0,080 $\pm$0,01 for silica-saturated slags without fluxing additions. From Table 5.2 it appears that Fe_2O_3 is more susceptible to replacement by Al_2O_3 and FeO because it is less basic and the result is a decrease in the Fe^{3+}/Fe^{2+} ratio.

The result for this investigation can be compared with the work of Toguri and Santander (17). Toguri and Santander equilibrated a high silica slag in alumina crucibles at 1573° K over a range of partial pressures of oxygen and the resulting slag had an average alumina content of 6 mass per cent. The alloy and slag were equilibrated over a period of 48 hours and it is possible that for each run the slag was at alumina saturation. Assuming that the slags of Toguri and Santander were close to silica saturation and that the slag is saturated with 6 mass per cent Al_2O_3 at 8×10^{-9} atm oxygen the result obtained by least squares analysis

$$\text{mass \% Cu in slag} = 29{,}3\ a_{CuO_{0,5}} \qquad 5.13$$

indicates that alumina depresses the solubility of copper in slag. It is most likely that the slags of Toguri and Santander were not at silica saturation at the completion of each experiment and this might account for the lower value of the solubility of copper in slag compared to this investigation in which

$$\text{mass \% Cu in slag} = 34{,}18\ a_{CuO_{0,5}} \qquad 5.15$$

Bailey (16) showed that for silica-saturated slag at an oxygen partial pressure of 10^{-8} atm containing no fluxing additions and additions of 5 mass per cent Al_2O_3 and 10 mass per cent Al_2O_3

respectively

$$\text{mass \% Cu in slag} = 31{,}80\ a_{CuO_{0,5}} \quad \text{(flux-free)} \qquad 5.16$$

$$= 27{,}52\ a_{CuO_{0,5}} \quad (5\%\ Al_2O_3) \qquad 5.17$$

$$= 42{,}63\ a_{CuO_{0,5}} \quad (10\%\ Al_2O_3) \qquad 5.18$$

The result for the addition of 5 mass per cent Al_2O_3 shows a decrease in the solubility of copper compared with the solubility for alumina-free slag. The results for 10 mass per cent Al_2O_3 show a significant increase in the solubility of copper. Bailey used a run time of 20 hours with a stock slag showing a high degree of oxidation. This relatively short equilibration time and the highly oxidized slag could have given substantial errors. Figure 6 indicates that the values of copper solubility are too high at low oxygen potentials and too low at high oxygen potentials.

5.2.3 Lime

The solubility of copper in silica-saturated slag containing about 4,5 mass per cent, 7,5 mass per cent and 10,5 mass per cent lime is shown in Figure 30. Lime additions significantly decrease the solubility of copper in slag. At about 70 mass per cent copper in the alloy the copper content of the slag was decreased from 2,10 mass per cent for the silica-saturated slag without fluxing additions to values of 1,89 mass per cent, 1,64 mass per cent and 1,40 mass per cent for fluxing additions of 4,5 , 7,5

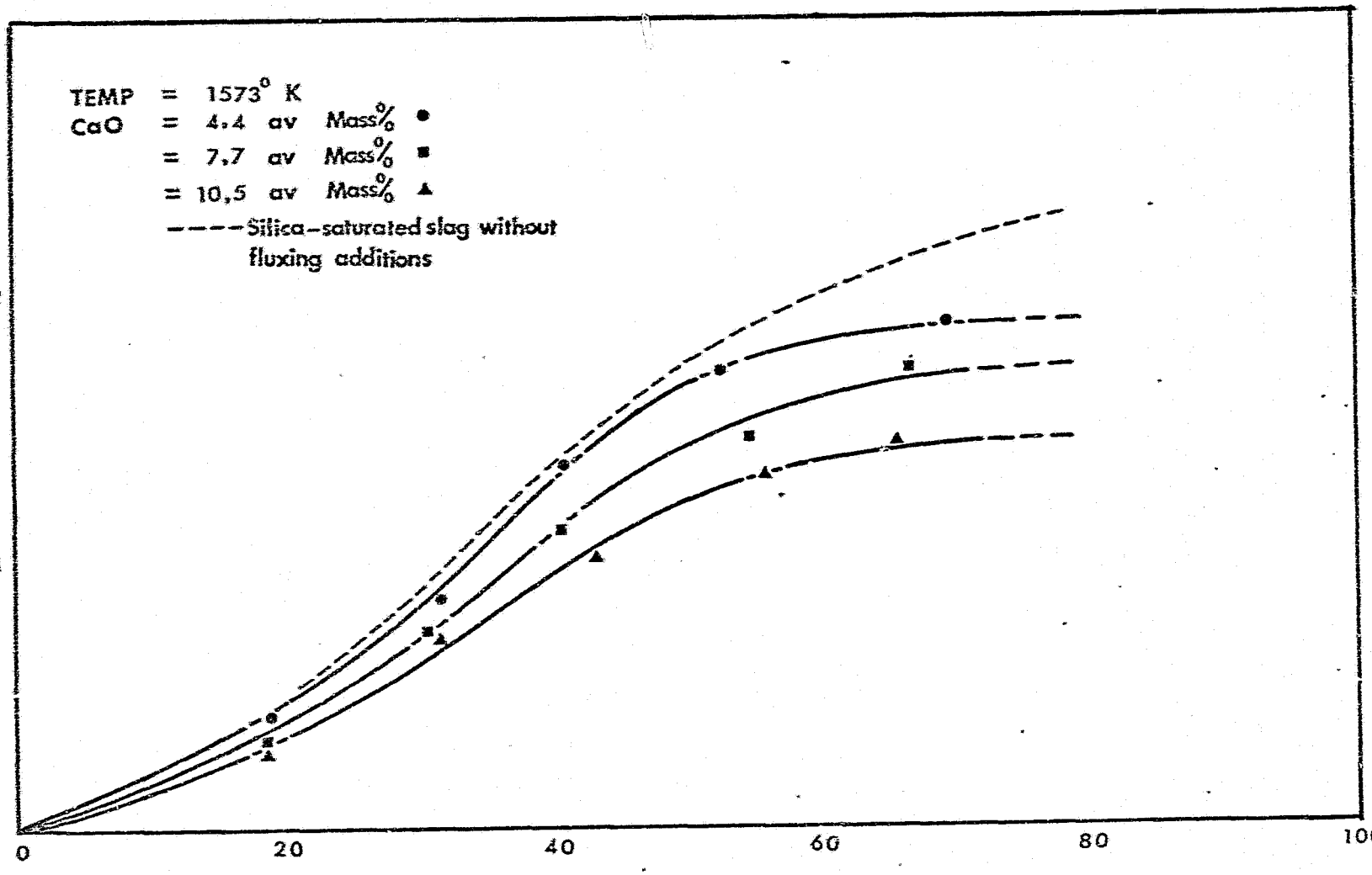

FIGURE 30 SOLUBILITY of COPPER in SILICA – SATURATED SLAG CONTAINING CaO

and 10,5 mass per cent CaO respectively.

The calculated results for the activities and activity coefficients of copper oxide are presented in Table 5.1 whilst the activity coefficient of copper oxide is plotted against the mole fraction $N_{CuO_{0,5}}$ in Figure 31. The figure shows that for successive increases in lime content of the slag there is an increase in the activity coefficient of copper oxide. The averages of the activity coefficients of copper oxide are respectively 2,86 (4,4 mass % CaO), 3,28 (7,7 mass % CaO) and 3,81 (10,5 mass % CaO).

Two points marked on Figure 31 with question marks appear to be inconsistent with the other results. The high values for these activity coefficients might possibly have been due to entrapment of alloy in the slag. However from the relation $\gamma = a/N$ it can be seen that when the amount of copper in the slag increases then the activity coefficient decreases. In both cases the gold content of the slag was below 80 ppm. Therefore, metallic entrapment is not the explanation for the high activity coefficient values. The copper content of the slag for these two points is low and therefore most susceptible to analytical error. It is considered that error in copper analysis at these low values resulted in low copper values and therefore anomously high activity coefficients of copper oxide. Figure 31.

The mass per cent copper for each addition of lime is plotted against the activity coefficient of copper oxide in Figure 32 and the following expressions were obtained

$$\text{mass \% Cu in slag} = 34,60\ a_{CuO_{0,5}} \quad \text{(CaO-free slag)} \quad 5.12$$

$$= 32,13\ a_{CuO_{0,5}} \quad \text{(4,5 mass \% CaO)} \quad 5.19$$

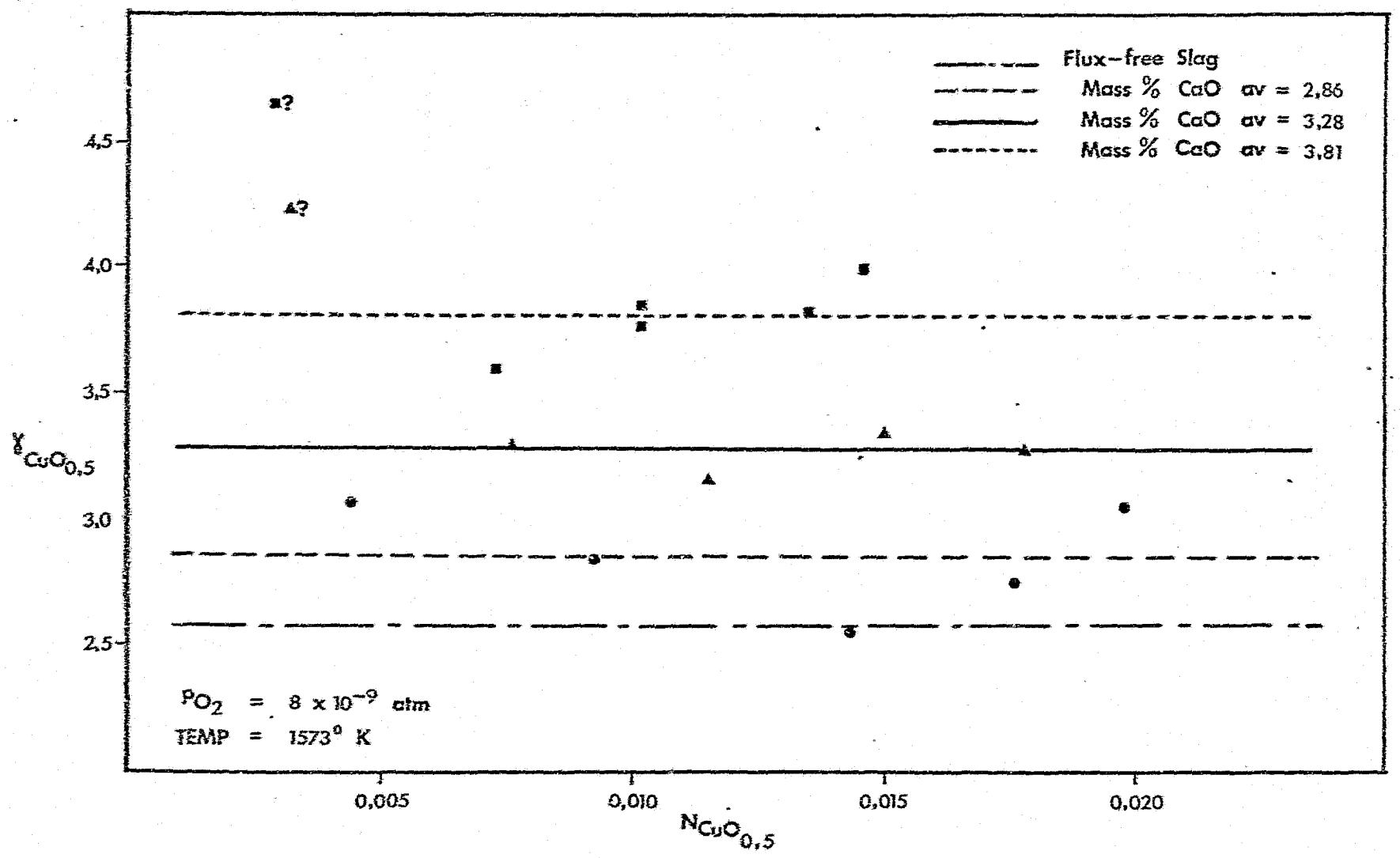

FIGURE 31 ACTIVITY COEFFICIENT of COPPER OXIDE as a FUNCTION of MOLE FRACTION of COPPER OXIDE in SILICA-SATURATED SLAG CONTAINING LIME

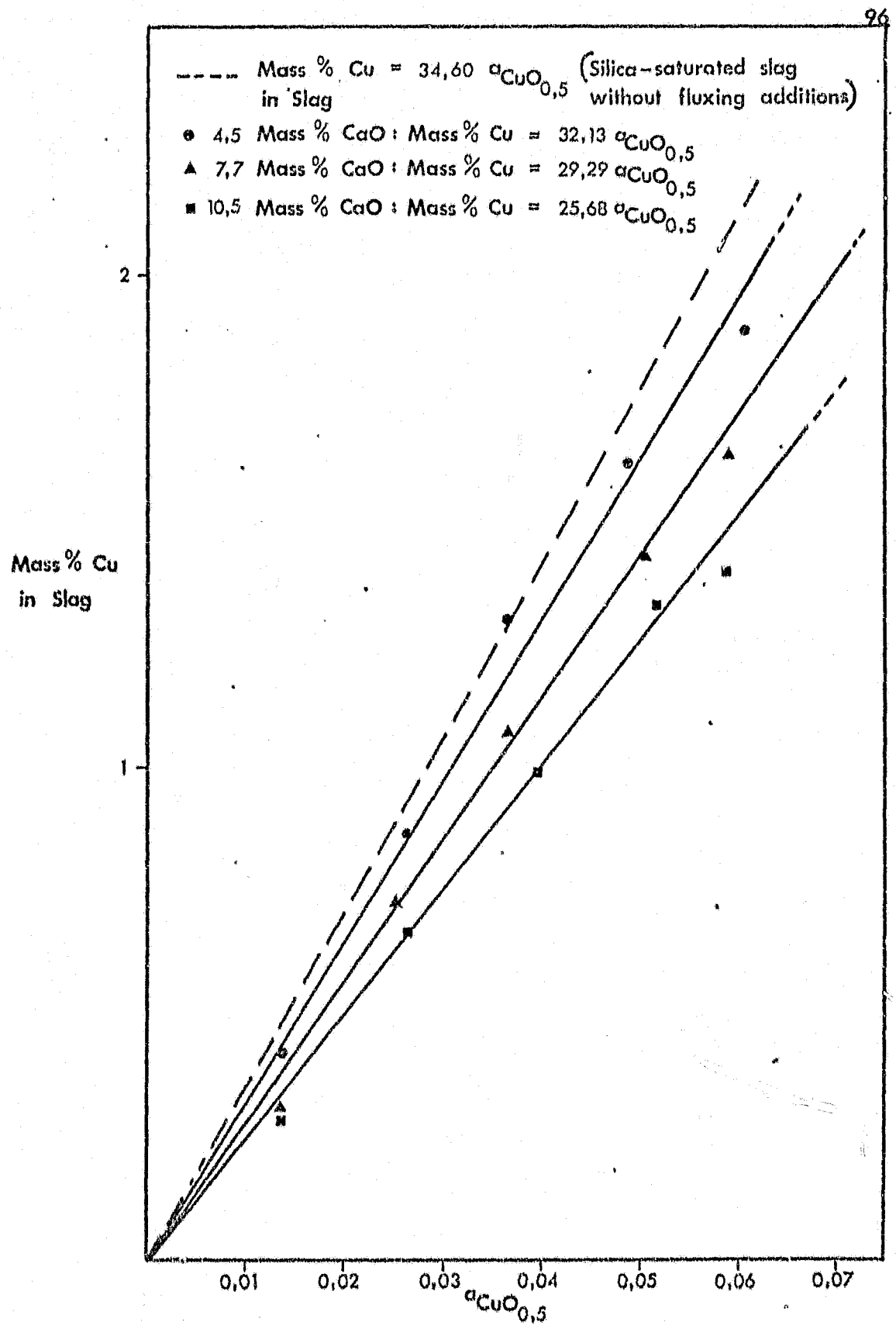

FIGURE 32 RELATIONSHIP BETWEEN MASS PER CENT COPPER and $a_{CuO_{0,5}}$ in SILICA-SATURATED SLAG CONTAINING 4,5 7,5 and 10,5 MASS PER CENT LIME at 1573° K

$$\text{mass \% Cu in slag} = 29{,}29\ a_{CuO_{0,5}} \quad (7{,}5 \text{ mass \% CaO}) \quad 5.20$$
$$= 25{,}68\ a_{CuO_{0,5}} \quad (10{,}5 \text{ mass \% CaO}) \quad 5.21$$

The increase in activity coefficient of copper oxide and decrease in the solubility of copper for successive increases in the lime content of the slag can again be explained in terms of the acid-base theory of slags. From Table 5.2 it can be seen that lime is more basic than either MgO or Al_2O_3 and hence lime additions can be expected to give a greater decrease in the solubility of copper oxide in the slag than either of these two oxides because of the greater replacement of copper cations with calcium cations in the silicate complex.

Lime also affected the FeO and Fe_2O_3 contents of the slag. For additions of 4,5 , 7,5 and 10,5 mass per cent CaO the Fe^{3+}/Fe^{2+} ratio was 0,068 $\pm$ 0,01 , 0,073 $\pm$ 0,01 and 0,070 $\pm$ 0,01. As expected, all three values are lower than the value of 0,080 $\pm$ 0,01 for silica-saturated slag. There was not a decrease in the ratio for increasing lime additions probably because of analytical difficulties with small samples.

5.3 Practical Implications of Results

In copper smelting it is intended that the copper content of the slag should be as low as possible especially when high slag to matte or slag to metal ratios are encountered. In order to minimise the copper content of slags it is important to maintain a relatively low viscosity in order to avoid entrapped particles of metal or matte in the

slag. The other important consideration is thermodynamic where an increase in the activity coefficient of copper oxide in the slag results in a lowering of the solubility of copper oxide in the slag.

This investigation shows that thermodynamically the copper content of a silica-saturated slag can be altered significantly by additions of MgO, Al_2O_3 and CaO. The additions of up to about 4 mass per cent MgO and 8 mass per cent Al_2O_3 has a slightly beneficial effect on copper losses to silica-saturated slags by raising $\gamma CuO_{0,5}$ from 2,58 to 2,83 and 2,75 respectively. The addition of lime has a more pronounced affect in lowering the copper solubility in slag. Additions of about 4,5 , 7,5 and 10,5 mass per cent CaO increased the activity coefficient from 2,58 for flux-free slag to 2,86 , 3,28 and 3,81 with respective decreases in the maximum copper solubility from 2,10 mass per cent to 1,89 , 1,64 and 1,40 respectively.

The results of the investigation are most relevant to reverberatory smelting since these slags are silica-saturated. The incorporation of MgO, Al_2O_3 and especially CaO in reverberatory slags is desirable to maximize $\gamma CuO_{0,5}$ provided the physical characteristics of the slag, particularly the viscosity and melting point, are not adversely altered.

6. FUTURE WORK

This outline of future work is only for silica-saturated slags.

1. In silica-saturated slags the oxygen potential of the slag at a particular oxygen partial pressure is reflected by the Fe^{3+}/Fe^{2+} ratio. When MgO, Al_2O_3 or CaO are added to silica-saturated slag at the same partial pressure of oxygen the Fe^{3+}/Fe^{2+} ratio is altered. It would be of interest to know the change in activities and activity coefficients of both FeO and Fe_2O_3 for additions of MgO, Al_2O_3 and CaO. To do this accurately more refined techniques are required for the analysis of FeO.

2. As an extension of this investigation, information on the effects of Al_2O_3 and CaO on copper solubility in silica-saturated slag up to the limits of solubility of Al_2O_3 and CaO would be of interest at a partial pressure of oxygen of 10^{-8} atm.

3. Little information is available about the effect of additions of MgO and CaO on copper solubility in silica-saturated slags over the range of partial pressures of oxygen from 10^{-11} to 10^{-6} atm. The information on the effect of alumina is conflicting and investigations at different partial pressures of oxygen for additions of MgO, Al_2O_3 and CaO are necessary to clarify the effect of these fluxing additions on the solubility of copper in silica-saturated iron silicate slags.

4. If the variation in the solubility of copper was established for single additions of MgO, Al_2O_3 and CaO over a range of oxygen potentials, the study could then be extended to investigate the effect of additions of more than one of these components on the solubility of copper in silica-saturated slags.

7. CONCLUSION

The effect of fluxing additions of MgO, Al_2O_3 and CaO on the solubility of copper in silica-saturated slag was investigated by varying the activity of copper in a copper-gold alloy equilibrated with a fayalite slag in a silica crucible at a partial pressure of oxygen of 8×10^{-9} atm and a temperature of 1573° K.

The results of this investigation confirm that copper is dissolved as $CuO_{0,5}$ in silica-saturated fayalite slag and the solubility of copper in this slag without any fluxing additions can be expressed by the relationship

$$\text{mass \% Cu in slag} = 34,60 \; a_{CuO_{0,5}}$$

for copper contents in the slag between 0 to 2,4 mass per cent. The average value of the activity coefficient of $CuO_{0,5}$ in silica-saturated slag without fluxing additions was 2,58.

The addition of 4 mass per cent MgO increased the activity coefficient of $CuO_{0,5}$ to 2,83 in the slag and slightly decreased the solubility of copper in silica-saturated slag at 1573° K. The expression obtained for copper solubility was

$$\text{mass \% Cu in slag} = 33,74 \; a_{CuO_{0,5}}$$

The addition of about 8 mass per cent Al_2O_3 also increased the activity coefficient of $CuO_{0,5}$ in the slag to 2,75 and slightly reduced the solubility of copper in the slag. The expression obtained for copper solubility was

$$\text{mass \% Cu in slag} = 34,18 \; a_{CuO_{0,5}}$$

Additions of lime (4,5 , 7,5 and 10,5 mass per cent) reduced the solubility of copper in the slag and increased the activity coefficient of $CuO_{0,5}$ to 2,86 , 3,28 and 3,81 respectively. The solubility of copper in slag for additions of about 4,5 , 7,5 and 10,5 mass per cent CaO respectively can be expressed by the following equations

$$\text{mass \% Cu in slag} = 32{,}13\ a_{CuO_{0,5}} \quad \text{(4,5 mass per cent CaO)}$$

$$= 29{,}29\ a_{CuO_{0,5}} \quad \text{(7,5 mass per cent CaO)}$$

$$= 25{,}68\ a_{CuO_{0,5}} \quad \text{(10,5 mass per cent CaO)}$$

At 10,5 mass per cent CaO in silica-saturated slag the copper solubility in slag is decreased by about 30 per cent.

The observed increase in the activity coefficient of $CuO_{0,5}$ with additions of MgO, Al_2O_3 and CaO results in an increase in the activity of copper oxide in the slag with a subsequent decrease in the solubility of copper in the slag.

The effect of the fluxing additions MgO, Al_2O_3 and CaO on the solubility of copper in silica-saturated slag and on the activity coefficients of $CuO_{0,5}$ can be explained by their relative basicities. In silica-saturated iron silicate slags both MgO and Al_2O_3 act as basic oxides. Their relative position on an acid-base classification table (Table 5.2) indicates that the effect of both fluxes on the solubility of copper in slag will not be particularly pronounced and indeed this is the case. Lime is more basic than Cu_2O and its effect on the solubility of copper in slag is far greater.

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9. APPENDIX

1. Flowrate Calibrations

The intimate mixing of carbon dioxide and carbon monoxide at elevated temperature provides an extremely small yet accurately controlled oxygen partial pressure through the equilibrium reaction.

$$CO + \tfrac{1}{2}O_2 = CO_2 \qquad A.1$$

for which the equilibrium constant is

$$\log K = \log \frac{P_{CO_2}}{P_{CO}\, P_{O_2}^{\frac{1}{2}}} \qquad A.2$$

The standard free energy for equation A.1 is given by

$$\Delta G^o = -RT\ln K \qquad A.3$$

$$\log K = -\Delta G^o/(RT)(2,303) \qquad A.4$$

Standard free energy data in calories for the reaction is available (42). Therefore,

$$\log K = \log \frac{P_{CO_2}}{P_{CO}\, P_{O_2}^{\frac{1}{2}}} \qquad A.5$$

$$= \frac{14550}{T} - 4,405 \qquad A.6$$

The above relationship can be expanded to a third order relationship as a more accurate function of temperature. However this is not necessary within the accuracy required. Table A.1 gives the CO and CO_2 flowrates used at 1573° K.

TABLE A.1 CALIBRATION TABLE FOR CO_2/CO FLOWRATES

P_{O_2} (atm)	P_{CO_2}/P_{CO}	CO (ml/min)	CO_2 (ml/min)
10^{-6}	69,98	10	700
10^{-7}	22,13	30	664
10^{-8}	6,998	50	350
10^{-9}	2,213	100	221,3
10^{-10}	0,70	300	210
10^{-11}	0,221	400	88,4
10^{-12}	0,07	400	28

Data from Table A.1 are plotted on Figure 33 as CO_2/CO ratio versus the partial pressure of oxygen. An accurate measurement of the CO and CO_2 flowrates at the settings used showed that the CO_2/CO ratio was 6,240 which corresponded to an oxygen partial pressure of $8,0 \times 10^{-9}$ atm.

2. Evaluation of Thermodynamic Data

For the reaction $CO + \frac{1}{2}O_2 = CO_2$ A.1

$$\Delta G^0 = a - b\,T \quad !$$

$$= -RT\ln K \qquad \text{A.3}$$

Thus from the data of Thompson (42)

$$R\ln K = \frac{66560}{T} - 20,15 \qquad \text{A.7}$$

and $$\log K = \frac{14550}{T} - 4,405 \qquad \text{A.6}$$

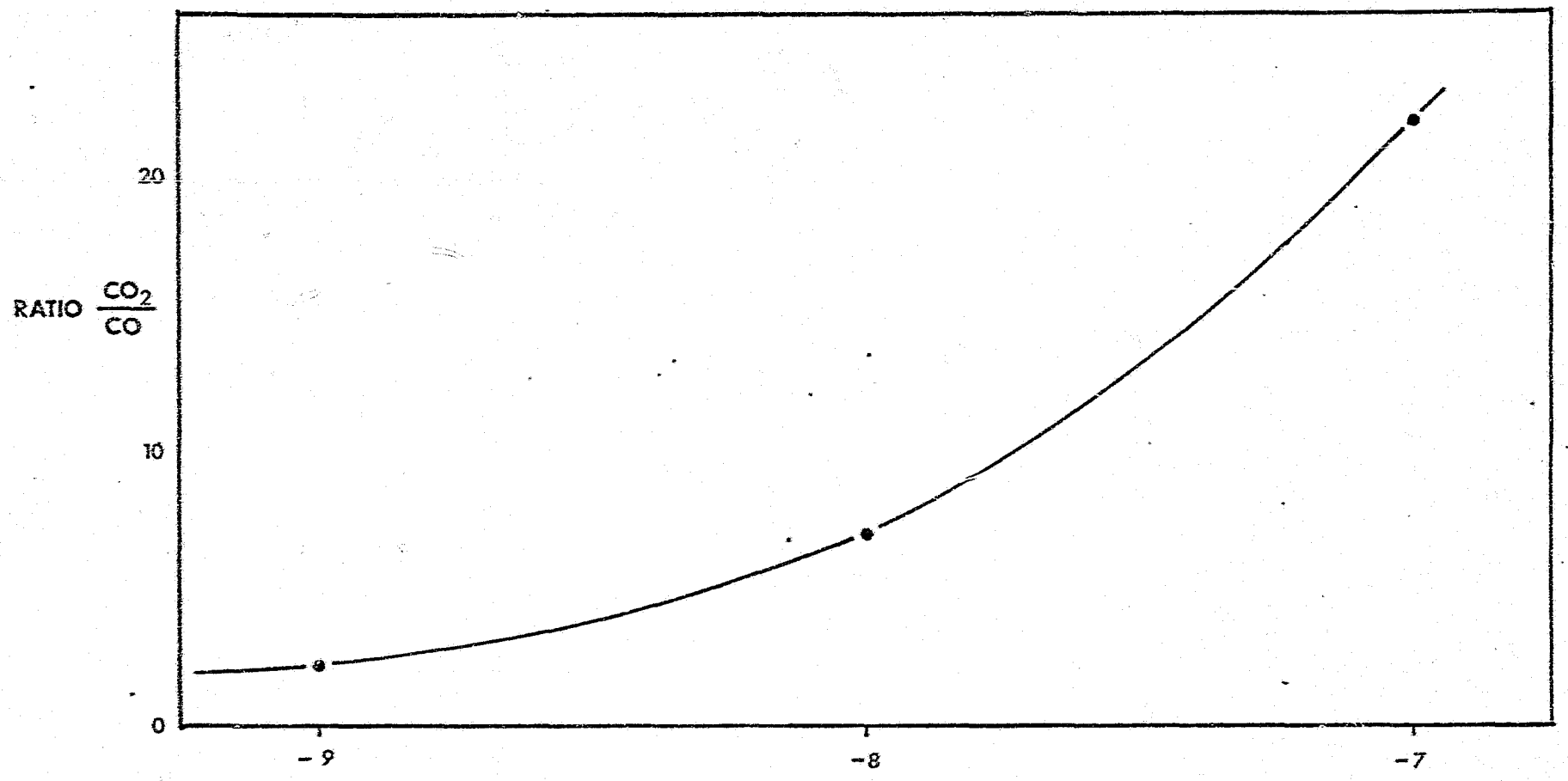

FIGURE 33 CALIBRATION CURVE for ESTIMATION of OXYGEN PARTIAL PRESSURE

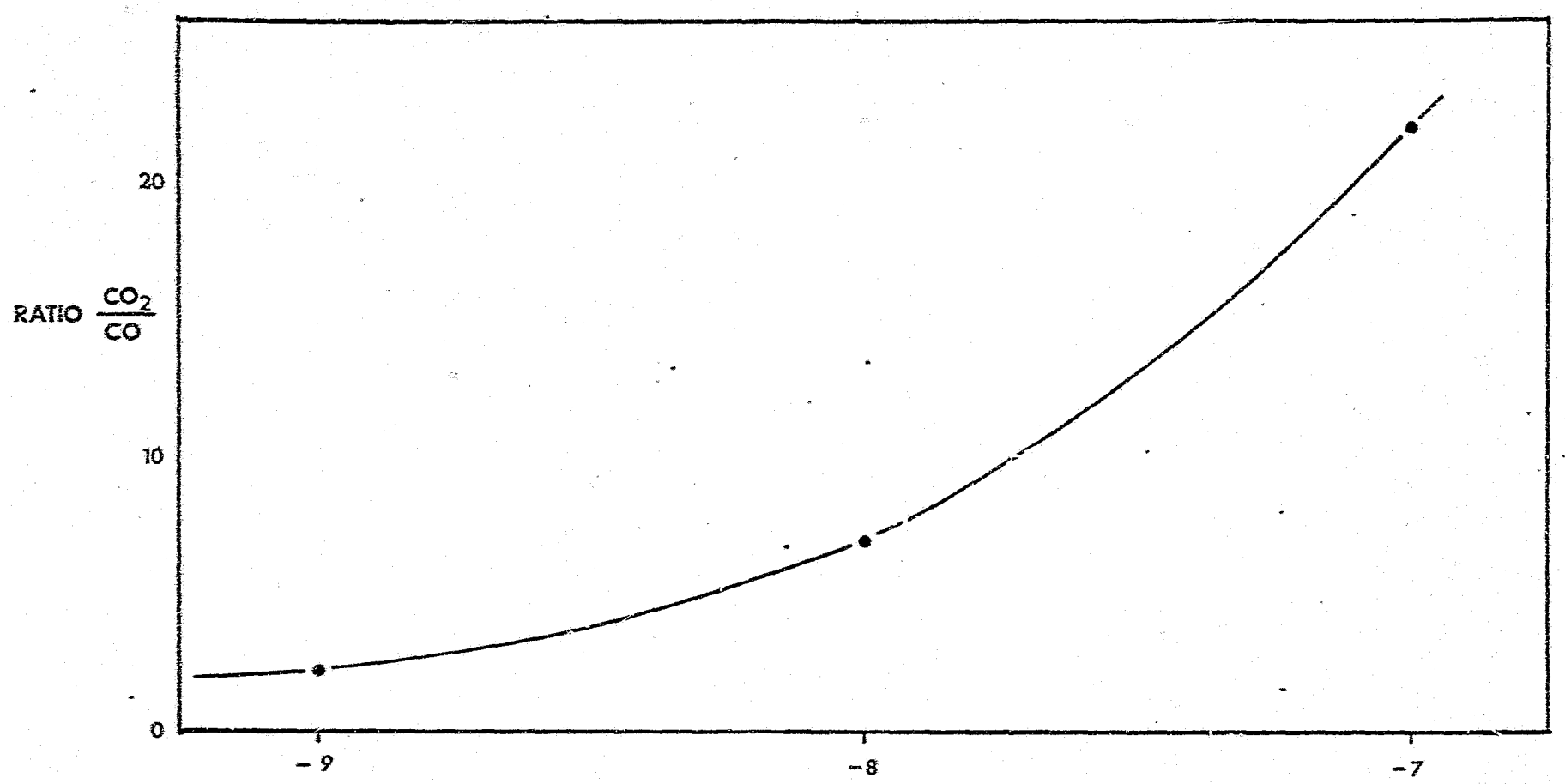

FIGURE 33 CALIBRATION CURVE for ESTIMATION of OXYGEN PARTIAL PRESSURE

from which $K = 6,995 \times 10^{-4}$ at 1573^{0} K

Compared to the value obtained by Kubaschewski (43)

$$K = 6,981 \times 10^{-4}$$

For the reaction

$$Cu(l) + \tfrac{1}{2}O_2\,(g) = CuO_{0,5}(l) \qquad A.8$$

The data of Elliott and Gleiser (44) gives

$$\Delta G^{0}(cal) = -17490 + 7,21\,T \qquad A.9$$

for which $K = 7,158$ at 1573^{0} K

From Mah (45) the following information was obtained.

$$2Cu(l) + \tfrac{1}{2}O_2 = 2CuO_{0,5}$$

$$\Delta G^{0}(cal) = -29260 + 10,488\,T \qquad A.10$$

for which $K = 59,36$ at 1573^{0} K

Thus the equilibrium constant for equation A.8 is

$$K = 7,7045$$