Transport Properties of CO$_2$ in Different Reactivity Coke Solution Loss Reaction Based on Stefan Flow Theory

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1. INTRODUCTION

Metallurgical coke is a kind of porous carbon material used in the ironmaking blast furnace. It holds the key as a stock column skeleton, and as heating, carburizing, and reducing agent. The functions of heating, carburizing, and carbon reduction of other fuels such as coal and biochar can be partly replaced. However, as the skeleton of the blast furnace charging column, coke plays an increasingly prominent role, which is the key to the efficiency and service life of a blast furnace.

The degradation of coke strongly dominates the gas permeability of the upper part of the blast furnace and impacts liquid permeability of the lower part of the blast furnace. The main reason for coke degradation is the solution loss reaction of coke, which is the gasification reaction with carbon dioxide (C$_{\text{coke}}$ + CO$_2$ = 2CO). Furthermore, the degradation of coke results from the increase of the gasification rate, the expansion of pores, and the thinning of pore walls. Therefore, it is of great significance to deeply study the coke gasification mechanism for understanding and controlling the reaction process.

At present, the coke quality evaluation method developed by Nippon Steel Corporation in the early 1970s, coke reacts with carbon dioxide at 1100 °C for 2 h, is the most widely used method around the world. It is generally accepted that the coke with a low coke reactivity index% (CRI) and high coke strength after reaction% (CSR) is of high quality. However, the difference in CRI and CSR of coke used by ironmaking enterprises is very significant in different countries. Even in China, the difference in CRI and CSR of coke used by ironmaking plants in different regions is quite distinct. This leads some scholars to question the validity of CRI and CSR experimental conditions. In the blast furnace, the coke solution loss reaction occurs under the conditions of temperature and gas composition change, accompanied by iron ore reduction and other chemical reactions. Cheng and Negro believed that the coke solution loss reaction in a blast furnace is mainly determined by the concentration of carbon dioxide. Goleczka and Barnaba considered that the amount of coke solution loss in the blast furnace should be approximately 20–30 and 25%, respectively. Nomura et al. suggested that CSR was not suitable for evaluating the post-reaction strength of highly reactive coke. The solution loss reaction should stop when the mass loss of coke was 20%, and the reaction temperature should be adjusted accordingly. For solving the confusion of CRI and CSR, Wang et al. proposed a new testing method for coke...
quality evolution, namely, coke gasifies with carbon dioxide at 1100, 1150, 1200, 1250, and 1300 °C, respectively, until the mass loss of coke reaches 25%. Although the new evaluation method solves the problem of coke quality evaluation, it is necessary to investigate the mechanism of the coke solution loss reaction at different temperatures for solving the problem fundamentally.

In the process of solid or liquid combustion, there is a phase interface between the external gas flow and the fuel. Due to the concentration gradient of the external multicomponent gas at the phase interface, the external gas generates normal diffusion flow at the phase interface. This phenomenon of normal diffusion flow along with the physical or chemical process at the interface of two phases can be called Stefan flow. It was proposed by Stefan in 1871 while investigating water evaporation.26

Since then, the notion of Stefan flow has been broadly used in multicomponent mass transfer, particularly in combustion. The calculation results of Caram et al.27 showed that for air combustion, whether or not to consider Stefan flow at low oxygen concentrations had a slight influence on the results. Baum et al.28 studied the combustion of pulverized coal under the mixed control of diffusion and the reaction under the air conditions. It was considered that the existence of Stefan flow contributed to making the combustion of carbon particles controlled by the mass transfer process, and its effect on mass and heat transfer was related to the size of the carbon particle. Kalinchkak et al.29 also proved that the effect of Stefan flow was mainly related to the decrease in the size of the carbon particle during the combustion process, but had little dependence on the temperature. Paterson et al.30 concluded that Stefan flow had a great influence on the prediction of the reaction rate, and ignoring Stefan flow would lead to 85% error. Förtsch et al.31 found that the mass transfer coefficient error was up to 17% depending on whether the Stefan flow was considered or not when the oxidation reaction of carbon particles was considered under the air combustion conditions. Edge et al.32 pointed out that the Stefan flow was helpful in controlling the combustion process by the mass transfer process when carbon was consumed. Yu et al.33 studied the mass transfer coefficient of CO₂ and O₂ under the oxygen-enriched combustion conditions. The results showed that the mass transfer coefficient of CO₂ was more sensitive to the surface reaction rate and reaction products of carbon particles when considering Stefan flow. Whether to consider Stefan flow or not could cause 74% mass transfer coefficient error.

Besides, Stefan flow is well-studied not only in the field of combustion but is also being studied in other fields. Wang et al.34 introduced Stefan flow theory into the oxidation-reduction reaction of carbon-bearing pellets, defined the concept of a bidirectional gas-solid reaction, and established the kinetic model with Stefan flow. During the coke solution loss reaction, the outer surface of coke is the phase interface of the gas-solid phase, and there is a concentration gradient of carbon dioxide and carbon monoxide in the gas boundary layer. When coke reacts with carbon dioxide, the normal diffusion flow of carbon dioxide and carbon monoxide is generated on the outer surface of coke. This mass transfer phenomenon during the coke solution loss reaction is similar to solid fuel combustion. Therefore, in the external diffusion course of the coke solution loss reaction, the diffusion of carbon dioxide and carbon monoxide in the boundary layer will facilitate the Stefan flow phenomenon. However, this has not been reported in many previous studies on coke gasification.35–39 This results in an inaccurate and incomplete understanding of the mass transfer mechanism of the coke solution loss reaction. Huang et al.40 successfully applied the Stefan flow theory into the coke solution loss reaction process and established a kinetic model to discuss the influence of Stefan flow on the coke solution loss reaction. However, the focus of his work was to establish a kinetic model for only one kind of coke, but the application of other kinds of coke is not mentioned. In other words, the effect of Stefan flow on different reactivity coke and its severity need to be further elucidated.

Hence, this paper aims to study how Stefan flow affects the solution loss reaction of coke with different reactivities at different temperatures. The external diffusion coefficient, the mass transfer coefficient, the carbon dioxide concentration on the outer surface of coke, and the carbon dioxide concentration in coke with and without Stefan flow of three kinds of coke with different reactivities are analyzed and compared for illustrating the severity of the Stefan flow effect. It is hoped that this work can expand as well as deepen the understanding of the mechanism of the solution loss reaction of coke with different reactivities, and provide certain theoretical reference for coke quality evaluation.

2. EXPERIMENTAL SECTION

2.1. Samples. Three kinds of metallurgical coke with various CRI and CSR were selected to perform the gasification experiments. Their proximate analyses according to Chinese standards GB/T 2001−2013 and ash composition analyses according to Chinese standards GB/T 1574-2007 are shown in Tables 1 and 2, respectively. Their CRI and CSR, according to Chinese standards GB/T4000-2008 are shown in Table 3. Some other physical properties, such as original apparent density, original surface area, original porosity, and original mean pore radius were measured by mercury intrusion based on the national standard GB/T 21651.1-2008, and the results are exhibited in Table 4. Only one coke sample (diameter: 26−28 mm) was used in each gasification experiment.

![Table 1. Proximate Analyses of Coke Samples (on Air Dry Basis wt %)](https://dx.doi.org/10.1021/acsomega.0c03913)
Therefore, the gasification furnace resulting from solution loss was approximately 20

\[
\text{calculated using the following equation}
\]

\[
m = \frac{m_0 - m_1}{m_0} \times 100\%
\]

(1)

where \( m_0 \) represents the coke weight (g) before the gasification reaction and \( m_1 \) represents the coke weight (g) in real-time.

The average values of the experimental results exhibited in Figure 2 are obtained through three experiments. The repeatability of coke gasification experiments is acceptable. It can be seen from Figure 2 that under any temperature, the reaction time of Coke A is the longest, followed by Coke B, and that of Coke C is the shortest. This indicates that the reactivity of Coke A is less than that of Coke B, which is less reactive than Coke C at each temperature.

The kinetic model of the coke solution loss reaction with the effect of Stefan flow has been established in the previous study. For the external and internal diffusion processes of the coke solution loss reaction, the original calculation method was presented, and the corresponding modified calculation method with the effect of Stefan flow was established. By comparing and analyzing the external diffusion coefficient, mass transfer coefficient, carbon dioxide concentration on the outer surface of coke, and the concentration distribution of carbon dioxide inside coke with and without Stefan flow, the influence of Stefan flow on coke solution loss reaction was illustrated.

Therefore, the above experimental results are used to construct the mathematical model. Through the analysis and comparison of the calculation results, how Stefan flow affects different reactive coke can also be explained.

3. RESULTS AND DISCUSSION

3.1. Experimental Results. Weight loss (\%) \( x \) can be calculated using the following equation

\[
x = \frac{m_0 - m_1}{m_0} \times 100\%
\]

heating rate of 10 °C/min. During the heating process, the moisture was completely removed. When the target temperature was stable and the electronic balance was unchanged, the value on the electronic balance was denoted as the mass of the coke sample before the gasification reaction. The gas was then switched from nitrogen to carbon dioxide (99.99%, 5.0 L/min). In previous studies,\(^{25,41}\) the gasification of coke in the blast furnace resulting from solution loss was approximately 20–30%; therefore, the gasification experiment was stopped before the mass loss of the coke sample reached 25%. Finally, the gas was rapidly switched from carbon dioxide to nitrogen, and the furnace was cooled to room temperature.

Figure 1. Experimental devices. (1) integrated control cabinet, (2) electronic balance, (3) suspension wire, (4) heater, (5) reaction tube, (6) furnace, (7) basket, (8) coke, (9) gas inlet, and (10) thermocouple.

Table 2. Ash Analyses of Coke Samples (wt %)

| coke samples | SiO\(_2\) | Al\(_2\)O\(_3\) | Fe\(_2\)O\(_3\) | CaO | MgO | SO\(_2\) | TiO\(_2\) | K\(_2\)O | Na\(_2\)O | P\(_2\)O\(_5\) | MnO\(_2\) |
|--------------|---------|--------------|--------------|-----|-----|--------|--------|-------|--------|---------|---------|
| coke A       | 47.15   | 35.42        | 6.25         | 2.77| 0.33| 1.74   | 1.62   | 0.51   | 0.47   | 0.51    | 0.05    |
| coke B       | 47.21   | 35.62        | 6.48         | 3.25| 0.53| 1.13   | 1.41   | 0.57   | 0.61   | 0.54    | 0.07    |
| coke C       | 46.72   | 35.35        | 7.12         | 3.39| 0.58| 1.42   | 1.55   | 0.65   | 0.86   | 0.34    | 0.08    |

Table 3. CRI and CSR of Coke Samples

| coke samples | CRI (%) | CSR (%)|
|--------------|---------|--------|
| coke A       | 23.04   | 68.12  |
| coke B       | 28.02   | 56.45  |
| coke C       | 33.27   | 49.47  |

Table 4. Physical Properties of Coke Samples

| coke samples | coke original density \( \rho_0 \) (g/cm\(^3\)) | original surface area \( S_\rho \) (m\(^2\)/g) | original porosity \( \varepsilon_0 \) (%) | original mean pore radius \( R_p \) (nm) |
|--------------|------------------------------------------------|---------------------------------|-----------------------------------|-----------------------------------|
| coke A       | 1.07                                           | 10.29                           | 44.89                             | 81.1                              |
| coke B       | 1.13                                           | 15.38                           | 48.66                             | 55.9                              |
| coke C       | 1.064                                          | 10.26                           | 47.63                             | 70.8                              |
corresponding temperatures, and the degree of decrease is very obvious. This indicates that regardless of the kinds of coke, the influence of Stefan flow on gas diffusion and mass transfer capacity in the boundary layer is very serious and cannot be ignored. Furthermore, at each temperature, $D_{CO,CO,SF}$ and $h_{DSF}$ are changing and fluctuating during the reaction process. The values of $D_{CO,CO,SF}$ and $h_{DSF}$ are not equal for the different kinds of coke. This indicates that the diffusion and mass transfer capacity of the gas in the boundary layer of different kinds of coke are distinct, which illustrates the difference in coke types when considering Stefan flow.

To more intuitively display how Stefan flow affects the diffusion coefficient and mass transfer coefficient of different kinds of coke, the average deviation of $D_{CO,CO}$ and $D_{CO,CO,SF}$ and $h_D$ and $h_{DSF}$ needs to be calculated, and the equation is shown below

$$ADEV(X_{org})\% = \frac{\sum_{i=1}^{N} (X_{org,i} - X_{org,mod,i} \times 100)}{N}$$

(2)

where ADEV is the average deviation (%), $X_{org}$ is the original parameter, and $X_{mod}$ is the modified parameter. The average deviations of $ADEV(D)$ and $ADEV(h)$ of Coke A, B, and C at different temperatures are shown in the following figure.

For the convenience of comparison, the ordinate of Figure S4a,b is unified. It can be observed that $ADEV(D)$ and $ADEV(h)$ of Coke A, B, and C are decreased with the increase of temperature. It indicates that the influence of Stefan flow on the external diffusion coefficient and mass transfer coefficient is weakened as the temperature is increased. At the same temperature, the $ADEV(D)$ of the three kinds of coke is higher than that of $ADEV(h)$, indicating that the difference of the external diffusion coefficient is larger than that of the mass transfer coefficient. This is because the external diffusion coefficient and mass transfer coefficient are correlated by the Sherwood number, and there is a relationship between the Sherwood number, Reynolds number, and Schmidt number, which is similar to the Ranz–Marshall equation. This leads to different reductions of the external diffusion coefficient and mass transfer coefficient under the influence of Stefan flow. The final result is that $ADEV(D)$ and $ADEV(h)$ are not equal at the same temperature. The values of $ADEV(D)$ and $ADEV(h)$ show that Stefan flow has a significant influence on the external diffusion coefficient and mass transfer coefficient.

The results of $ADEV(D)$ of Coke A, B, and C ranged from 48.71 to 44.57%, 47.61 to 43.27%, and 47.56 to 43.03%, respectively, from 1100 to 1300 °C. The values of $ADEV(h)$ of Coke A, B, and C change in the range of 46.04~42.57, 43.17~39.47, and 43.15~39.15% in the same temperature range. Based on the above results, the average deviations of $ADEV(D)$ and $ADEV(h)$ are very significant and not negligible. Furthermore, if Stefan flow is ignored in the external diffusion process, the external diffusion coefficient and mass transfer coefficient of gas will be evidently different. This will lead to a marked difference in the understanding of the diffusion and mass transfer ability of gas in the boundary layer. The reality is that the diffusion and mass transfer capacity of gas in the boundary layer is not as strong as previously thought when considering Stefan flow.

The order of $ADEV(D)$ and $ADEV(h)$ of three kinds of coke at each temperature is as follows: $ADEV(D)$ and $ADEV(h)$ of Coke A (low reactivity) are greater than those of Coke B (medium reactivity), and $ADEV(D)$ and $ADEV(h)$ Coke B are greater than those of Coke C (high reactivity). This shows that the influence of Stefan flow on gas diffusion and mass transfer capacity in the boundary layer decreases as coke reactivity increases, that is, as the coke reactivity decreases, the Stefan flow influence becomes greater. The difference in $ADEV(D)$ of three kinds of coke is not very obvious at the same temperature. However, the difference of $ADEV(h)$ of three kinds of coke is somewhat significant at each temperature, that is, the $ADEV(h)$ of Coke A with low reactivity is significantly higher than that of

Figure 2. Experimental results. (a) the weight loss (%) of Coke A, (b) the weight loss (%) of Coke B, and (c) the weight loss (%) of Coke C.
the other two kinds of coke. This indicates that the gas mass transfer capacity of low reactivity coke decreases more apparently, and the influence of Stefan flow on low reactivity coke is greater than that of medium and high reactivity coke. The lower the reactivity of coke, the stronger the Stefan flow effect will be.

To compare the values of \( D_{CO_2,CO,CO} \) and \( h_{D,CO} \) of three kinds of coke more intuitively, the average value of \( D_{CO_2,CO,CO} \) and \( h_{D,CO} \) at different temperatures is calculated to compare the difference in the external diffusion coefficient and mass transfer coefficient of coke with different reactivities after modification.

Figure 6 is the comparison of the average values of \( D_{CO_2,CO,CO} \) and \( h_{D,CO} \) of all coke at various temperatures. Figure 6a,b exhibits that \( AVE(D_{CO_2,CO,CO}) \) and \( AVE(h_{D,CO}) \) of three kinds of coke are different at different temperatures. Although the difference of \( AVE(D_{CO_2,CO,CO}) \) of three kinds of coke at each temperature is not much greater than that of \( AVE(h_{D,CO}) \), the existence of the difference in \( AVE(D_{CO_2,CO,CO}) \) reflects the differences among coke types when Stefan flow is taken into account. Additionally, the numerical order of \( AVE(D_{CO_2,CO,CO}) \) and \( AVE(h_{D,CO}) \) has a positive correlation with the order of coke reactivity. \( AVE(D_{CO_2,CO,CO}) \) and \( AVE(h_{D,CO}) \) are increased with coke reactivity at each temperature. According to the previous study, considering Stefan flow, the calculation of \( D_{CO_2,CO,CO} \) involves the molar fraction of carbon dioxide on the outer surface of coke. The lower the reactivity of coke is, the higher the molar fraction of carbon dioxide on the outer surface of coke is, which leads to the smaller values of \( D_{CO_2,CO,CO} \). Moreover, \( h_{D,CO} \) is calculated from \( D_{CO_2,CO,CO} \), and thus the case of \( h_{D,CO} \) is the same as that of \( D_{CO_2,CO,CO} \). The higher the reactivity of coke, \( D_{CO_2,CO,CO} \) and \( h_{D,CO} \) will appear. Therefore, this indicates that when Stefan flow is considered, the higher the coke reactivity is, the stronger the gas diffusion and mass transfer capacity will be in the boundary layer at the same temperature.

3.3. Carbon Dioxide Concentration on the Outer Surface of Coke. Figure 7 gives the information about the concentration of carbon dioxide on the outer surface of three kinds of coke with and without Stefan flow at different temperatures. All ordinates in Figure 7 are in the same data range. On the left side, Figure 7a,c,e shows the original concentration of carbon dioxide on the outer surface of Coke A, Coke B, and Coke C, with and without Stefan flow at different temperatures. (\( D_{CO_2,CO: original external diffusion coefficient, D_{CO_2,CO,CO}: external diffusion coefficient with Stefan flow \)) (a) \( D_{CO_2,CO} \) of Coke A; (b) \( D_{CO_2,CO,CO} \) of Coke A; (c) \( D_{CO_2,CO} \) of Coke B; (d) \( D_{CO_2,CO,CO} \) of Coke B; (e) \( D_{CO_2,CO} \) of Coke C; and (f) \( D_{CO_2,CO,CO} \) of Coke C.
dioxide concentration on the outer surface of coke $C_{CO_2,S}$ of Coke A, B, and C. On the other side, it shows the carbon dioxide concentration on the outer surface of coke with Stefan flow $C_{CO_2,S,SF}$ of three kinds of coke. The figure shows that the carbon dioxide concentration on the outer surface of the three kinds of coke, whether original or modified, decreases as the temperature increases, and this general rule is unchanged. At each temperature, the $C_{CO_2,S,SF}$ of three kinds of coke is always less than that of $C_{CO_2,S}$, which indicates that the influence of Stefan flow on the carbon dioxide concentration on the outer surface of coke is very significant, and Stefan flow leads to the decrease of carbon dioxide amount on the outer surface of coke. For comparing how Stefan flow affects $C_{CO_2,S}$ and $C_{CO_2,S,SF}$ of three
kinds of coke more intuitively, the average deviation of $C_{CO,S}$ and $C_{CO,S,SF}$ in the above figure is calculated.

Figure 8 shows the average deviations of carbon dioxide concentration on the outer surface of three kinds of coke with and without Stefan flow at different temperatures. In the figure, the ADEV($C_{CO,S}$) of the three kinds of coke increases as temperature increases, which indicates that the impact of Stefan flow on the carbon dioxide concentration on the outer surface of coke increases as the temperature increases. This is because, as the temperature increases, the reaction rate becomes faster, and the chemical reaction on the outer surface should be more intense, and thus, the influence of Stefan flow will be more
serious. Therefore, the carbon dioxide concentration on the outer surface of coke is increasingly affected. Besides, for Coke A, B, and C individually, the values of ADEV(C_{CO2,S}) of Coke A is less than that of Coke B, which is less than that of Coke C at each temperature. This shows that the lower the coke reactivity, the smaller the influence of Stefan flow on the carbon dioxide concentration on the outer surface of coke will be, and this impact is increased as the coke reactivity increases, and the gradient law is relatively obvious at each temperature. This is also because of the higher reactivity of coke, whether at low or high temperature, the chemical reaction intensity on the outer surface of coke will be higher than that of coke with lower reactivity. Therefore, the effect of Stefan flow on high reactivity coke is greater than that of coke with low reactivity. The intensity gradient of such influence is clear at each temperature.

To illustrate the relationship between the coke reactivity and the modified carbon dioxide concentration on the outer surface of coke, Figure 9 exhibits the average values of carbon dioxide concentration on the outer surface of three kinds of coke under different temperatures. (AVE(C_{CO2,S,SSF}): average values of C_{CO2,S} and C_{CO2,SSF}).

Figure 8. Comparison of average deviations of ADEV(C_{CO2,S}) of three kinds of coke at different temperatures. (ADEV(C_{CO2,S}): average deviation of C_{CO2,S} and C_{CO2,SSF}).

Figure 9. Comparison of average values of AVE(C_{CO2,SSF}) of three kinds of coke at different temperatures. (AVE(C_{CO2,SSF}): average values of C_{CO2,SSF}).

3.4. Distribution of Carbon Dioxide Concentration in Coke. The influence of Stefan flow on the external diffusion process of the coke gasification reaction has an indirect impact on the internal diffusion-chemical reaction process. This effect is embodied in the concentration distribution of carbon dioxide in coke. Therefore, according to the previous work, the concentration distribution of carbon dioxide inside the three kinds of coke is calculated, and the original and modified carbon dioxide concentration distribution in the coke is shown in Figure 10.

Figure 10 shows the distribution of carbon dioxide concentration on the three kinds of coke with and without Stefan flow at various temperatures. All of the ordinates are unified for comparison. The distribution of carbon dioxide concentration in coke in Figure 10 is different from conventional ones. A new concept is introduced, namely, the carbon dioxide concentration region in coke. The upper limit of each carbon dioxide concentration region is the carbon dioxide concentration curve at a weight loss of 25%, which means that all of the other carbon dioxide concentration curves at weight loss of less than 25% are in the region. Therefore, the effect of Stefan flow on the distribution of carbon dioxide concentration in coke can be more directly reflected by the size of the carbon dioxide concentration region.

As seen in Figure 10, both the C_{CO2} region and C_{CO2,SSF} region decrease as the temperature increases. This rule is always unchanged irrespective of Stefan flow considerations. Besides, all of the C_{CO2} regions of three kinds of coke under each temperature are larger than that of the C_{CO2,SSF} region. However, the difference between the C_{CO2} region and C_{CO2,SSF} region is not so large. This indicates that Stefan flow has a certain influence on the distribution of carbon dioxide concentration of inner coke, but this effect does not seem very significant. The area of the C_{CO2} region and C_{CO2,SSF} region of three kinds of coke at each temperature, has no relationship with coke reactivity because the distribution of carbon dioxide concentration in coke is determined by the effective internal diffusion coefficient of carbon dioxide and the reaction rate constant.

To present more clearly the influence of Stefan flow on the distribution of carbon dioxide concentration in three kinds of coke, the C_{CO2} region area and C_{CO2,SSF} region area in Figure 10 are integrated, and their deviation is calculated as follow

\[
\text{DEV}(C_{CO2})\% = \frac{S(C_{CO2}) - S(C_{CO2,SSF})}{S(C_{CO2})} \times 100\% \quad (3)
\]

In the equation, DEV(C_{CO2}) is the deviation of the integral area of the C_{CO2} region and C_{CO2,SSF} region (%), S(C_{CO2}) is the integral.
Figure 10, continued
area of the $C_{CO_2}$ region, $S(C_{CO_2, SF})$ is the integral area of the $C_{CO_2, SF}$ region. And the calculated results are plotted as follows.

The deviations of the integral area of the $C_{CO_2}$ region and $C_{CO_2, SF}$ region of three kinds of coke under various temperatures, are shown in Figure 11. This shows that regardless of the type of coke, $DEVS(C_{CO_2})$ increases linearly with the increase of temperature, and this trend remains unchanged. The $DEVS(C_{CO_2})$ values of Coke A, B, and C vary from 6.62 to 22.85%, 7.74 to 25.17%, and 8.62 to 26.74% at $1100 - 1300$ °C, respectively. This suggests that the effect of Stefan flow on carbon dioxide concentration distribution in coke increases as the temperature increases. At $1150$ °C, the $DEVS(C_{CO_2})$ is higher than 10%, indicating that the effect of Stefan flow is significant and not negligible. From the location of the fitting lines in Figure 11, the effect of Stefan flow on the carbon dioxide concentration distribution in coke increases with increasing coke reactivity at any temperature, which cannot be obtained from Figure 10. Thus, it can be concluded that the higher the coke reactivity is, the more significant the influence will be.
Figure 11. Deviations of DEVS(C_{CO2}) of three kinds of coke at different temperatures. (DEVS(C_{CO2}): deviations of the integral area of C_{CO2} region and C_{CO2,SSF} region).

4. CONCLUSIONS
How Stefan flow affected carbon dioxide and coke gasification with different reactivities was discussed, analyzed, and compared in the temperature range of 1100–1300 °C. Regardless of coke types, the external diffusion coefficient and mass transfer coefficient with Stefan flow were smaller than those without Stefan flow. Especially the modified external diffusion coefficients of the three kinds of coke were different at any temperature, which aligned more with the actual situation and reflected the individuality of the gasification process of coke with different reactivities. The minimum average deviations of the external diffusion coefficient and mass transfer coefficient with and without Stefan flow of Coke A, B, and C were 44.57/43.27/ 42.23 and 42.57/39.47/39.15%, respectively. This indicates the influence of Stefan flow on gas diffusion and mass transfer capacity in the boundary layer decreases as the coke reactivity increases. The average values of the modified external diffusion coefficient and the modified mass transfer coefficient had a positive correlation with the reactivity order of coke at any temperature. The carbon dioxide concentration on the outer surface of three kinds of coke was decreased with Stefan flow at any temperature. The average deviations of the carbon dioxide content on the outer surface of Coke A, B, and C before and after modification ranged from 2.87 to 10.91%, 4.81 to 12.49%, and 5.18 to 12.67%, respectively. These were increased with coke reactivity, which means, the effect of Stefan flow on the carbon dioxide concentration on the outer surface of coke is increased as the coke reactivity increases. The average values of carbon dioxide concentration on the outer surface of three kinds of coke with Stefan flow decreased as coke reactivity increased. The area of the carbon dioxide concentration region of three kinds of coke at any temperature with Stefan flow was less than that without Stefan flow. The deviations of the integral area of carbon dioxide concentration region with and without Stefan flow of Coke A, B, and C varied from 6.62 to 22.85%, 7.74 to 25.17%, and 8.62 to 26.74% at a temperature range of 1100–1300 °C, respectively. This indicates that the effect of Stefan flow on carbon dioxide concentration distribution in coke increases as coke reactivity increases.

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