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Relationship between copper content of slag and matte in the SKS copper smelting process

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Abstract: In the newly developed oxygen-enriched bottom-blowing copper smelting process (also known as the SKS copper smelting process), Cu loss in slag is one of the most concerning issues. This paper presents our research results concerning the relationship between the Cu content of the matte and slag in the SKS process; the results are based on actual industrial production in the Dongying Fangyuan copper smelter. The results show that the matte grade strongly influences Cu losses in slag. The dissolved and entrained losses account for 10%–20% and 80%–90% of the total SKS industrial Cu losses in slag, respectively. With increasing matte grade, the dissolved and entrained Cu losses in the SKS slag both increase continuously. When the matte grade is greater than 68%, the content of Cu in the smelting slag increases much more dramatically. To obtain a high direct recovery of copper, the matte grade should be less than 75% in industrial SKS copper production.

Keywords: copper smelting; oxygen-enriched bottom-blowing; matte grade; Cu loss; SKS process

1. Introduction

The pyrometallurgical process, because of its high production efficiency and adaptability for complex raw materials, still plays a dominant role in copper production [1–3]. Currently, the development of a cleaner and more efficient copper metallurgical process is a key R&D trend [4–6]. The oxygen-enriched bottom-blowing bath smelting process (also known as the SKS process), which originated in China, is an achievement of independent research and development for modern intensifying metallurgical techniques. The process has been widely used to deal with complex resources and produce Cu [7–9], Pb [10], Sb [11], and other metals. The earliest industrial oxygen-enriched bottom-blowing copper bath smelting furnace (SKS furnace) was put into production in the Fangyuan smelter (Dongying, China) in 2008. The SKS furnace was initially designed to manufacture 50 thousand tons of copper per year. In 2010, the product capacity was expanded to 100 thousand tons per year [12]. Since then, the SKS copper smelting process has been rapidly adopted in China. The process has thus far been successfully applied in more than a dozen smelters, including the Baotou Huading smelter, Yantai Hengbang smelter, Jiyuan Yuguang smelter, Hengyang Minmetals copper smelter, and the Sanmenxia Zhongyuan gold smelter. Currently, the process is being promoted overseas. Many smelters in South America (such as Codelco in Chile) are planning to use the SKS process to replace their traditional Teniente process.

Although the SKS process has been widely applied in China, Cu loss during the SKS process is still a problem. Table 1 summarizes the composition of mattes produced and the copper content of the corresponding smelter slag in several typical copper smelting processes worldwide [13]. Reducing slag Cu loss in the smelting process could greatly improve copper recoveries and reduce the burden of the copper slag cleaning process afterward. The smelting temperature, slag type (Fe/SiO_2 ratio), and matte grade strongly

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influence slag Cu losses. Chen *et al.* [14] discussed the effects of smelting temperatures and slag type (Fe/SiO_2 ratio) on Cu losses in actual industrial copper smelting slag from the SKS process in the Dongying Fangyuan smelter. Liu *et al.* [15] investigated the slag phase diagram under the SKS smelting conditions. In a certain range, increasing the content of ZnO or Al_2O_3 could substantially increase the smelting liquid temperature of the SKS slag and further increase the viscosity of slag and the entrained Cu losses in the slag. Tan [16] researched copper losses in rotary holding furnace (RHF) slag and developed four key performance indicators (slag layer thickness, plan of skimming and tapping cycle, settle time, and slag skimming rate) to reduce the Cu losses in RHF slag. Jalkanen *et al.* [17] analyzed the mineralogy and morphology of Cu in solidified smelter slags. Ruşen *et al.* [18–19] discussed the effects of some additives (CaO, B_2O_3, and colemanite ($2CaO{\cdot}3B_2O_3{\cdot}5H_2O$)) on Cu losses to matte smelting slag. Coursol *et al.* [20] researched the optimal operating conditions of minimization of Cu losses in Cu smelting slag during electric furnace treatment.

Table 1. Compositions of matte and slag in typical copper smelting processes [13] wt%

Smelter	Cu in matte	Fe in matte	S in matte	Cu in slag	S in slag	Furnace type
Outokumpu	70	8	21	1.4	0.5	Flash
Naoshima	65.7	9.2	21.9	0.6	0.3	Mitsubishi
Cyprus	58	15.9	—	0.6	—	Isasmelt
Inco	45	26	25	0.63	—	Flash
Lubumbashi	73	6	20	1.1	—	Blast
El Teniente	50.3	21	—	0.88	1.2	Reverb-wet
Mufulira	47	24	23.8	0.66	0.9	Reverb-wet
Anaconda	52	20	—	0.75	—	Elec

However, thus far, no research on the relationship between the Cu content of matte and slag in the SKS Cu bath smelting process has been reported. We studied this relationship, gaining a better understanding of how Cu losses occur in the SKS Cu bath smelting slag so that we can provide guidance on how to decrease Cu losses in slag.

2. Experimental

2.1. Commercial SKS furnace

The SKS smelting furnace is shown in Fig. 1. It is a horizontal cylindrical vessel with adjustable gas injection lances [12]. Nine lances were equipped on the bottom of the SKS furnace in two rows in the length direction. Five lances were installed in the lower row at 7°, and four lances were installed in the upper row at 22°. The external dimensions of the furnace were ϕ4.4 m × 16.5 m. Chrome–magnesia bricks (380 mm) were lined inside the furnace to avoid corrosion of the furnace by the melt. The feed rate of dry mixed concentrates was 65 t/h. The production capacity (copper) of the SKS reactor in Fig. 1 is approximately 100000 t annually.

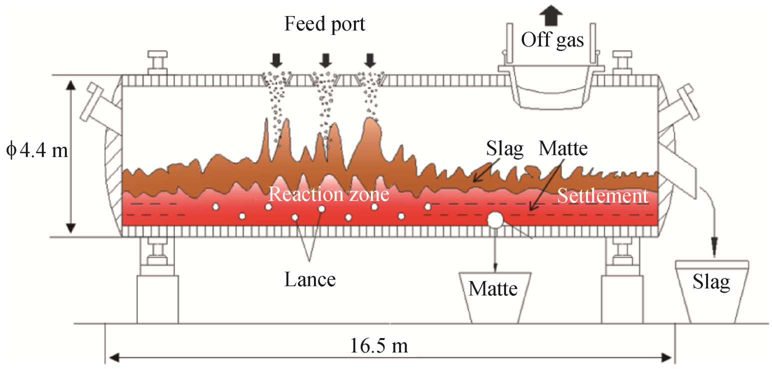


Fig. 1. Dimensions of the first-stage SKS furnace at the Fangyuan smelter [12].

2.2.Characteristics of the process

On the basis of the operational requirements for charging compositions, the Cu concentrates, silica flux, and some recycling materials containing Cu were blended together. The raw materials, without being ground, dried, or pelletized before mixing, were charged directly into the SKS furnace from three top feed ports of the furnace. Air enriched with oxygen was blown into the matte layer directly and constantly through the lances. The oxygen-enriched air was then split into numerous tiny flows with high speed and dispersed in the molten matte and slag. The oxygen-enriched air contacted the melt sufficiently and accelerated mixing, as depicted in Fig. 2. The oxidation–reduction reaction proceeded violently, enhancing the reaction efficiency in the SKS reactor.

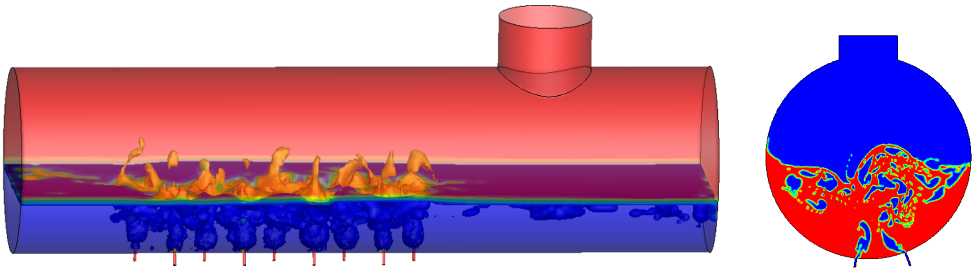


Fig. 2. Gas–liquid interaction in an SKS furnace.

2.3. Research methodology and calculation process

On the basis of an investigation of the characteristics [12] in an SKS smelting furnace and the total Gibbs free energy minimization thermodynamic theory, a thermodynamic mechanism model [21] for the process was established to illustrate the distribution behavior of the elements. Meanwhile, specific simulation software [22] named SKSSIM (SKS Simulation) was developed to carry out the present study, as depicted in Fig. 3. The SKSSIM software has been repeatedly verified and successfully applied in the Fangyuan smelter and the Minmetals copper smelter. The SKSSIM software is a convenient way to analyze the SKS process. To study the behavior of Cu, we performed SKS process calculations by regulating the oxygen–ore ratios to study the influence of the matte grade on Cu losses in SKS smelting slag.

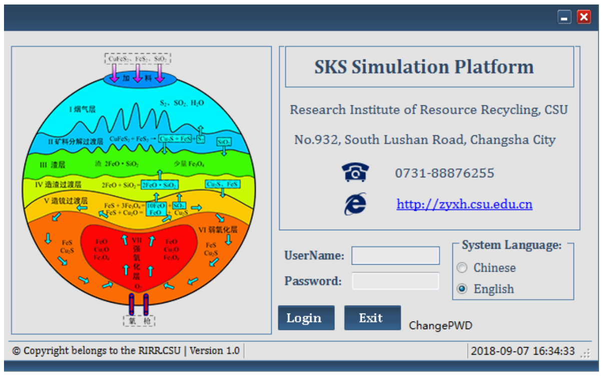


Fig. 3. SKSSIM software.

The compositions of the mixed concentrates in an actual plant were provided by the Fangyuan smelter, as given in Table 2. The normal and stable operation parameters of actual industrial production are listed in Table 3.

Table 2. Compositions of the mixed copper concentrates wt%

Fe	Cu	S	SiO_2	MgO	CaO	Al_2O_3	As	Sb	Bi	Pb	Zn	Bal.
26.77	24.38	28.67	6.44	1.95	2.45	2.27	0.35	0.13	0.12	0.98	1.91	3.58

Table 3. Industrial operation parameters in the Fangyuan smelter

Stable industrial parameter	Value
Feed rate of dry raw concentrates / $(t \cdot h^{-1})$	65
Moisture content in concentrates / wt%	9.85
Feed rate of quartz sand flux / $(t \cdot h^{-1})$	5.3
Temperature / °C	1180–1200
Gas relative pressure in SKS furnace / Pa	−48 – −185
Flow rate of oxygen / $(m^3 \cdot h^{-1})$	10800
Flow rate of air / $(m^3 \cdot h^{-1})$	5700
Volume fraction of O_2 in the final air / vol%	73
Matte grade / %	70

3. Results and discussion

3.1. Verification of calculation results

A comparison between the results calculated using the SKSSIM software and the industrial data provided by the Fangyuan smelter under stable production conditions is presented in Table 4.

The industrial data (compositions of matte and slag) under the stable production conditions were also taken from the Fangyuan smelter.

The calculated chemical compositions of the matte and slag agree well with the industrial values. Accordingly, the SKSSIM software is reliable and demonstrates valuable practice capacity. The software can be used to simulate the slag Cu losses and to determine the optimum parameters for the SKS process.

3.2. Relationship between matte grade and the Fe/SiO_2 ratio of slag

In our study, by regulating the flow rate of air or oxygen, as shown in Table 3, we investigated a wide range of the flow rate of the whole oxygen (the combined oxygen of air and pure oxygen) from 5450 m^3/h to 12800 m^3/h. Meanwhile, the oxygen concentration in oxygen-enriched air was 73vol%. As shown in Fig. 4, as the flow rate of total oxygen increased, the matte grade increased gradually. More FeO was generated and entered the slag; thus, the Fe/SiO_2 ratio also increased. The matte grade changed from 42.61% to 76.40%, and the Fe/SiO_2 ratio changed from 0.81 to 1.73. In addition, the feeds compositions charged into the SKS furnace remained constant in the calculation.

Table 4. Comparison between the industrial data and the calculation results wt%

Data sources	Material	Fe	Cu	SiO_2	S	As	Pb	Bi	Zn	Sb
Industrial data from Fangyuan smelter	Matte	5.48	70.81	0.56	20.01	0.08	1.66	0.07	1.05	0.03
	Slag	42.55	3.19	25.22	0.85	0.06	0.41	0.03	2.17	0.12
Calculated result by SKSSIM	Matte	4.85	70.42	0.77	20.28	0.06	1.72	0.06	1.03	0.04
	Slag	42.33	3.01	25.3	0.81	0.07	0.39	0.02	2.09	0.11

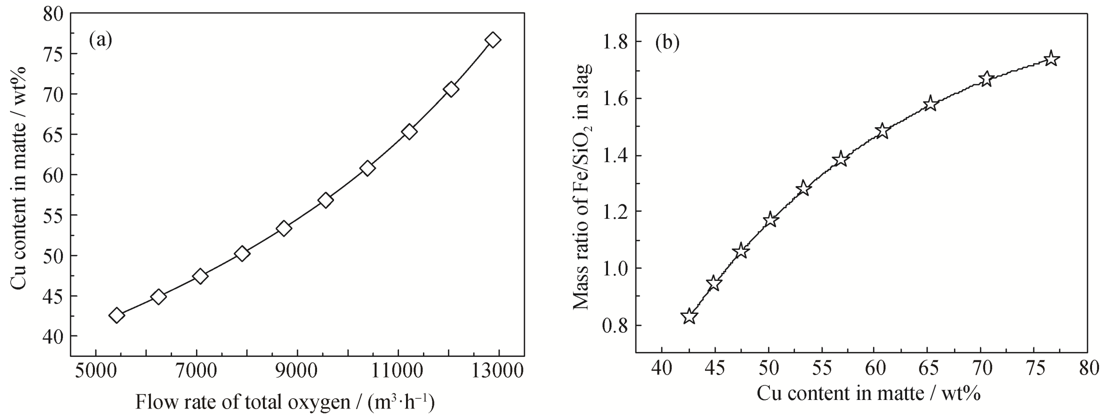


Fig. 4. Relationship of different process parameters: (a) matte grade and flow rate of total oxygen; (b) matte grade and Fe/SiO_2 ratio in slag.

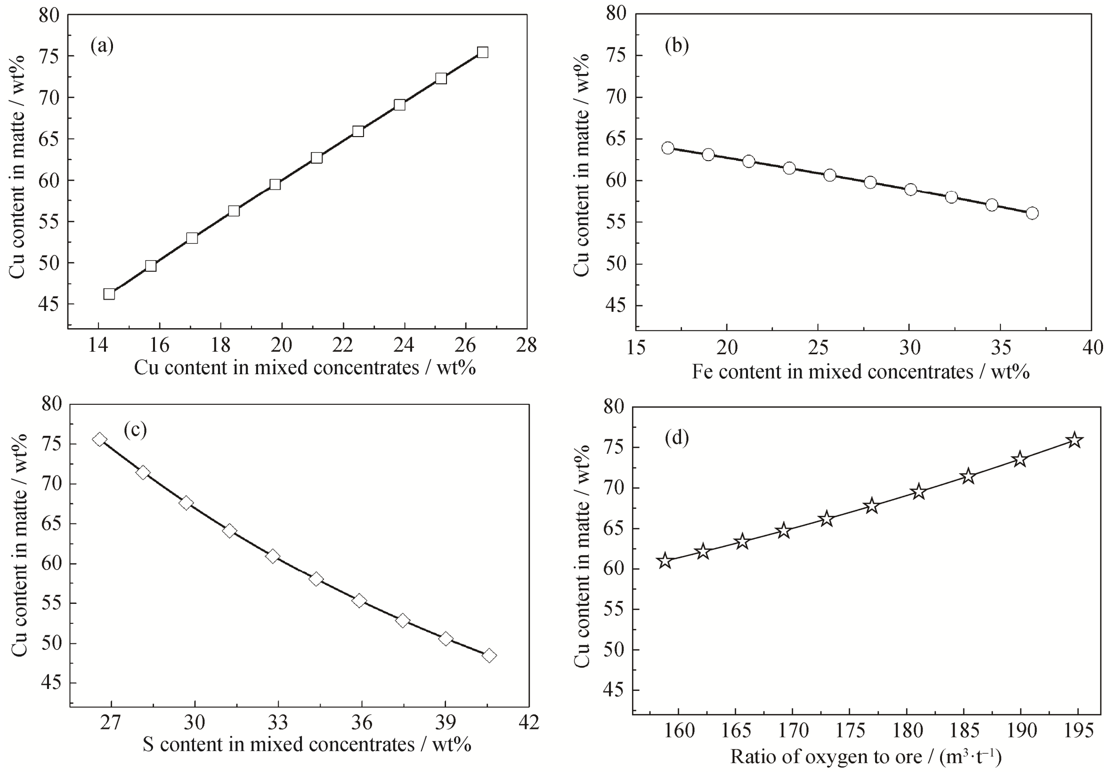
In actual production, the matte grade can also be affected by changes of other parameters such as the Cu, Fe, and S content in the copper concentrates, the oxygen/ore ratio, and the oxygen content in the blown air, as shown in Fig. 5. However, in the present work, we mainly discuss the relationship between the matte grade and copper losses in slag.

3.3. Influence of matte grade on Cu loss in slag

The relationship between the Cu content of slag and matte is an important index to assess the property and capability of a smelter. As shown in Table 4, the Dongying Fangyuan smelter was producing high-grade matte (> 70% Cu), and the content of Cu in the slag was approximately 3wt% (mass percentage). Because the matte grade strongly influences the Cu loss to slag, the SKSSIM software was used to calculate the variation tendency of the Cu loss to the SKS smelting slag with increasing matte grade.

Commonly, Cu losses in slag are divided into two categories [14,23]: (1) Cu^+ dissolved in molten liquid slag in the form of Cu_2O or Cu_2S; (2) matte droplets mechanically entrained in the slag. The copper losses to slag in the SKS industrial production experiment with the matte grade fixed at approximately 75% are shown in Fig. 6. All of the dissolved, entrained, and total Cu losses in slag increased with increasing Fe/SiO_2 ratio of the slag.

In addition to the Fe/SiO_2 mass ratio of slag, numerous other physical and chemical factors can potentially influence the Cu loss in slag. Yazawa *et al.* [24] and Shimpo *et al.* [25] have discussed the dissolved Cu losses in slag on the basis of lab test data. The data from these researchers [24–25] and the calculated dissolved Cu losses are plotted in Fig. 7. As shown in Fig. 7, when the matte grade is less than 69%, the results given by SKSSIM are similar to the previous data [24]. However, when the matte grade is nearly 75%, the calculation results are consistent with the data of Chen *et al.* [14], which are intermediate between the results of previous reports [24–25]. Although the simulation conditions in this study differ from the selected experiments by researchers [24–25], their research results demonstrate that the dissolved Cu loss in slag is mainly less than 1wt% and much less than the entrained loss.



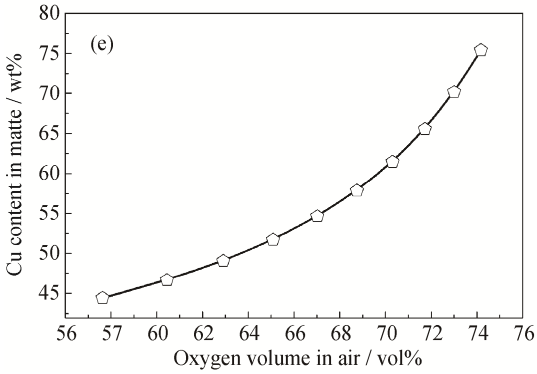


Fig. 5. Relationship between the matte grade and other parameters in the SKS process: (a) Cu content in copper concentrates; (b) Fe content in copper concentrates; (c) S content in copper concentrates; (d) oxygen/ore ratio; (e) oxygen content in air.

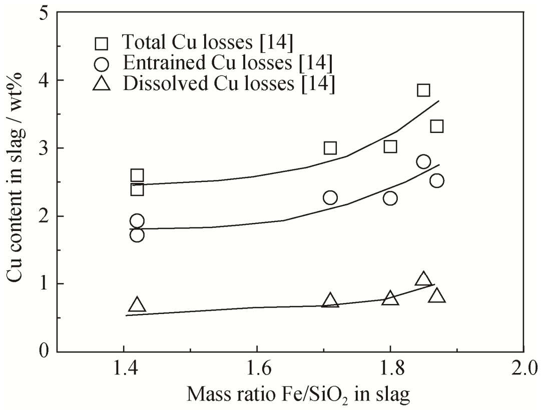


Fig. 6. Relationship of Cu losses and the Fe/SiO$_2$ ratio of slag in SKS industrial production experiments.

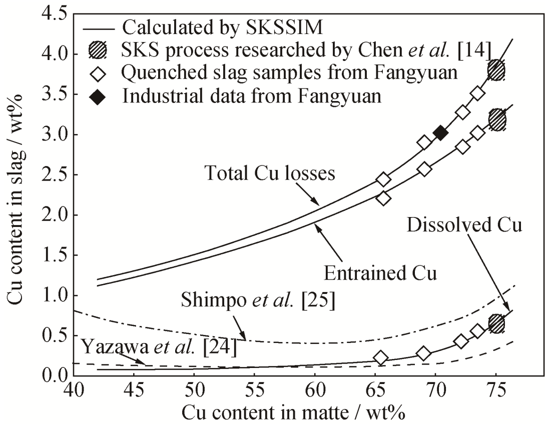


Fig. 7. Cu losses in slag in the SKS copper smelting process.

Fig. 7 also shows that the Cu content in slag under normal industrial production conditions of the SKS process is very similar to the result calculated by SKSSIM. With increasing content of Cu in matte, the content of Cu in slag increases. When the content of Cu in the matte increases to 69wt%, total slag Cu losses increase much more dramatically. The alteration of the chemical and physical properties of the iron-silicate slag can explain the present phenomenon. When the content of Cu in the matte increases, the oxygen potential in the SKS smelting system and the Fe/SiO_2 mass ratio in the slag both increase. More Fe_3O_4 in the slag has been oxidized from FeO, which results in increasing viscosity of iron-silicate slag; thus, the settling velocity of droplets of copper matte in molten liquid slag decreases. A high slag viscosity can result in a mass of physical entrainment Cu losses in the slag. This conclusion is demonstrated by the research of Chen *et al.* [14], who found that the entrained part could account for > 72% of the total Cu losses under the conditions of an Fe/SiO_2 ratio of 1.7 and a matte grade of approximately 75%. Therefore, for better Cu recovery, the matte grade should be less than 75%.

3.4. Impacts of matte grade on Fe_3O_4 and FeO content in slag

"FeO"–SiO_2 is a base system for copper smelting slag. Some researchers [26–28] have reported the phase diagram of this system, as shown in Fig. 8. The fayalite (Fe_2SiO_4) primary phase at iron saturation is replaced by a spinel primary phase when the partial pressure of oxygen (Po_2) is fixed at 10^{-3} Pa (similar to the SKS smelting condition). Under normal and stable production conditions of the SKS smelting process in the Fangyuan smelter, the smelting temperature is approximately 1200°C (1473 K) and the Fe/SiO_2 mass ratio of the slag is approximately 1.7 (the SiO_2 content in the slag is approximately 25wt%). Fig. 8 shows that the operating temperature is lower than the liquid temperature line. A substantial amount of spinel (Fe_3O_4) phase is present in the liquid phase, which results in an increase of the slag viscosity.

To further analyze the relationship between Cu loss and matte grade in the SKS process, some quenched slag samples were taken from the Dongying Fangyuan smelter. As shown in Fig. 9, at the beginning of slag tapping, through a tapping hole, a cold and long iron bar was used to dip into the molten liquid slag to collect the quenched slag sample. Before the next sampling, the iron rod was immersed in water and cooled. The sampling techniques have been described elsewhere [14].

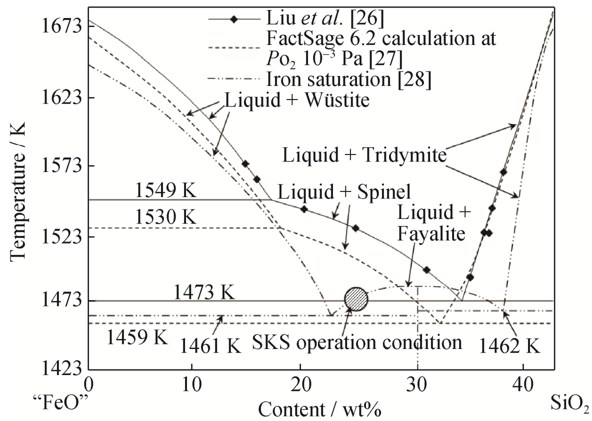


Fig. 8. "FeO"–SiO_2 system at $Po_2 = 10^{-3}$ Pa.

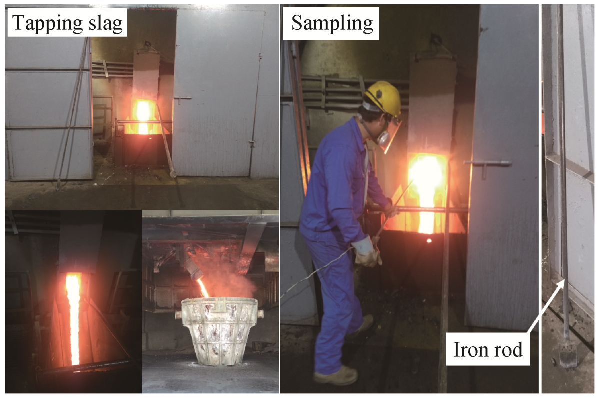


Fig. 9. Sampling slag from the bottom-blowing furnace.

The total content of Fe in slag was obtained by X-ray fluorescence analysis. The content of FeO in the slag was measured by the potassium dichromate titration method [29]. The Fe^{3+} or Fe_3O_4 content in slag was calculated on the basis of the Fe^{2+} content and the total Fe content. The microstructures of four quenched samples of slag are shown in Fig. 10. Detailed concentrations of iron oxides and Cu losses from the compositional analysis are listed in Table 5. The dissolved, entrained, and total Cu losses in the quenched slag sample are very similar to the SKSSIM calculation results. As depicted in Fig. 7, the entrained Cu losses account for 85%–92% of the total Cu losses.

With increasing matte grade, more FeO is generated and enters into the slag and the Fe/SiO_2 mass ratio of the slag increases. The variation trends of the Fe_3O_4 and FeO contents in the slag with the content of Cu in the matte are given in Fig. 11 and are compared with the values reported by Wang and Zhang for the flash smelting process [30]. The changing tendencies of the Fe_3O_4 content in the SKS process and the flash smelting process are similar. As the matte grade increases but is no more than 56%, the content of FeO in slag increases slightly because more FeS in the matte is oxidized into slag. However, when the matte grade is greater above 56%, because the oxygen potential in the SKS furnace is sufficiently high to oxidize more FeO in the slag to Fe_3O_4, the FeO content of the slag decreases. The Fe_3O_4 content of the slag increases continuously, which results in an increase of the solid fraction of slag and the viscosity of the slag [14]. This behavior reveals that the Cu content of the matte should be optimized to enhance the direct recovery rate of copper in the smelting system, which could also reduce the burden of the grinding and flotation cleaning process of the copper slag afterward.

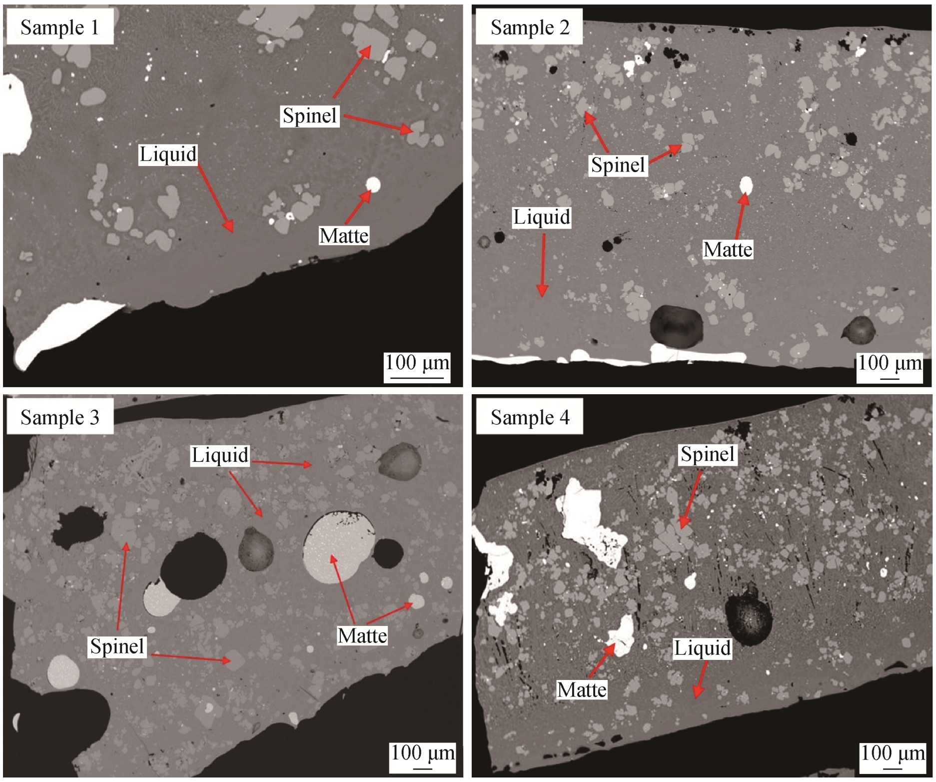


Fig. 10. Microstructures of quenched slag sample.

Table 5. Compositions of the quenched slag samples wt%

Sample No.	Cu (total)	Cu (dissolved)	Cu (entrained)	FeO	Fe_3O_4
Sample 1	2.42	0.22	2.20	32.41	23.67
Sample 2	2.88	0.31	2.65	32.10	25.18
Sample 3	3.21	0.35	2.81	31.21	30.55
Sample 4	3.55	0.54	3.01	28.73	31.24

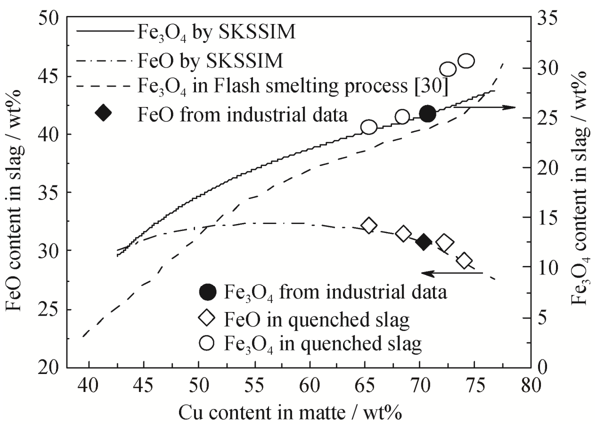


Fig. 11. Contents of FeO and Fe_3O_4 in SKS smelting slag.

4. Conclusions

In the SKS smelting process, the Cu loss to slag is one of the most concerning issues and is divided into two parts: (1) Cu^+ ions dissolved into slag in the form of Cu_2O or Cu_2S and (2) matte droplets entrained in slag. The dissolved and entrained Cu loss to SKS slag account for 10%–20% and 80%–90% of the total Cu loss, respectively. The matte grade strongly influences the Cu losses in the smelting slag. The mass ratio of Fe/SiO_2 of the smelting slag and the total Cu losses in the slag increase with increasing Cu content in the matte. When the matte grade is greater than 68%, the content of Cu in the slag increases much more dramatically. As the matte grade increases but is 56% or less, the content of FeO in the slag increases slightly. However, when the matte grade is greater than 56%, the content of FeO decreases. The content of Fe_3O_4 in the slag continuously increases, which increases both the solid fraction of molten liquid smelting slag and its viscosity. To improve the direct recovery rate of Cu in the bottom-blowing copper smelting system, the content of Cu in the matte should be less than 75wt%.

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