Sb distribution in the phases of SiO\(_2\) saturated Sb-Fe-O-SiO\(_2\)-CaO system in air

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Abstract
Sb distribution in the phases of SiO\(_2\) saturated Sb-Fe-O-SiO\(_2\)-CaO system has been determined for the first time through high-temperature experiment and quenching techniques, followed by Electron probe X-ray microanalysis (EPMA) in air (P\(_{\text{tot}}\) = 1 atm, P\(_{O_2}\) = 0.21 atm). The phases were quantified in the temperature range of 900 °C-1200 °C and the effects of Fe/SiO\(_2\) (mass fraction) and CaO/SiO\(_2\) (mass fraction) on Sb\(_2\)O\(_3\) content in the Sb-Fe-O-SiO\(_2\)-CaO system were investigated at 1200 °C. The results indicate that the Sb-containing phase primarily existed in the solid solution phase at 1200 °C. With the increase of temperature from 1100 °C to 1200 °C, the Sb\(_2\)O\(_3\) content in the solid solution phase increased drastically from 7.52 wt% to 17.36 wt%. Lowering the values of CaO/SiO\(_2\) and Fe/SiO\(_2\) in the smelting process effectively reduced Sb\(_2\)O\(_3\) content in the slag. The verification experiment results suggest that the antimony content in slag was 0.57 wt%, the crude antimony yield rate was lower than 4%, and the crude antimony grade was beyond 94 wt%, which can achieve the reduction of antimony content in the slag.

Keywords: Phase relations; Liquidus; Microstructure; Pyrometallurgy

1. Introduction
Antimony is considered an important strategic resource that is widely used in the production of ceramics, fire retardants, medicaments and alloys [1]. Currently, antimony sulfide concentrate is the main raw material for the antimony production. The metallic antimony is primarily produced via different techniques, such as the antimony blast furnace and volatilization bath smelting processes [2-4]. Due to the shortage of antimony resources and increase in amount of secondary raw materials in metallurgical industry, the production processes urgently need to be re-designed or optimized [5]. Regarding the raw material composition during smelting, the Sb-Fe-O-SiO\(_2\)-CaO system is widely adopted in the pyrometallurgical processing for antimony extraction. Phase relations of the Sb-Fe-O-SiO\(_2\)-CaO system are significant in the development of new flow sheets and reduction of operating costs. Also, the data of antimony content in Sb-Fe-O-SiO\(_2\)-CaO system is important to limit the antimony content in the slag predictions and process philosophy improvement. However, the
experimental phase relations regarding this system are still ambiguous.

To develop accurate descriptions of the high-order systems, the phase relations of simple binary and ternary systems should be investigated. The phase relations of the CaO-SiO$_2$, FeO-Fe$_2$O$_3$-SiO$_2$, and FeO$_x$-CaO-SiO$_2$ subsystems have been systematically studied by several researchers using high-temperature experiments and rapid quenching technique [6-13]. The phase relations of FeO-Fe$_2$O$_3$-SiO$_2$ system in equilibrium with O$_2$ pressures ranging from $10^{10.9}$ to 1 atm have been studied and two ternary invariant points were determined in this system. The phases of tridymite (SiO$_2$), fayalite (2FeO·SiO$_2$), magnetite (FeO·Fe$_2$O$_3$), a liquid with composition 35 wt% SiO$_2$, 11 wt% Fe$_2$O$_3$, and 54 wt% FeO coexisted in equilibrium at 1140 °C, while the phases of fayalite (2FeO·SiO$_2$), magnetite (FeO·Fe$_2$O$_3$), wüstite with a composition corresponding to 35 wt% Fe$_2$O$_3$, a liquid with composition 22 wt% SiO$_2$, 14 wt% Fe$_2$O$_3$, and 64 wt% FeO coexisted in equilibrium at 1150 °C [7]. The phase relations in the FeO$_x$-CaO-SiO$_2$ system at intermediate oxygen partial pressures and the temperatures ranging from 1200 °C to 1350 °C were analyzed via equilibration/rapid-quenching/Electron probe X-ray microanalysis [8]. As per the above-mentioned works, the high-temperature experiment and rapid quenching technique is one of the most precise and flexible technique employed to obtain accurate experimental phase relations for systems with “sluggish” phase change [14,15].

Inspired by the above approach, herein, Sb distribution in the phases of SiO$_2$ saturated Sb-Fe-O-SiO$_2$-CaO system in air was investigated via the high-temperature experiment and quenching techniques, followed by EPMA and the verification experiments. The work would provide novel experimental data for the fundamental research.

2. Experiments
2.1. Preparation of initial mixtures

In the high-temperature experiments, chemically pure calcium oxide powder (CaO), silicon dioxide powder (SiO$_2$), antimony trioxide powder (Sb$_2$O$_3$), and ferrous oxalate dehydrate powder (FeC$_2$O$_4$·2H$_2$O) were employed as raw materials. The powders were dried at 373 K, weighed in desired proportions, mixed thoroughly in an agate mortar, and then, flaked at 5 MPa with a 9 mm-diameter pressing tool. The total weight of each flake was 0.15 g. The thin flakes have advantage in the molten phase quenching. The flakes reacted at a fixed temperature for 4 h. Thereafter, flakes were quenched in ice water, dried and analyzed.

2.2. Experimental apparatus and technique

The experiments were carried out in a horizontal laboratory tube furnace with silicon carbide heating elements (Figure 1). The experiments were conducted with the left and right gates of the reaction tube open to ambient atmosphere ($P_{tot} = 1$ atm, $P_{O2} = 0.21$ atm). Firstly, the specimen was placed into a quartz crucible. Subsequently, the quartz crucible containing the specimen was placed into an alumina ark. Then, the alumina ark was pushed into the uniformly hot temperature zone of the furnace with a steel rod. The temperature of the uniformly hot zone was measured using a calibrated thermocouple. The furnace temperature accuracy was ±2 K.

To avoid the effect of the quartz crucible on the experimental results, a spacer that had the same composition as the specimen was placed at the bottom of the quartz crucible. The specimen was taken out and quenched rapidly into the ice water at the end of the reaction time. It was consequently dried and inlaid into the resin. The specimen was analyzed after it was prepared by pre-grinding and polishing techniques.
2.3. Analysis

The microstructures and phase compositions were examined via Electron probe X-ray microanalysis (EPMA-1720H) using the cross section of the polished samples. Cross sections of polished samples were also characterized using X-ray diffraction analysis (Cu Kα, λ=0.154056 nm; Rigaku-TTR III).

3. Results and discussion

3.1. Chemical compositions and microstructures of samples

The compositions of the initial mixtures are listed in Table 1. All of the well-quenched samples were carefully examined and analyzed and each sample consisted of four homogeneous phases. The compositions of the phases were measured using EPMA. At least five independent measuring points from each phase were analyzed with EPMA for each sample, taken from their different well-quenched parts to quantify the chemical composition. The obtained EPMA results of the high-temperature experiments of SiO₂ saturated Sb-Fe-O-SiO₂-CaO system in the temperature range from 900 °C to 1200 °C in air are listed in Table 2.

Four phases were found in the samples reacted within the temperature range from 900 °C to 1200 °C. As listed in Table 2, the Sb-containing phase primarily existed in the solid solution phase at 1200 °C. When the temperature dropped to 1100 °C, the silicate phase began to appear. The solid solution phase vanished and Sb-silicate phase appeared at the temperature of 900 °C and 1000 °C. Moreover, as the temperature increased, the Sb₂O₃ content in solid solution phase increased dramatically, from 7.52 wt% at 1100 °C to 17.36 wt% at 1200 °C. The possible reason for the phenomenon is that high temperature is beneficial for the increase of Sb₂O₃ solubility in the solid solution phase. The Sb₂O₃ content in SiO₂ and High T.Fe phase displayed a stable tendency within the temperature range of 900 °C to 1200 °C, wherein, the average contents were approximately 1.3 wt% and 3.3 wt%, respectively. Also, the Sb-containing phase primarily changed from the solid solution phase to the Sb-silicate phase at the temperatures below 1100 °C. The Sb₂O₃ contents in the Sb-silicate phases were 48.32 wt% and 54.32 wt% at 1000 °C and 900 °C, respectively, indicating
that as the temperature decreased during the reaction, the Sb-containing phases primarily existed in the Sb-silicate phases.

Selected microstructures of the quenched samples from the polished sections are presented in Fig. 2. The Sb-containing phase mainly existed in the off-white block area (solid solution phase) at 1200 °C. Subsequently, when the reaction temperature was decreased from 1200 °C to 1100 °C, the Sb-containing phase (solid solution phase) changed from the off-white block to strip structure. Finally, at 1000 °C and 900 °C, the Sb-containing phase (the Sb-silicate phase) gathered into bright white block structure from strip structure.

Figure 3 illustrates the XRD patterns of the selected samples reacting at 900 °C-1200 °C. As presented in Fig. 3, the phases of Fe₃O₄, SiO₂ and Fe₂SiO₄ were determined at 1200 °C. The phases of Fe₂O₃, SiO₂, Fe₂SiO₄, and Ca₃Fe₂Si₃O₁₂ were decided at 1100 °C. The phases of Fe₂O₃, SiO₂, Fe₂SiO₄ and CaFeSi₂O₆ were detected at 1000 °C and 900 °C. Therefore, it can be inferred from the aforementioned analysis that the phases of Fe₂O₃ and Fe₂SiO₄ corresponded to the High T.Fe phase at 900 °C-1200 °C, and the phase of Ca₃Fe₂Si₃O₁₂ corresponded to the silicate phase at 1100 °C (Fig. 2(b)). Notably, the Sb-containing phase mainly existed in the solid solution phase and Sb-silicate at the temperature of 1100 °C-1200 °C and 900 °C-1000 °C, respectively. However, the Sb-containing phases were not detected in the XRD patterns, may be due to low Sb content in the slag system.

**Table 1. The composition of the initial mixtures (wt%)**

|     | FeO  | SiO₂ | CaO  | Sb₂O₃ |
|-----|------|------|------|-------|
|     | 28.66| 53.07| 13.27| 5.00  |

**Table 2. Phase composition of SiO₂ saturated Sb-Fe₂O₃-SiO₂-CaO system at 900-1200 °C in air**

| No. | T/ °C | Time/ h | Phase             | EPMA (wt%) | FeO | CaO | SiO₂ | Sb₂O₃ |
|-----|-------|---------|-------------------|------------|-----|-----|------|-------|
| 1   | 1200  | 4       | Solid solution    |            | 28.60| 24.65 | 29.39 | 17.36 |
|     |       |         | Liquid            |            | 20.99| 22.37 | 53.47 | 3.17  |
|     |       |         | SiO₂              |            | 5.29 | 2.79  | 90.67 | 1.25  |
|     |       |         | High T.Fe phase   |            | 75.49| 2.60  | 18.88 | 3.03  |
| 2   | 1100  | 4       | Solid solution    |            | 23.71| 26.82 | 41.94 | 7.52  |
|     |       |         | SiO₂              |            | 7.00 | 3.03  | 88.75 | 1.22  |
|     |       |         | High T.Fe phase   |            | 75.34| 2.93  | 18.94 | 2.79  |
|     |       |         | Silicate          |            | 9.75 | 38.91 | 51.34 | 0.00  |
| 3   | 1000  | 4       | Silicate          |            | 17.14| 30.80 | 52.05 | 0.00  |
|     |       |         | SiO₂              |            | 5.99 | 3.43  | 89.22 | 1.36  |
|     |       |         | High T.Fe phase   |            | 78.36| 2.36  | 15.80 | 3.48  |
|     |       |         | Sb-silicate       |            | 11.97| 21.03 | 18.68 | 48.32 |
| 4   | 900   | 4       | Silicate          |            | 15.90| 29.80 | 50.97 | 3.33  |
|                  | SiO₂   | Fe₂O₃ | SiO₂ | CaO  |
|------------------|--------|-------|------|------|
| High T. Fe phase | 4.76   | 3.01  | 90.81| 1.42 |
| Sb-silicate      | 79.36  | 2.23  | 14.43| 3.98 |

**Figure 2.** BSE micrographs of selected quenched and polished Sb-Fe-O-SiO₂-CaO samples of the present study at: (a) 1200 °C; (b) 1100 °C; (c) 1000 °C, and (d) 900 °C. (Fe/SiO₂ 0.42, CaO/SiO₂ 0.25)

**Figure 3.** The XRD patterns of selected samples reacted at: (a) 1200 °C; (b) 1100 °C; (c) 1000 °C, and (d)
3.2. Effect of Fe/SiO$_2$ and CaO/SiO$_2$ on Sb$_2$O$_3$ content in Sb-Fe-O-SiO$_2$-CaO system

The selection of slag type during smelting has an important impact on Sb$_2$O$_3$ content in the slag. To explore the change of Sb$_2$O$_3$ content during the reaction, the effect of Fe/SiO$_2$ and CaO/SiO$_2$ on the Sb$_2$O$_3$ content were investigated for the current system. Selected slag phase compositions are listed in Table 3.

The selected samples were carefully analyzed and the phase compositions were measured using EPMA. The obtained EPMA results of SiO$_2$ saturated Sb-Fe-O-SiO$_2$-CaO system at 1200 °C in air with various values of CaO/SiO$_2$ and Fe/SiO$_2$ are listed in Table 3. The Sb-containing phase mainly existed in the solid solution phase at 1200 °C. The Sb$_2$O$_3$ content in the solid solution phase demonstrated a stable tendency within the content range from 14.45 wt% to 17.36 wt% with CaO/SiO$_2$ 0.84-1.03 and Fe/SiO$_2$ 0.76-0.97. Moreover, the Sb$_2$O$_3$ content in SiO$_2$ and High T.Fe phase also demonstrated a stable tendency at 1200 °C. The average contents were approximately 1.45 wt% and 3.1 wt%, respectively, which is consistent with the above results. Also, as presented in Table 3, Sb$_2$O$_3$ content in the liquid phase was lower than that in the solid solution phase within the content range from 0.13 wt% to 3.17 wt% with CaO/SiO$_2$ 0.42-0.50 and Fe/SiO$_2$ 0.31-0.39.

Figure 4 presents the BSE micrographs of the selected quenched and polished SiO$_2$ saturated Sb-Fe-O-SiO$_2$-CaO samples at 1200 °C in air with various values of CaO/SiO$_2$ and Fe/SiO$_2$. As shown in Fig. 4, the High T.Fe phase disappeared when the CaO/SiO$_2$ increased to 0.55 with Fe/SiO$_2$ fixed at 0.40. The possible reason for the phenomenon may be that as the CaO content increased, CaO combined with other phases to form a low melting phase in the liquid phase. In contrast, when the CaO/SiO$_2$ increased from 0.25 to 0.70 with Fe/SiO$_2$ fixed at 0.80, the phase composition only changed slightly. Therefore, the above analysis illustrates that lowering the values of CaO/SiO$_2$ and Fe/SiO$_2$ in the smelting process can effectively reduce Sb$_2$O$_3$ content in the slag.

### Table 3. Selected slag phase compositions in the present study (wt%)

| Slag No. | CaO/SiO$_2$ mass ratio | Fe/SiO$_2$ mass ratio | Composition/ wt% |
|---------|------------------------|----------------------|-----------------|
|         |                        |                      | FeO   | CaO   | SiO$_2$ | Sb$_2$O$_3$ |
| 1       | 0.25                   | 0.40                 | 28.66 | 13.27 | 53.07   | 5.00       |
| 2       | 0.25                   | 0.80                 | 42.22 | 10.56 | 42.22   | 5.00       |
| 3       | 0.40                   | 0.40                 | 26.44 | 19.59 | 48.97   | 5.00       |
| 4       | 0.40                   | 0.80                 | 39.58 | 15.83 | 39.58   | 5.00       |
| 5       | 0.55                   | 0.40                 | 24.55 | 25.00 | 45.45   | 5.00       |
| 6       | 0.55                   | 0.80                 | 37.25 | 20.49 | 37.25   | 5.00       |
| 7       | 0.70                   | 0.40                 | 22.90 | 29.69 | 42.41   | 5.00       |
| 8       | 0.70                   | 0.80                 | 35.19 | 24.63 | 35.19   | 5.00       |

Table 4. Phase composition of SiO$_2$ saturated Sb-Fe-O-SiO$_2$-CaO system at 1200 °C in air with various values of
| No. | Time/ h | Phase        | EPMA (wt%) | Fe/SiO₂ mass ratio | CaO/SiO₂ mass ratio |
|-----|---------|--------------|------------|--------------------|---------------------|
|     |         |              | FeO  | CaO  | SiO₂  | Sb₂O₃ |                |                |
| 1   | 4       | Solid solution | 28.60 | 24.65 | 29.39 | 17.36 | 0.76           | 0.84           |
|     |         | Liquid        | 20.99 | 22.37 | 53.47 | 3.17  | 0.31           | 0.42           |
|     |         | SiO₂          | 5.29  | 2.79  | 90.67 | 1.25  | /              | /              |
|     |         | High T.Fe phase | 75.49 | 2.60  | 18.88 | 3.03  | /              | /              |
| 2   | 4       | Solid solution | 31.71 | 23.60 | 29.27 | 15.42 | 0.84           | 0.81           |
|     |         | Liquid        | 24.51 | 22.44 | 51.77 | 1.28  | 0.37           | 0.43           |
|     |         | SiO₂          | 9.21  | 3.29  | 86.33 | 1.17  | /              | /              |
|     |         | High T.Fe phase | 79.53 | 2.27  | 14.60 | 3.60  | /              | /              |
| 3   | 4       | Solid solution | 28.91 | 25.51 | 29.46 | 16.12 | 0.76           | 0.87           |
|     |         | Liquid        | 22.05 | 23.97 | 53.01 | 0.96  | 0.32           | 0.45           |
|     |         | SiO₂          | 6.25  | 4.27  | 88.15 | 1.33  | /              | /              |
|     |         | High T.Fe phase | 76.34 | 3.65  | 17.01 | 3.00  | /              | /              |
| 4   | 4       | Solid solution | 30.56 | 24.70 | 30.29 | 14.45 | 0.78           | 0.82           |
|     |         | Liquid        | 24.36 | 23.67 | 51.26 | 0.71  | 0.37           | 0.46           |
|     |         | SiO₂          | 9.71  | 4.49  | 84.30 | 1.50  | /              | /              |
|     |         | High T.Fe phase | 79.44 | 3.46  | 14.14 | 2.96  | /              | /              |
| 5   | 4       | Solid solution | 29.04 | 26.77 | 28.12 | 16.07 | 0.80           | 0.95           |
|     |         | Liquid        | 23.71 | 25.59 | 50.33 | 0.37  | 0.37           | 0.51           |
|     |         | SiO₂          | 6.83  | 7.34  | 84.23 | 1.60  | /              | /              |
| 6   | 4       | Solid solution | 30.77 | 26.41 | 26.77 | 16.05 | 0.89           | 0.99           |
|     |         | Liquid        | 24.30 | 25.06 | 49.97 | 0.67  | 0.38           | 0.50           |
|     |         | SiO₂          | 6.96  | 5.47  | 85.69 | 1.88  | /              | /              |
|     |         | High T.Fe phase | 78.67 | 4.39  | 13.77 | 3.17  | /              | /              |
| 7   | 4       | Solid solution | 29.06 | 27.33 | 28.71 | 14.90 | 0.79           | 0.95           |
|     |         | Liquid        | 23.51 | 26.25 | 50.11 | 0.13  | 0.36           | 0.52           |
|     |         | SiO₂          | 7.67  | 8.51  | 82.68 | 1.14  | /              | /              |
| 8   | 4       | Solid solution | 31.68 | 26.25 | 25.48 | 16.59 | 0.97           | 1.03           |
|     |         | Liquid        | 25.04 | 24.80 | 49.45 | 0.71  | 0.39           | 0.50           |
|     |         | SiO₂          | 8.31  | 5.61  | 84.44 | 1.64  | /              | /              |
|     |         | High T.Fe phase | 78.99 | 4.70  | 13.46 | 2.85  | /              | /              |
Figure 4. BSE micrographs of selected quenched and polished Sb-Fe-O-SiO$_2$-CaO samples of the present study at 1200 °C

(a) CaO/SiO$_2$ 0.25, Fe/SiO$_2$ 0.40; (b) CaO/SiO$_2$ 0.25, Fe/SiO$_2$ 0.80;
(c) CaO/SiO$_2$ 0.40, Fe/SiO$_2$ 0.40; (d) CaO/SiO$_2$ 0.40, Fe/SiO$_2$ 0.80;
(e) CaO/SiO$_2$ 0.55, Fe/SiO$_2$ 0.40; (f) CaO/SiO$_2$ 0.55, Fe/SiO$_2$ 0.80;
(g) CaO/SiO$_2$ 0.70, Fe/SiO$_2$ 0.40; (h) CaO/SiO$_2$ 0.70, Fe/SiO$_2$ 0.80
3.3. Verification experiment

As per the foregoing theoretical analysis, the verification experiment on the bath smelting of antimony sulfide concentrate was performed at the smelting temperature of 1200 °C, oxygen concentration of 80%, gas pressure of 0.18 MPa, oxygen flow rate of 0.16 m³/h, slag composition of Fe/SiO₂: 0.4, CaO/SiO₂: 0.5, and reaction time of 40 min. The experimental results are listed in Table 5. The antimony content in slag was 0.57 wt%, which is consistent with the above analysis. Also, the crude antimony yield rate was lower than 4% and crude antimony grade was beyond 94 wt%, demonstrating that the antimony-containing phases were mainly volatilized into dust during smelting, which can achieve the reduction of antimony content in slag.

| Project                             | Test results |
|-------------------------------------|--------------|
| Antimony content in slag            | 0.57 wt%     |
| Crude antimony rate                 | 3.6%         |
| Crude antimony grade                | 94.83 wt%    |

4. Conclusion

(1) The effect of temperature on the phase compositions within the temperature range of 900 °C to 1200 °C was systematically investigated. The results suggest that the Sb-containing phase mainly existed in the solid solution phase at 1200 °C. At the temperature below 1100 °C, the Sb-containing phase primarily changed from the solid solution phase to the Sb-silicate phase.

(2) As the temperature increased, the Sb₂O₃ content in the solid solution phase increased drastically from 7.52 wt% at 1100 °C to 17.36 wt% at 1200 °C. The Sb₂O₃ content in SiO₂ and High T Fe phase demonstrated a stable tendency within the temperature range of 900 °C to 1200 °C, while the average contents were approximately 1.3 wt% and 3.3 wt%.

(3) The effects of Fe/SiO₂ and CaO/SiO₂ on the Sb₂O₃ content in the Sb-Fe-O-SiO₂-CaO system were explored. The results demonstrate that lowering the values of CaO/SiO₂ and Fe/SiO₂ in the smelting process can effectively reduce Sb₂O₃ content in the slag. The verification experiment results imply that the antimony content in slag was 0.57 wt%, which is consistent with the above theoretical analysis.

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