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Distribution and Control of Arsenic during Copper Converting and Refining

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Abstract: Arsenic content in copper concentrates is continuously increasing worldwide. It is desirable to remove arsenic from copper in the earlier stages of copper making due to the deposition of arsenic to cathode copper during the electrorefining process. Effects of temperature, flux, and oxygen on the distribution of arsenic during copper converting and fire refining processes were studied using FactSage 8.2. The results showed that arsenic can be effectively removed by proper selection of converting and refining slags. The decrease in Fe/SiO_2 or Fe/CaO ratio in the converting slag is favorable for arsenic distributed to slag. CaO is more effective than SiO_2 in decreasing the liquidus temperature of the slag and arsenic content in the blister copper during the converting process. Na_2O or CaO as a flux is effective to remove arsenic in the fire refining process.

Keywords: arsenic distribution; arsenic control; FactSage; blister copper; anode copper

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1. Introduction

Most copper is extracted from concentrates from pyrometallurgical processes which include smelting, converting, and fire and electro refining steps to obtain high-purity cathode copper [1,2]. The Outokumpu flash furnace and bath smelting furnaces are widely used in copper matte smelting processes [3]. The Peirce–Smith batch converting process dominates the production of blister copper from matte. However, the Peirce–Smith batch converting process has the disadvantages of low SO_2 utilization, gas fugitive emissions, and discontinuous operation. The flash converting and bath converting technologies have been developed to overcome the disadvantages [4,5]. Copper fire refining is mostly carried out in a rotary refining furnace, blowing gas, such as O_2, CO_2, and N_2, for the removal of sulfur and adding reductants for the removal of oxygen [6,7].

Arsenic is a harmful impurity present in copper concentrates [8]. In copper smelting, converting, and fire refining processes, arsenic can be present in the condensed phases and gas [8]. Arsenic-containing materials in the off gas is usually collected in the form of dust [9]. In the condensed phases, the arsenic can be distributed between matte and slag in the smelting process, between the blister copper and slag in the converting process, and between the anode copper and slag in the fire refining process [10]. With the high-quality copper concentrates exhausted worldwide, the content of arsenic in the copper concentrates is increasing continuously [11]. If arsenic is not handled properly in copper production, it will cause difficulty for copper extraction and endanger the environment [12,13]. Arsenic from anode copper can accumulate in the electrolyte during copper electrorefining. Deposit of the arsenic on the cathode copper significantly affects its quality and production rate during the copper electrorefining [14]. Removal of arsenic before the electrorefining process is important to produce high-quality cathode copper.

Swinbourne and Kho [15] studied the distribution of arsenic during copper flash converting using HSC Chemistry 7.1 software. It was found that the distribution of ar-

senic among copper, slag, and waste gas depends on their thermodynamic properties. Swinbourne and Kho reported that one-half of arsenic is distributed in blister copper due to the arsenic activity coefficient being lower than in slag. Li et al. [16,17] studied the distribution behavior of arsenic during copper flash converting with the multi-phase equilibrium mathematical model. It was found that arsenic is mainly present in blister copper (51.4%) and slag (41.9%). All of these thermodynamics studies only used properties of pure substances and did not consider solution phases. Yuan and Liu [18] and Liu [19] studied the distribution of arsenic in the bottom-blowing smelting and converting processes. It was found that increase in SiO_2 in slag was beneficial to reduce the arsenic in blister copper. Guo et al. [20] established a mass balance model and substance flow charts of arsenic in the bottom-blowing smelting and Peirce–Smith converting processes. It was found that arsenic mainly existed in both smelting and converting dusts. Zhou et al. [21] investigated the distribution of arsenic in ISA smelting, Peirce–Smith converting, and rotary refining processes in detail. It was found that the addition of alkaline fluxes was efficient to remove the arsenic from blister copper during the refining process. Kucharski [6] studied the effect of Na_2CO_3 on the removal of arsenic from blister copper during the refining process. It was found that arsenic from the blister copper reacted with Na_2CO_3 to produce Na_3AsO_4. Hidayat [22] summarized the experimental research of Kaur et al. [23], Nagamori and Mackey [24], and Avarmaa et al. [25], and determined the distributions of Arsenic between liquid copper and silicate slag at tridymite saturation. It was found that arsenic preferentially dissolved into copper instead of slag. Moreover, the arsenic in the slag was increased with increasing the oxygen partial pressure.

Most of the studies focused on the distribution of arsenic between different phases. Few studies discussed the control of arsenic by slag in copper-converting and -refining processes. Iron silicate is the major component of copper-smelting slags with variable Fe/SiO_2 ratios. It is not considered to use slag to control arsenic distribution in the smelting process. However, different fluxes can be used in the converting and fire refining processes. It is possible to evaluate the ability of different slags for the removal of arsenic from blister copper and anode copper. Recently updated FactSage 8.2 with extensive slag and alloy solution phases are used to investigate the optimal conditions to fix the arsenic in the copper-converting and -refining slags [26]. This study aims to understand the distribution of arsenic in matte converting and blister copper refining from a thermodynamics aspect.

2. Thermodynamic Predictions

FactSage 8.2 software was widely used in pyrometallurgical processing of different minerals [27–29]. Propper selection of the databases and phases is important to obtain reliable results. In the present study, the databases selected included "FactPs", "Ftmisc", and "Ftoxide". The solution phases selected in the smelting of copper concentrate included "FTmisc-PYRRB", "Ftmisc-MeS2", "Ftmisc-BCCS", "Ftmisc-FCCS", "Ftoxid-SLAGA", "Ftoxid-SPINA", and "FToxid-Oliv". The solution phases selected in the matte converting included "FTmisc-CuLQ", "FTmisc-FeCu", "FToxid-SLAGA", "FToxid-SPINA", "FToxid-MeO", and "FToxid-Oliv". The solution phases selected in fire refining of blister copper included "FTmisc-CuLQ", "FToxid-SLAGA", "FToxid-SPINA", and "FToxid-MeO."

3. Results and Discussion

3.1. Converting of Copper Matte

Peirce–Smith converter is still the major converting technology in copper smelters due to its advantages of easy operation and high adaptability of complex feeds. However, the P–S converter has the disadvantages of low SO_2 concentration, gas fugitive emissions, and discontinuous operation. Several continuous converting technologies have been developed in recent years, including flash converting and bottom-blowing converting processes. These new technologies have good automation and more efficient capture and utilization of SO_2. The published data on arsenic distribution on different technologies during converting are shown in Table 1. It can be seen from the table that more arsenic turns to the gas

phase in the P–S converting process because of the strong stirring. In the condensed phases, arsenic tends to stay in the blister copper with a small proportion of removal by the slag. Guo et al. [20] simulated the distribution of arsenic in the converting process. They indicated that up to 77% could go to gas with less than 10% arsenic left in the blister. It seems that all plant data are far away from the simulation. Comparing with the P–S converter, flash-converting and bottom-blowing-converting furnaces retain more than 95% arsenic in the condensed phases where higher arsenic enters the blister.

Table 1. The published data on arsenic distribution during converting.

Authors	Technology	Distribution (wt%)		
		Blister Copper	Slag	Gas
Li [16]	Flash	52	42	6
Swinbourne [15]	Flash	60	37	3
Liu [19]	Bottom blowing	65.7	31.4	2.9
Guo [20]	Peirce-smith	12.9	9.4	77.7
Zhou [21]	Peirce-smith	39.0	17.0	44.0
Zhang [30]	Peirce-smith	28.0	13.0	58.0
Wang [31]	Peirce-smith	66.4	5.4	28.2
Wang [32]	Peirce-smith	59.9	5.1	34.9
Vogt J [33]	Peirce-smith	50.0	32.0	18.0

Typical composition of copper concentrate, matte, and blister copper from a smelting plant were used in the present study as shown in Table 2 [21]. A total of 50% copper in matte was produced in the smelting stage and the slag had Fe/SiO_2 1.2. The slag temperature was 1200 °C and O_2 content in the gas was 22.83%. It can be seen from Table 2 that the arsenic content in the concentrate, matte, and blister copper was 0.30, 0.37, and 0.43%, respectively.

Table 2. Typical compositions of copper concentrate, matte, and blister copper.

	Cu	Fe	S	As	O	Ca	Al_2O_3	CaO	MgO
Concentrate	29.50	32.60	31.80	0.30	0.00	0.00	2.60	2.30	0.90
Matte	50.01	23.70	24.45	0.37	1.18	0.28	0.00	0.00	0.00
Blister copper	98.50	0.00	0.73	0.43	0.34	0.00	0.00	0.00	0.00

Matte converting was conducted at 1290 °C with a conventional slag containing Fe/SiO_2 ratio of 1.7. The effect of copper content in blister copper on arsenic distribution during the converting is shown in Figure 1. Oxygen partial pressures during the converting are also shown in the figure. Note that Figure 1 only shows the end of the converting stage where the matte phase has been converted to the blister copper. It can be seen that the Po_2 increases with increasing copper content in the blister. At the same time, the arsenic increases in the slag and decreases in the blister. According to the thermodynamic predictions, arsenic in the gas phase is neglected as shown in Figure 1. It seems that the converting reactions in the flashing and bottom furnaces approached equilibrium as indicated in Table 1.

Figure 2 shows the phase proportion as a function of copper content in blister copper under the same conditions as Figure 1. It can be seen that the proportion of gas remains constant when the Cu in the blister is increased from 97 to 99%. However, the proportion of the slag increases slowly initially and then rapidly with increasing copper content in the blister. In contrast, the proportion of the blister decreases slowly initially and then rapidly with increasing copper content in the blister. The reason is that high Cu in blister corresponds to high oxygen partial pressure as shown in Figure 1. Cu in the blister is oxidized to Cu_2O which is dissolved into the slag resulting in copper loss in the converting process. Arsenic in the blister can be reduced from 63 to 20% when the Cu in the blister is increased from 97 to 99% according to Figure 1. However, it can be seen from Figure 2 that

the proportion of the blister is reduced from 30 to 20%. The direct recovery of copper in the converting process is decreased by 33% if a low-As and high-Cu blister is produced. More effort will be required to recover the copper from the converting slag and the productivity of the converting furnace is significantly affected. It is a choice of the operator to balance the purity and productivity of the blister during the converting process.

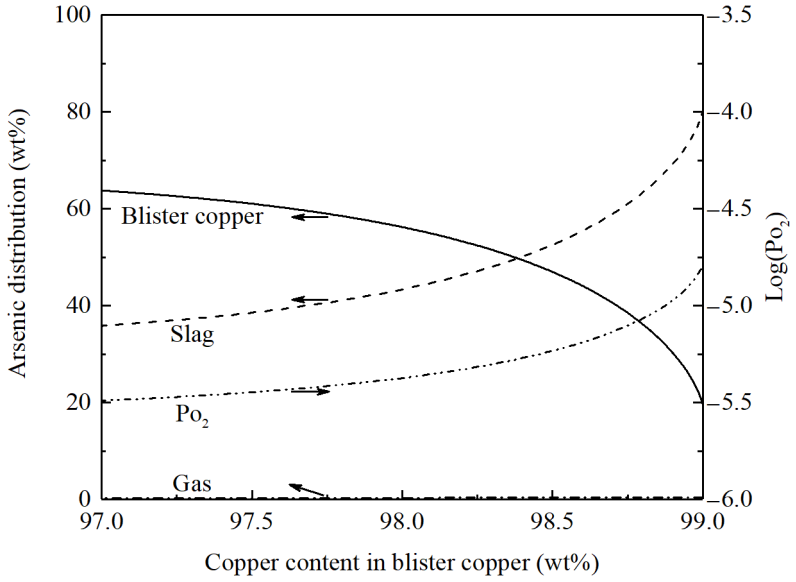


Figure 1. Effect of copper content in blister copper on arsenic distribution at 1290 °C with a slag Fe/SiO_2 ratio of 1.7.

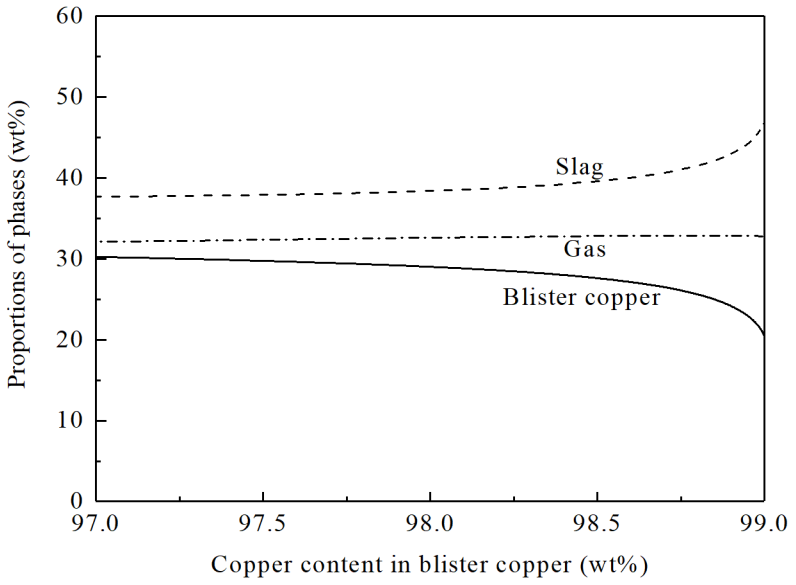


Figure 2. Effect of copper content in blister copper on proportions of the phases at 1290 °C with a slag Fe/SiO_2 ratio of 1.7.

Figure 3 shows the effect of temperature on arsenic distribution during converting at a slag Fe/SiO_2 1.7 and 98.5% Cu in blister copper. Little arsenic is present in the gas phase. The arsenic is decreased slightly in the slag and increased slightly in the blister with increasing temperature. Thermodynamically, a high converting temperature is not beneficial to remove the arsenic from the blister.

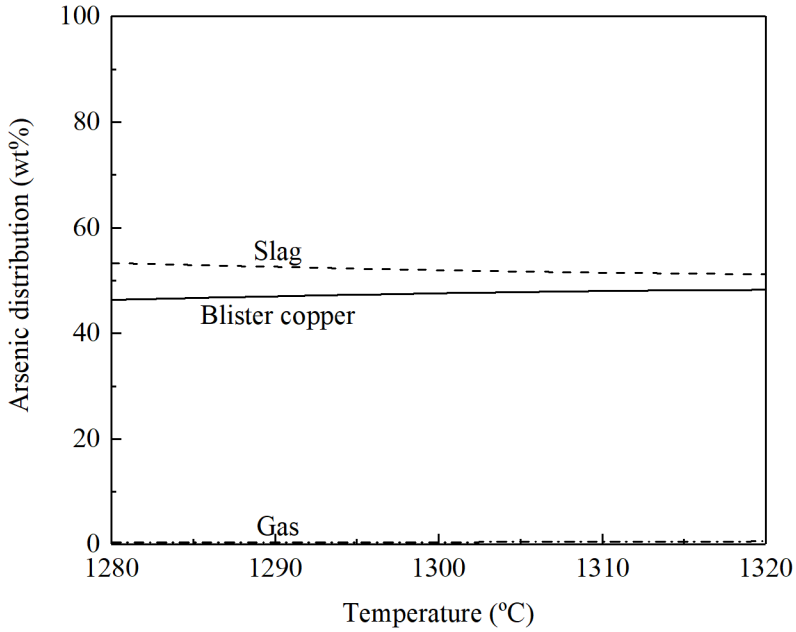


Figure 3. Effect of temperature on arsenic distribution at slag Fe/SiO_2 1.7 and 98.5% Cu in blister copper.

Both SiO_2 and CaO can be used in the copper converting process as a flux. The primary role of a flux is to form a liquid slag with the impurities. Effects of SiO_2 and CaO on liquidus temperature of the converting slag are shown in Figure 4 at a fixed copper content 98.5% in blister copper. Fe/SiO_2 or Fe/CaO ratio is usually used in the operation to control the addition of the flux and the composition of the converting slag. Figure 4 shows that, when SiO_2 is used as a flux, spinel is the only primary phase in the Fe/SiO_2 ratios of 1.5 to 4.0. The liquidus temperatures increase with increasing Fe/SiO_2 ratio. $Ca_2Fe_2O_5$ and spinel are the primary phases when CaO is used as a flux. In the $Ca_2Fe_2O_5$ primary phase field, liquidus temperatures decrease with increasing the Fe/CaO ratio. On the other hand, the spinel is the primary phase when the Fe/CaO ratio is hgher than 2.65. The liquidus temperatures in the spinel primary phase field increase with increasing the Fe/CaO ratio in the slag. It is obvious that CaO is a more efficient flux to lower the liquidus temperature of the converting slag. Much less CaO is required to control the liquidus temperature. As a result, more scrap can be treated in the converting furnace.

Figure 5 illustrates the arsenic distribution as a function of Fe/SiO_2 or Fe/CaO ratio at 1290 °C and copper content in blister copper 98.5%. In both cases the arsenic in the gas phase is neglected. The arsenic always increases in the slag and decreases in the blister copper with decreasing Fe/SiO_2 or Fe/CaO ratio. This means that both SiO_2 and CaO can fix the arsenic in the slag and decrease arsenic in the blister copper. Comparing with SiO_2, more arsenic can be fixed in the slag by adding CaO. Fe/SiO_2 and Fe/CaO are usually controlled between 1–2 and 3–4, respectively, in the converting process [34]. It can be seen from Figures 4 and 5 that, the liquidus temperature is 1264 °C and 52% arsenic is dissolved in the blister copper when the Fe/SiO_2 ratio is 1.5. In contrast, the liquidus temperature is 1217 °C and only 34% arsenic is dissolved in the blister copper when the Fe/CaO ratio is 3.0.

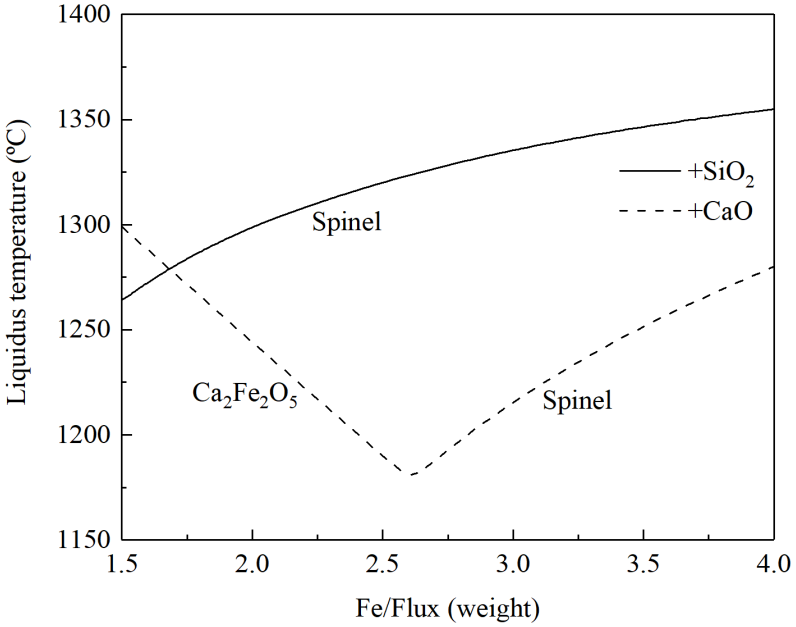


Figure 4. Effect of SiO_2 or CaO on liquidus temperatures of converting slag at a fixed copper content 98.5% in blister copper.

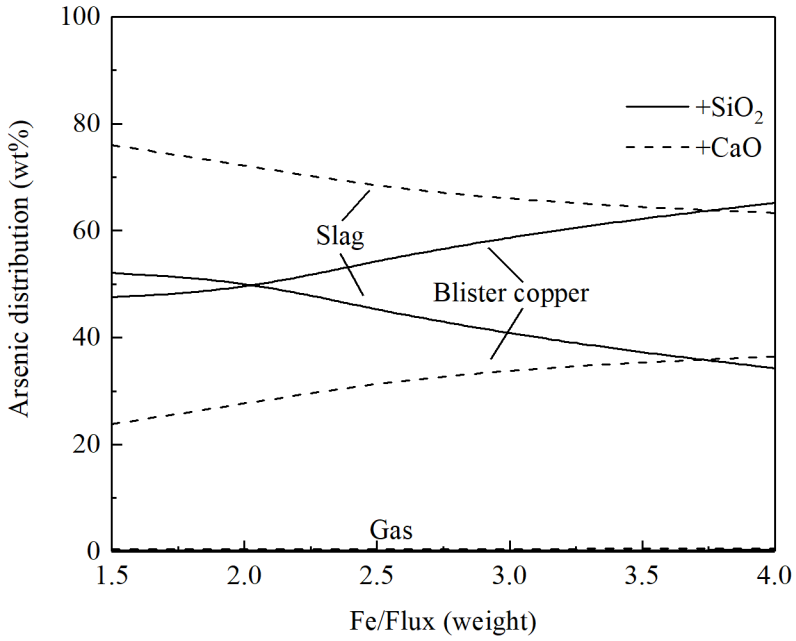


Figure 5. Effect of Fe/Flux ratio on arsenic distribution at 1290 °C and 98.5% Cu in the blister copper.

3.2. Fire refining of Blister Copper

After the converting process, blister copper containing 98.5% Cu is taken as an example for refining. It can be seen from Table 2 that arsenic, sulfur, and oxygen are the major impurities in blister copper and their concentrations are 0.43%, 0.73%, and 0.34%, respectively.

All blister copper needs to be fire refined to produce anode copper for electrorefining. The purpose of fire refining is to produce liquid copper which can be cast with minimal gas porosity, since sulfur and oxygen can disrupt the downstream electrorefining process. The conventional fire refining process usually includes an oxidation stage to remove sulfur and a reduction stage to remove oxygen. It can be seen from Figure 6 that in the oxidation stage, sulfur content in the anode copper decreases rapidly at the beginning of oxygen

blowing and then slowly after the sulfur content is lower than 0.1%. On the other hand, oxygen content in the anode copper increases slowly with increasing oxygen blowing. However, continuous blowing of oxygen will increase the oxygen in copper quickly, which will require more reductant and time to remove oxygen in the reduction stage.

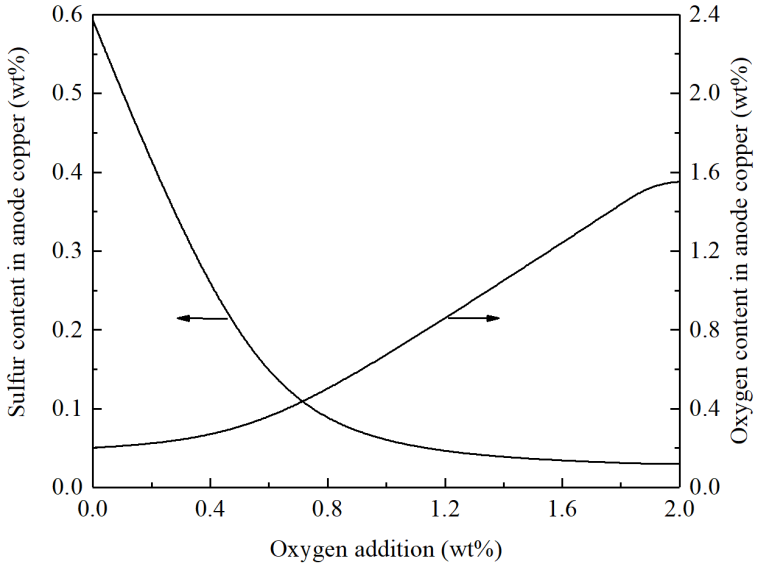


Figure 6. Effect of O_2/blister on sulfur and oxygen contents in anode copper at 1200 °C.

With increased arsenic content in the copper concentrates, it can be seen from Tables 1 and 2 that arsenic cannot be completely removed in the smelting and converting process. Arsenic in the anode copper will be brought into the electrorefining process which will affect the quality of the cathode copper and the efficiency of the process. The electrolyte needs to be purified frequently which will reduce the productivity of the electrorefining process. Flux is not usually used in the conventional fire refining process because the removal of sulfur and oxygen does not require a flux. It can be seen from Table 3 that over 80% arsenic is present in the anode copper after fire refining [20,21]. When Na_2CO_3 was used as a flux, the arsenic in the anode copper can be reduced to 1% [6]. It is therefore necessary to add a flux in the fire refining process to control arsenic in the anode copper.

Table 3. The published data on arsenic distribution during fire refining.

Authors	Flux	Mass Fraction (%)		
		Copper Anode	Slag	Gas
Zhou [21]	-	83.5%	-	-
Guo [20]	-	96.9	3.1	-
Kucharski [6]	Na_2CO_3	1	-	-

It can be seen from Figure 7 that if only oxygen is used in the refining process, arsenic in the anode copper is almost constant. In contrast, the addition of Na_2O or CaO can significantly reduce arsenic in the anode copper. Na_2O is more effective than CaO for removing arsenic from the copper.

The main reactions between arsenic and flux (CaO or Na_2O) are

$$3CaO + 2[As] + 5[O] = Ca_3(AsO_4)_2 \quad (1)$$

$$3Na_2O + 2[As] + 5[O] = 2Na_3AsO_4 \quad (2)$$

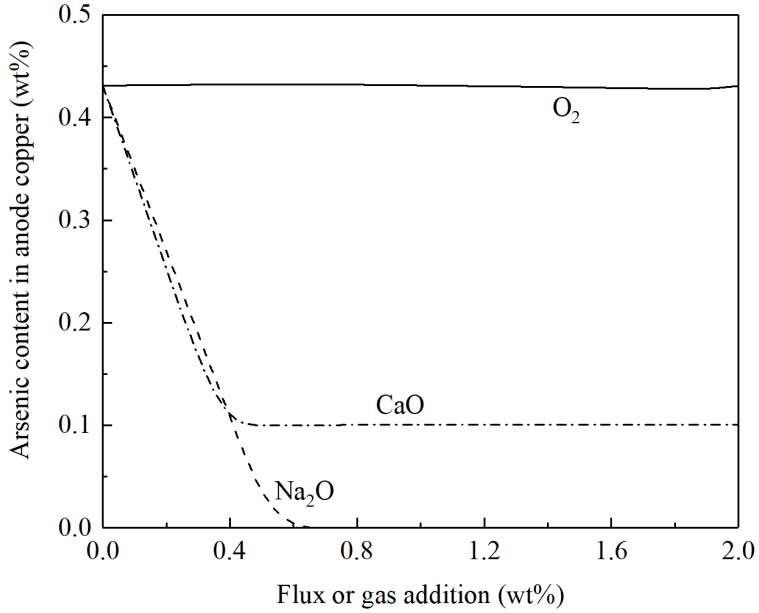


Figure 7. Effect of flux and oxygen on arsenic content in anode copper at 1200 °C.

It can be seen from the reactions that oxygen plays an important role in the removal of arsenic. Figure 8 shows the effect of oxygen on arsenic content in anode copper with 0.5% flux addition. It can be seen that oxygen does not affect the arsenic content in anode copper when Na_2O is a flux. However, the arsenic content in anode copper decreases significantly with increasing oxygen/blister ratio. When the oxygen addition is higher than 0.6%, CaO is a more effective flux than Na_2O to remove arsenic. When the oxygen addition is lower than 0.6%, it can be seen from Figure 6 that 0.15–0.6% S is present in the copper, which has a competing reaction with As to react with CaO. The addition of more oxygen removes sulfur from copper to enable the arsenic to react with CaO. On the other hand, the same weight of Na_2O has less mole% than CaO due to its higher molecular mass. Na_2O can react with both of sulfur and arsenic in the copper, which consumed all of the 0.5% Na_2O added without oxygen addition. More CaO in mole can remove more arsenic from copper when sulfur in copper is low.

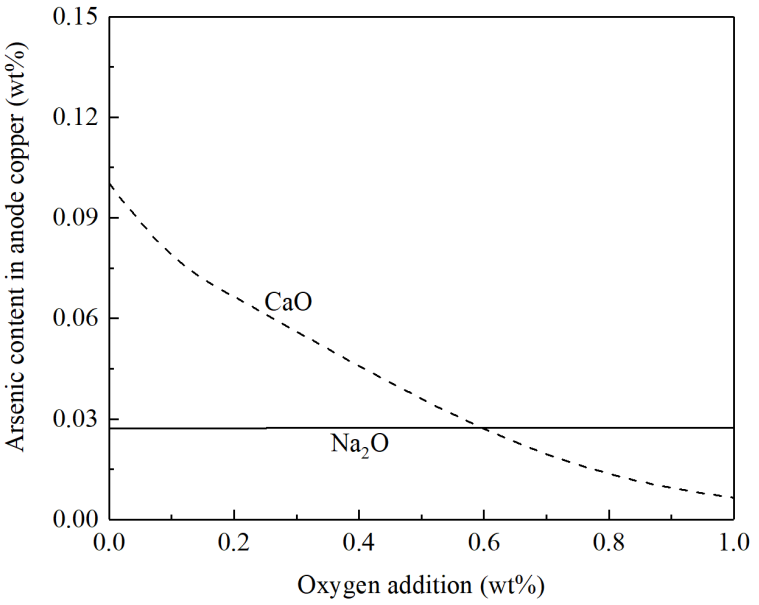


Figure 8. Effect of O_2/blister on arsenic content in anode copper with 0.5% flux/blister.

Effect of temperature on arsenic content in anode copper is shown in Figure 9 with 0.2% flux addition. It can be seen that at lower temperature, the arsenic content in anode copper can be reduced with increasing the temperature. The effect of temperature on arsenic content in anode copper is not significant at high temperatures and high flux additions.

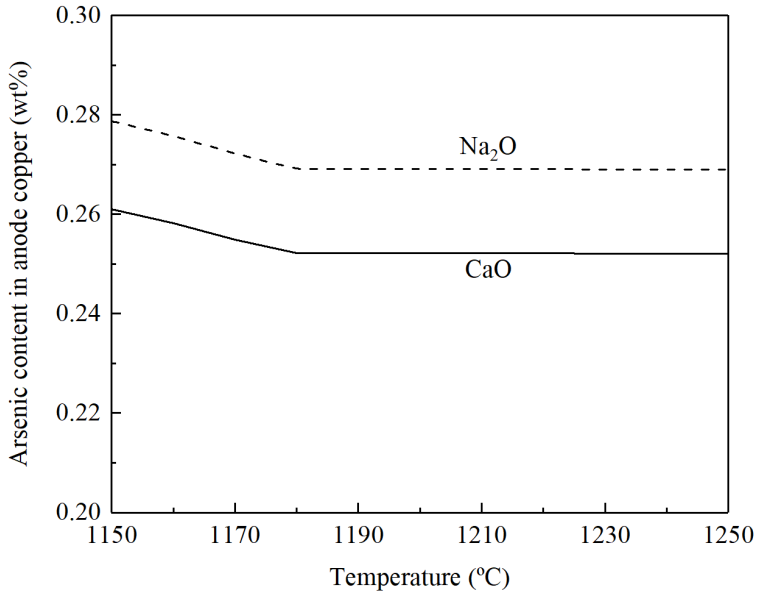


Figure 9. Effect of temperature on arsenic content in anode copper with 1.5% O_2/blister and 0.2% flux/blister.

Figure 10 shows the effect of flux addition on arsenic distribution at 1200 °C with O_2 addition 1.5%. It can be seen that CaO is a better flux than Na_2O in the removal of arsenic if oxygen is added which is the condition of oxidation stage in the fire refining process. With 0.55% CaO or Na_2O addition, over 99% arsenic brought from blister can be removed in the slag phase and a clean anode copper is obtained. The molecular mass of Na_2O (61.98) is larger than the molecular mass of CaO (56.08). When the same weight of Na_2O or CaO is added to the copper refining furnace with sufficient oxygen, CaO is a better flux than Na_2O to remove arsenic from the anode copper.

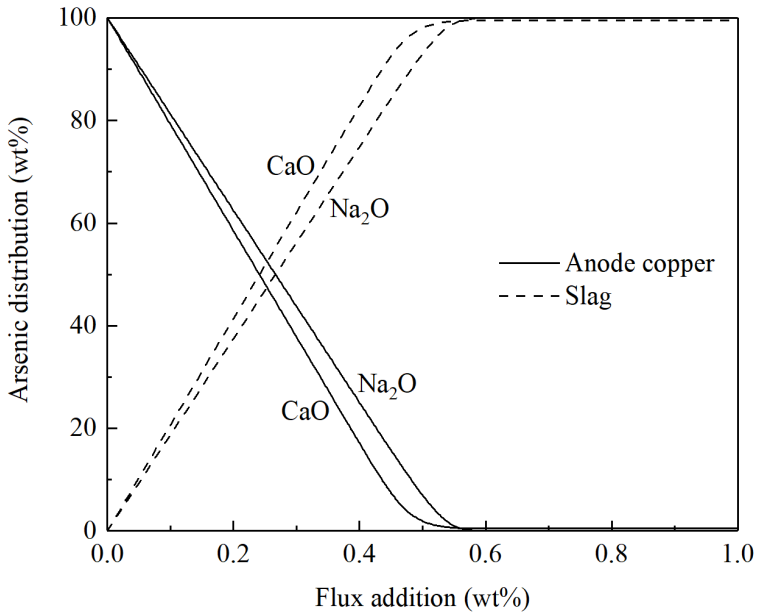


Figure 10. Effect of flux on arsenic distribution with 1.5% O_2/blister at 1200 °C.

4. Conclusions

Control of arsenic in the copper production is becoming to be a more and more important issue. Most of the studies were focused on the smelting process. The distribution and control of arsenic during copper converting and refining have been investigated by a review of the industrial data and thermodynamic calculations. Conventional converting and fire refining processes cannot remove arsenic efficiently. CaO is found to be a more effect flux in decreasing the liquidus temperature of the slag and arsenic content in the blister copper during the converting process. Both CaO and Na_2O can fix arsenic in the slag during the fire refining process. When oxygen is present, CaO is a better flux than Na_2O to remove arsenic from copper. Arsenic in anode copper can be slightly decreased with increasing temperature when the flux is added. More oxygen is beneficial for the removal of arsenic from anode copper with CaO addition.

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