the SKS process, Fe/SiO<sub>2</sub> should be around 1.7.

Smelting temperature with the change in the initial content of Cu and S in concentrate is given in Figure 8c. As both the Cu and S content in concentrate increase, the smelting temperature decreases. With Cu 18% and S 28%, Fe/SiO<sub>2</sub> is about 1200 °C. However, as Cu and S content in concentrate increases to 24% and 34%, respectively, Fe/SiO<sub>2</sub> is nearly 1050 °C. In the actual industrial production of the SKS copper smelting process, the smelting temperature should be above 1170 °C.

Copper loss to slag is an important factor in production. The mass fraction of copper in slag with the change in the initial content of Cu and S in concentrate is given in Figure 8d. As S content in concentrate decreases and Cu increases, the copper loss to slag increases. With Cu 24% and S 28% in concentrate, the mass fraction of copper in slag increases to about 2.9%. However, with Cu 18% and S 34% in concentrate, the mass fraction of copper in slag drops to around 1.5%. In industrial production, the mass fraction of copper in slag should be controlled below 2.5% for the high direct recovery rate of copper.

In slag,  $Fe_3O_4$  can increase the viscosity of slag and further increase the copper loss to slag, thus  $Fe_3O_4$  content in slag is a very important indicator. In Figure 8e,  $Fe_3O_4$  content in slag with the change in the initial content of Cu and S in concentrate is given. As both the Cu and S content in concentrate increase, the  $Fe_3O_4$  content in slag decreases. With Cu 18% and S 28%, the  $Fe_3O_4$  content in slag is about 27%. However, when the Cu and S contents in concentrate increase to 24% and 34%, respectively, the  $Fe_3O_4$  content in slag is nearly 24.5%.

The mass fraction of sulfur in slag with the change in the initial content of Cu and S in concentrate is given in Figure 8f. As S content in concentrate decreases and Cu increases, the S content in slag increases. With Cu 24% and S 28% in concentrate, the S content in slag increases to about 0.74%. However, with Cu 18% and S 34% in concentrate, the S content in slag drops to around 0.63%. The S element exists in slag mainly in the form of Cu<sub>2</sub>S and FeS by the mechanical entrainment of matte.

In short, under normal production or little change of other major process parameters (smelting temperature, copper loss to slag, matte grade, copper loss to slag, slag type Fe/SiO<sub>2</sub>, Fe<sub>3</sub>O<sub>4</sub> content in slag, etc.), the proportions of ore should be optimized to remove more arsenic from the matte to the gas or slag phase in the actual industrial production.

#### 3.7. Industrial Application and Verification

The commercial SKS smelting furnace in the Fangyuan 1<sup>#</sup> smelter (Dongying, China) is shown in Figure 9. In the Fangyuan 1<sup>#</sup> smelter, gold concentrates bearing arsenic are treated with copper sulfide concentrates by using the SKS smelting process. As more and more gold concentrates bearing arsenic are added to copper sulfide concentrates, the Cu content in the final mixed concentrate decreases gradually. Therefore, the composition of the final mixed concentrate should be optimized.

The impurity element As could be greatly eliminated from the matte to gas phase by adjusting the concentrate composition. In this work, the oxygen rate was 12,072  $\text{Nm}^3 \cdot \text{h}^{-1}$ , the oxygen concentration in oxygen-enriched air was 73%, the feed rate of dry concentrate was 66 t·h<sup>-1</sup>, and the ratio of oxygen/ore was 183  $\text{Nm}^3 \cdot \text{t}^{-1}$ . The matte grade, smelting temperature, slag type Fe/SiO<sub>2</sub> were around 70%, 1200 °C and 1.7, respectively.

Through optimization, as shown in Figure 10, the concentrate composition was changed from its original proportions (Cu 24.4%, Fe 26.8%, S 28.7%, and other 20%) to optimized proportions (Cu 19%, Fe 32%, S 29%, and other 20%), and the distribution of arsenic among three phases was changed from the original (gas 82.01%, slag 12.08%, matte 5.91%) to optimized results (gas 94.37%, slag 3.45%, matte 2.18%). The mass fraction of arsenic in matte was reduced from 0.07% to 0.02%.



**Figure 9.** Commercial SKS smelting furnace in the Fangyuan 1<sup>#</sup> phase smelter.



**Figure 10.** Optimization of ore proportions for arsenic removal in the Fangyuan 1<sup>#</sup> smelter.

The research results and change rules were also confirmed by actual production at several other copper smelters, such as the Hengbang smelter, Yuguang smelter and Fangyuan 2<sup>#</sup> smelter. By decreasing the Cu content in concentrate, the arsenic removal ratio to gas increases, and residual ratios in slag and matte decrease. The change tendency is shown clearly in Table 3 and Figure 11.

Table 3. Main composition of concentrate and distribution ratios of arsenic.

Name of Smelter	Composition of C	Distribution Ratios of Arsenic (%)			
	Cu	Other	Gas	Slag	Matte
Yantai Hengbang [13]	16.10	83.90	88.35	7.51	4.14
Fangyuan 2 <sup>#</sup>	20.46	79.54	72.50	20.37	7.13
Jiyuan Yuguang	22.67	77.33	55.89	36.08	8.03



Figure 11. Arsenic distribution in different copper smelters.

Therefore, the optimized result identified in this work provides direct guidance to actual production, and can reduce the load of removing impurities in the conversion, refining and electrolytic processes in SKS copper smelters.

# 4. Conclusions

The main elements (Cu, Fe and S) in sulfide concentrates have a significant influence on the arsenic distribution among gas, slag and matte phases. When the Cu content in concentrate increases, the fractional distribution of arsenic in smelting products increases, except in the gas phase. As the Fe content increases, the proportion of arsenic reporting to the gas phase increases, however, there are opposite changes in the matte and slag phases. When the S content in the initial concentrate increases, the fractional distribution of arsenic in both the matte and slag phases decreases sharply, and reporting to the gas phase increases. Through optimization of the ore proportions, the concentrate composition was changed from its initial proportions (Cu 24.4%, Fe 26.8%, S 28.7%, and other 20%) to optimized proportions (Cu 19%, Fe 32%, S 29%, and other 20%), and the distribution of arsenic among three phases was improved from the initial (gas 82.01%, slag 12.08%, matte 5.91%) to an optimized distribution (gas 94.37%, slag 3.45%, matte 2.18%). The mass fraction of arsenic in matte was reduced from 0.07% to 0.02%. The optimized result could provide direct guidance for actual production.

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