Thermodynamic Calculation of Reaction and Equilibrium between Coal and FeO-containing Slag in the Atmosphere of CO₂ Gas

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High temperature slags could act as not only heat carrier and catalyst but also reactants in some chemical reactions to produce syngas due to the individual components in slags. A new method is put forward to utilize the thermal energy of converter slags to generate CO gas which could improve the energy utilization efficiency. Thermodynamics of the reactions among coal, FeO–CaO–SiO₂ slag and gas was studied by thermodynamic calculation under steady temperature condition or no enthalpy change condition. The effects of FeO amount in initial slag, mass%CaO/mass%SiO₂ ratio of slag, and slag temperature on the behavior of the production of H₂ and CO gases were clarified.

KEY WORDS: thermodynamic calculation; equilibrium; FeO-containing slag; fuel gas; coal.

1. Introduction

The ironmaking and steelmaking process by blast furnace and basic oxygen converter furnace consumes considerable amount of energy and generates CO₂ in large quantity. 1–3) Under this situation, the steel industry faces a serious challenge to meet the target that global CO₂ emissions are cut to less than 50% of 2 000 levels by 2 050. 4) Therefore, it is urgent to reduce CO₂ emission from steel industries and develop environmental-friendly ironmaking and steelmaking processes. 5)

Nowadays China has been the largest steel producer and its annual crude steel output reached 823 Mt in 2014. 6) Accordingly, about 110 Mt converter slags were discharged. The total waste heat associating with these slags is estimated to be more than 1.76 × 10¹⁷ J with the slag enthalpy of 1.6 GJ per ton of slag. 7, 8) which corresponds to around 5.85 million ton of coal. As high temperature slags carry a substantial amount of high quality thermal energy, they could represent the largest undeveloped energy source in the steel industry. 9, 10)

High temperature slags could act as not only thermal media but also good catalyst, for promoting decomposition of coal. 11) Steelmaking slag contains from 15 to 30 mass% FeO, which could contribute to chemical reactions to produce syngas. The conventional method based on sensible heat storage is an indirect process that uses ceramics, water, etc., and requires an additional heat exchange. 12, 13) From the viewpoint of energy cascade utilization, it is important to use high temperature waste heat as a heat source in the chemical industry. 14, 15) Some processes have been identified to recover thermal energy from molten slag by water vapor, because of its huge potential. Matsuura et al. 16) have calculated the generation of H₂ gas by the reaction between FeO in steelmaking slag and steam, and it was found that the increase of introduced gas temperature, the ratio of mass%CaO to mass%SiO₂ and FeO content in slag could improve the productivity of H₂ gas. Sato et al. 17) conducted the laboratory-scale experiments to examine this reaction. It was indicated that at 1 723 K the increase of FeO content and the ratio between mass%CaO and mass%SiO₂ enhanced the production of H₂ gas, which was consistent with the calculated results.

The conventional gas-solid coal gasification has a series of drawbacks for both the plant and the environment. Akiyama et al. 18, 19) pointed that gasification of carbon was more suitable for the recycling of molten BF slag and developed a process to promote the endothermic reaction to produce H₂ and CO using BF slag waste heat. 20) Then Purwanto and Akiyama 21) or Maruoka et al. 22) carried out a large number of experiments, the exergy recovery rate of which reached 85%. Li et al. 23–26) proposed a new system using the BF slag waste heat to ensure the requirement of heat for coal gasification. According to their analysis, about 0.4 Mt tons of CO was generated by 1 Mt slag and the conversion efficiency from thermal to chemical reached 35%.

According to all the references, the utilization of BF slag provides a valuable reference to our study of steelmaking slag. On the basis of these facts related to CO₂ emission and steelmaking slag, the utilization of steelmaking slag as a catalyst and heat carrier for coal reforming to produce CO,
H₂ gases and carbonaceous materials is attractive to solve these problems. To develop the new slag utilization method, sufficient understanding of the chemical reactions among FeO-containing slag, coal and gas is essential.

In the present study, equilibrium among coal, FeO–CaO–SiO₂ slag and gas was studied by thermodynamic calculations in the condition of steady temperature or no enthalpy change. Also, the effects of slag composition and slag temperature on the producing behavior of H₂ and CO gases were clarified. The results indicate the possibility of realizing a process of producing CO from coal and steelmaking slag. Furthermore, the proposed system will contribute to promoting energy conservation in the steelmaking industry by reducing CO₂ gas emission in the atmosphere, which will be useful for mitigating the global warming.

2. Thermodynamic Calculation at Steady Temperature Condition

2.1. Calculation Conditions

A commercial thermodynamic calculation software FactSage 6.3 was applied to estimate the equilibrium state of the system. Table 1 shows the details of calculation conditions. Firstly, the initial condensed phases including molten slag and solid oxides were prepared by inputting oxide constituents with prescribed compositions and equilibrating at initial slag temperature. Subsequently, the prepared condensed phases were equilibrated with coal and CO₂ gas. The composition of coal used for calculation is shown in Table 2.

2.2. Effect of FeO Amount in Initial Slag

Various conditions of slag temperature from 1 773 to 1 973 K and initial FeO content from 10 mass% to 40 mass% were examined in the steady temperature condition. In the prepared slag, 5 kg coal was added and 20 kg CO₂ was constantly introduced. The initial mass% CaO/mass%SiO₂ ratio of slag was 1.

The mole fraction of CO, H₂, CO₂ and H₂O in gas phase was around 86%, 3%, 10% and 1%, respectively. They were barely changed with the change of FeO content in initial slag, and the effect of temperature was unobvious on the mole fraction of gas.

Figure 1 shows the produced amount of CO, H₂ and H₂O gases and the amount of remaining CO₂ gas with FeO content in initial slag at steady temperature condition. Higher the reaction temperature was, more CO was produced and less H₂ was produced. The CO₂ gas left after equilibrium was the opposite with the produced amount of CO gas, while the produced amount of H₂O was the opposite with that of H₂. The following ΔG° was calculated by FactSage 6.3 Software and the calculated temperature range is from 1 600 K to 2 100 K.

\[ \text{C} (s) + \text{CO}_2 (g) \rightarrow 2 \text{CO} (g), \quad \Delta G^\circ = 161 000 - 168.1 T \quad \text{J/mol} \quad (1) \]

Coal reacted with CO₂ in the reduction reaction to produce the combustible gas. Firstly, CO₂ contacted with the surface of the coal granules and was absorbed by carbon to create intermediate complex. Then, CO was generated by the reduction reaction. Since this reaction is a highly endothermic reaction, the reduction of CO₂ is promoted and more CO is produced with the increase of temperature. At the same time, the increase of CO amount with increasing temperature restrained the following reaction. Therefore, the production of H₂ was suppressed.

\[ \text{C} (s) + \text{H}_2\text{O}(g) \rightarrow \text{CO}(g) + \text{H}_2(g), \quad \Delta G^\circ = 133 500 - 141.9 T \quad \text{J/mol} \quad (2) \]

In the case of 1 773 K, 1 823 K and 1 873 K, the produced amount of CO and H₂ decreased with the increase of FeO content higher than 30 mass% in the initial slag, which have the opposite change trends to the produced solid Fe amount as shown in Fig. 2. The activity of FeO in slag phase relative to liquid FeO with the increase of FeO content in initial slag is shown in Fig. 3. When FeO content in initial slag was 20 or 40 mass%, FeO content in slag phase after equilibrium was 19.2 or 33.5 mass% and the activity of FeO in slag phase relative to liquid FeO was 0.29 or 0.58, respectively. It could be seen that the activity of FeO in slag phase was not proportional to the FeO content when FeO content in initial slag was higher than 30 mass%, which was also a proof of the consumption of FeO. With the increase of the activity of FeO, metallic Fe can be formed more easily by carbon in coal. More CO could be produced through the CO₂ reduction compared with the FeO reduction according to the reaction equations. Therefore, in larger FeO activity case, some carbon was consumed for Fe formation and total generated CO amount decreased with the formation of metallic iron.

2.3. Effect of mass%CaO/mass%SiO₂ Ratio in Initial Slag

Various conditions of slag temperature from 1 773 to
1973 K and the initial mass% CaO/mass%SiO2 of slag from 1 to 4 were examined in the steady temperature condition. In the prepared slag, 5 kg coal was added and 20 kg CO2 gas was introduced. FeO content in initial slag was fixed as 20 mass%.

The effects of slag temperature and the initial mass%CaO/mass%SiO2 ratio of slag were unobvious on the mole fraction of H2 in gas phase after equilibrium, which was about 2.6%. Apart from the area where the initial mass%CaO/mass%SiO2 ratio of slag was from 1.4 to 2.1, the effects of slag temperature and the initial mass%CaO/mass%SiO2 ratio of slag were small on the mole fraction of CO and CO2 in gas phase after equilibrium, which was about 86.6% and 9.5%, respectively. The mole fraction of CO decreased while that of CO2 increased with the decrease of temperature in the range of mass%CaO/mass%SiO2 between 1.4 and 2.1.

Figure 4 shows the produced amount of CO, H2 and H2O gases and the amount of remaining CO2 gas with the initial mass%CaO/mass%SiO2 ratio of slag at steady temperature condition. In the case of 1973 K and 1923 K and 1873 K, the effect of mass%CaO/mass%SiO2 ratio of slag were unobvious. In the case of 1773 K, 1823 K and 1873 K, there was a smooth transition point of the amount of CO, CO2, H2 and H2O in gas phase after equilibrium around 1.8 of mass%CaO/mass%SiO2. The mole fraction of CO and H2 in gas phase after equilibrium initially decreased, and then turned to increase and finally became unchanged with the increase of the initial mass%CaO/mass%SiO2 ratio of slag. The change trend of CO2 was opposite to that of CO while the change trend of H2O was opposite to that of H2.

In the database of FactSage 6.3, “solid solution phase”
represents the phase formed by reaction among FeO, Fe₂O₃ and CaO, while “α-2CaO·SiO₂” presents one kind of 2CaO·SiO₂. The production behavior of “solid solution phase” and “α-2CaO·SiO₂” are shown in Fig. 5. The activity of FeO in slag phase relative to liquid FeO with the initial mass%CaO/mass%SiO₂ ratio in slag at steady temperature condition is shown in Fig. 6. When the mass%CaO/mass%SiO₂ ratio of slag was lower than 1.8, the activity of FeO increased with the increase of the mass%CaO/mass%SiO₂ ratio of slag and reached the maximum. Then the activity of FeO decreased and became stable with the further increase of the mass%CaO/mass%SiO₂ ratio of slag. When the mass%CaO/mass%SiO₂ ratio of slag was less than 1.8, the 2CaO·SiO₂ phase was formed continuously from the initial slag with the increase of mass%CaO/mass%SiO₂ ratio. So the FeO content in remaining liquid

Fig. 4. Produced amount of CO, H₂ and H₂O gases and the amount of remaining CO₂ gas with the initial mass%CaO/mass%SiO₂ ratio of slag at steady temperature condition.

Fig. 5. Amount of produced α-2CaO·SiO₂ phase and solid solution phase with the initial mass%CaO/mass%SiO₂ ratio in slag at steady temperature condition.

Fig. 6. Activity of FeO in slag phase relative to liquid FeO with the initial mass%CaO/mass%SiO₂ ratio in slag at steady temperature condition.
phase increased with the increase of solid phase fraction. More oxide ions would be taken in with the increase of CaO content, which made the increase of \( O^{2-} \) in slag. On the one hand, the complicated silicate anions would be depolymerized to form relatively simple structure. On the other hand, \( Fe^{2+} - O^{2-} \) was formed from the increased \( O^{2-} \) and \( Fe^{2+} \) while \( Ca^{2+} \) existed beside the relatively simple complex anions after depolymerization. This made the concentration of \( Fe^{2+} - O^{2-} \) in slag increase, so the activity of FeO increased.\(^{29}\) When the mass\%CaO/mass\%SiO\(_2\) ratio of slag was higher than 2.6, “solid solution phase” was produced because of the reaction between surplus CaO and FeO. Therefore, the activity of FeO was decreased and then became unchanged.

When the initial mass\%CaO/mass\%SiO\(_2\) ratio of slag was from 1.4 to 2.1, the activity of FeO in slag first increased and then decreased with the mass\%CaO/mass\%SiO\(_2\) ratio, which affected the reaction between FeO in slag and carbon for Fe formation. Therefore, the produced amount of CO and H\(_2\) has the according change trend.

3. Thermodynamic Calculation at No Enthalpy Change Condition

In the no enthalpy change calculation, condensed phases after an equilibrium calculation were input as the initial condensed phases in the next calculation and new gas was equilibrated again. The above equilibrium calculation between slag and gas was repeated until temperature decreased below 1 722 K. Coal was introduced together with CO\(_2\) gas at every step. The composition of coal used for calculation is shown in Table 2. Table 3 shows the calculation conditions by FactSage 6.3 in the case of no enthalpy change.

### Table 3. Conditions for calculations in the case of no enthalpy change.

| Slag preparation mass\%CaO/mass\%SiO\(_2\): from 1 to 4 |
|---------------------------------------------------------|
| FeO: from 10 mass\% to 40 mass\% step 5 mass\%         |
| Slag temperature: 1 773, 1 823, 1 873, 1 923 or 1 973 K |
| Total amount: 10 kg                                     |
| Reaction \( FeO - CaO - SiO_2 (slag) + 5 g \text{ coal} + 10 L \text{ CO}_2 (g) \rightarrow FeO - Fe_2O_3 - CaO - SiO_2 (slag) + \text{other condensed phases (exclude solid or liquid Fe)} + H_2O - H_2 - CO - CO_2 (g) \) in every calculation step |
| Gas condition                                         |
| Gas temperature: 298 K                                 |
| Gas composition: \( P(\text{CO}_2) = 1.0 \text{ atm} \) |
| Temperature Change to satisfy no enthalpy change       |

Fig. 7. Produced amount of CO, H\(_2\) and H\(_2\)O gases and the amount of remaining CO\(_2\) gas with the volume of constantly introduced CO\(_2\) gas under different FeO content in initial slag at no enthalpy change condition.
content in initial slag condition. This is because the amount of injected CO$_2$ was adequate to make coal react completely. When the FeO content in initial slag was 10 mass%, the mole fractions of CO, CO$_2$, H$_2$ and H$_2$O were 90.8%, 5.2%, 3.3% and 0.7%, respectively. When the FeO content in initial slag was 40 mass%, the mole fractions of CO, CO$_2$, H$_2$ and H$_2$O were 84.8%, 11.2%, 2.7% and 1.3%, respectively.

Figure 7 shows the produced amount of CO, H$_2$ and H$_2$O gas and the amount of remaining CO$_2$ gas with the volume of constantly introduced CO$_2$ gas under different FeO content in initial slag at no enthalpy change condition. Larger the initial FeO content was, smaller the amounts of produced CO and H$_2$ were. Effect on the amount of CO$_2$ was simply opposite to that of CO while the effect on the amount of H$_2$O was opposite to that of H$_2$. With the introduction of CO$_2$ gas, the activity of FeO in slag phase kept unchanged in any initial FeO content condition, which could be seen in Fig. 8. Liquid or solid Fe was generated from the beginning until 80 L of CO$_2$ gas supply as shown in Fig. 9. The produced amount of Fe reduced sharply when the introduced CO$_2$ gas reached 80 L. This is because the produced Fe was not included as the initial condensed phase in the next calculation. So the activity of FeO in slag phase kept unchanged although the Fe amount decreased. More the FeO content in initial slag was, more obvious the increasing trend of the reduced Fe amount with gas volume was.

![Fig. 8.](image1.png)

**Fig. 8.** Activity of FeO in slag phase relative to liquid FeO with the volume of constantly introduced CO$_2$ gas under different FeO content in initial slag at no enthalpy change condition.

![Fig. 9.](image2.png)

**Fig. 9.** Produced amount of solid or liquid Fe with the volume of constantly introduced CO$_2$ gas under different FeO content in initial slag at no enthalpy change condition.

![Fig. 10.](image3.png)

**Fig. 10.** Mole fraction of CO, CO$_2$, H$_2$ and H$_2$O in gas phase with the amount of constantly introduced CO$_2$ gas under different initial mass%CaO/mass%SiO$_2$ ratio in slag at no enthalpy change condition.
3.2. Effect of mass%CaO/mass%SiO₂ Ratio of Slag

For each calculation step, 10 L CO₂ gas was introduced together with 5 g coal. FeO content in initial slag was 20 mass%. Initial slag temperature was set as 1873 K and introduced gas temperature was 298 K. The initial mass%CaO/mass%SiO₂ ratio of slag was 1, 1.2, 1.5, 1.9, 2.3, 3, 4.

The mole fraction of H₂ in gas phase was about 3.3%, which was not affected by the initial mass%CaO/mass%SiO₂ ratio of slag. The mole fraction of CO and CO₂ also did not change with the supply of gas at each different mass%CaO/mass%SiO₂ ratio condition. Figure 10 shows the mole fraction of CO, CO₂, H₂ and H₂O in gas phase with the amount of constantly introduced CO₂ gas volume under different initial mass%CaO/mass%SiO₂ ratio in slag at no enthalpy change condition. The mole fraction of CO was higher than 83.2%. The mole fraction of CO₂ was more than 4.1% and less than 12.7%. The applicable condition of those results was that the initial mass% CaO/mass% SiO₂ ratios in slag were 1, 1.2, 1.5, 1.9, 2.3, 3 and 4. The effect of mass%CaO/mass%SiO₂ ratio was the same as the steady temperature calculation condition. When mass%CaO/mass%SiO₂ ratio was 1.9, the mole fraction of CO has the minimum value while the mole fraction of CO₂ was the highest.

Figure 11 shows the produced amount of CO, H₂ and H₂O gases and the amount of remaining CO₂ gas with the amount of constantly introduced CO₂ gas under different initial mass%CaO/mass%SiO₂ ratio in slag at no enthalpy change condition. The effect of the mass%CaO/mass%SiO₂ ratio on produced amount of gas was the same for CO and H₂, while that of CO₂ was affected oppositely. When mass%CaO/mass%SiO₂ ratio was 1.9, the produced amount of CO and H₂ were the minimum while that of CO₂ and H₂O were the maximum. When mass%CaO/mass%SiO₂ ratio was 3 or 4, the produced amount of CO and H₂ were the most and that of CO₂ and H₂O were the least.

The above results were related with the activity of FeO in the slag phase relative to liquid FeO, which is shown in Fig. 12. The activity of FeO in the calculation condition was the same as the aforementioned results in Fig. 6, which further demonstrated that the activity of FeO was affected by the mass%CaO/mass%SiO₂ ratio, the amount of produced α-2CaO·SiO₂ phase, and the existence of liquid or solid Fe and the solid solution phases. In large FeO activity...
case, some carbon was consumed by FeO reduction and total generated CO amount decreased with the formation of metallic iron. In the case of mass%CaO/mass%SiO2 ratio of 3 and 4, the FeO activity was the least and the generated CO amount was the most, which were primarily through the endothermic reaction between carbon and CO2. However, temperature change in one calculation step was smaller than others in those cases, which could be seen in Fig. 13. This is because temperature change of the system depends on not only the reduction of CO2 gas to form CO gas but also the reduction of iron oxide or formation of solid phase. In the case of mass%CaO/mass%SiO2 ratio of 3 and 4, significant amount of solid solution phases were formed.

3.3. Effect of Slag Temperature

For each calculation step, 10 L CO2 gas was introduced together with 5 g coal. Supplied gas temperature was 298 K. The initial mass%CaO/mass%SiO2 ratio of slag was 1 and FeO content in initial slag was 20 mass%. Initial slag temperature was varied as 1823 K, 1873 K, 1923 K or 1973 K.

Slag temperature almost has no effect on the mole fraction of CO, H2, CO2 and H2O. Mole fractions of CO, CO2, H2 and H2O were nearly constant at about 90.4%, 5.5%, 3.3% and 0.8%, respectively. Figure 14 shows the produced amount of CO, H2 and H2O gases and the amount of remaining CO2 gas with the amount of constantly introduced CO2 gas under different initial slag temperature at no enthalpy change condition. Although the lines of CO and H2 have the same slope, they are on the different position of the line with different slag temperature. The maximum amount of produced CO, H2 and H2O gases and the maximum amount of remaining CO2 gas under different initial slag temperature are marked by arrows in Fig. 14. Higher the slag temperature was, more CO and H2 gases were produced, because more CO2 gas can be introduced to the calculation system before the temperature reached 1772 K. When the slag temperature was higher, more energy could be reused and more CO2 gas can be introduced.

Slag temperature has no effect on the gas production amount of CO, H2 and H2O in every introduced 10 L gas. In every 10 L CO2 gas supply, about 0.073 mol, 0.00269 mol and 6.403 × 10−4 mol of CO, H2 and H2O was produced. Figure 15 shows the activity of FeO in slag phase relative
and slag in the atmosphere of CO2 gas has been studied and thermodynamic conditions of reactions between coal and thermal energy in steelmaking slag has been considered. The amount of CO2 gas was smaller as the introduced gas condition is higher after heat transfer with slag. Therefore, the amount of CO and H2 reached the minimum. When the initial mass%CaO/mass%SiO2 ratio varied from 1 to 4, the amount of produced CO and H2 became to decrease with the increase of the initial slag temperature as more CO2 gas would be left compared to the low initial slag temperature condition, and thus less CO2 gas would be left compared to the low initial slag temperature condition.

4. Conclusions

The production process of H2, CO gases by utilizing FeO and thermal energy in steelmaking slag has been considered and thermodynamic conditions of reactions between coal and slag in the atmosphere of CO2 gas has been studied by applying thermodynamic calculation in two different conditions.

In the steady temperature condition, the following results are obtained.

1) Lowering the FeO content, the mole fractions of CO and CO2 became larger and smaller, respectively, and more CO and H2 were totally produced.

2) In the present calculation conditions with mass%CaO/mass%SiO2 ratio varied from 1 to 4, the amount of produced CO and H2 reached the minimum at mass%CaO/mass%SiO2 = 1.9, while those were at maximum when mass%CaO/mass%SiO2 ratio was 3 or 4.

3) The amount of CO and H2 gases increased in total with the increase of the initial slag temperature as more CO2 gas can be introduced to the calculation system.

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