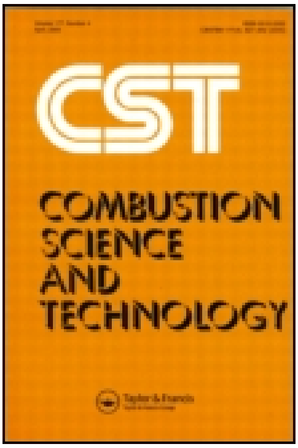

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Sen Li [a] & Xiaolin Wei [a]

[a] Institute of Mechanics, Chinese Academy of Sciences, Beijing, China

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NUMERICAL SIMULATION OF CO AND NO EMISSIONS DURING CONVERTER OFF-GAS COMBUSTION IN THE COOLING STACK

Sen Li and Xiaolin Wei
Institute of Mechanics, Chinese Academy of Sciences, Beijing, China

Oxygen converter steelmaking produces a large amount of high temperature off-gas. During pre- and postcombustion of converter off-gas in the cooling stack, CO concentration at the outlet is always over emissions standards, and NO emission is still paid little attention. In the article, CO and NO emissions are investigated during converter off-gas combustion by computational fluid dynamics (CFD). The simulation results indicate that CO emission is above 0.5%–2%, and NO emission is up to 150 ppmv at the precombustion stage of off-gas during 7–15% of blowing oxygen time. The poor mixing of both off-gas and the air that is sucked in, and too high combustion temperature (above 2000°C) result in high CO emission. In order to increase combustion efficiency of off-gas and to reduce CO concentration, the air supply mode must be improved to effectively organize the mixing of off-gas and air in the cooling stack. The high combustion temperature of off-gas and the poor mixing of both off-gas and the air that is sucked in facilitate the formation of thermal-NO during the pre- and postcombustion of off-gas. In order to reduce NO emission, both the air stoichiometry and temperature profiles in the cooling stack must be controlled.

Keywords: CO; Combustion; Converter off-gas; NO; Numerical simulation

INTRODUCTION

Many pollutants, such as dusts, SO_2, CO, NO_x, etc., are emitted from the iron making and steelmaking processes. Oxygen converter steelmaking is the major steelmaking method, which produces a large amount of high temperature off-gas (Aleksashin et al., 2007; Perlov and Nitskevich, 1965). The converter off-gas is a precious valuable fuel containing about 80% CO in the period of maximum gas production (Perlov and Nitskevich, 1965). However, off-gas formation is intermittent, and gas composition concentrations vary periodically, which makes the control of pollutant emissions very difficult.

In the converter steelmaking process, the converter decarburizes the carbon in the pig iron using pure oxygen at an approximate rate of $110\,m^3\,t^{-1}$ (molten steel), and about $100\,m^3$ of high temperature off-gas is generated, which issues from the mouth of the converter at temperature of about 1600°C. The average content of CO

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Address correspondence to Sen Li, Institute of Mechanics, Chinese Academy of Sciences, No. 15 Beisihuanxi Road, Beijing, 100190, China. E-mail: lisen@imech.ac.cn

Table 1 The rate constants of thermal-NO reactions

The rate constants of forward reaction	The rate constants of reverse reaction
$k_{1f} = 1.8 \times 10^8 e^{-38370/T}$	$k_{1r} = 3.8 \times 10^7 e^{-425/T}$
$k_{2f} = 1.8 \times 10^4 e^{-4680/T}$	$k_{2r} = 3.81 \times 10^3 e^{-20820/T}$
$k_{3f} = 7.1 \times 10^7 e^{-450/T}$	$k_{3r} = 1.7 \times 10^8 e^{-24560/T}$

in off-gas is about 70%, to which corresponds a heat of combustion of 9.21 MJ/m^3 (Roesler et al., 1994). Converter off-gas discharge is intermittent, and its discharge cycle is synchronous with the tap-to-tap cycle of converter steelmaking, of which tap-to-tap cycles are about 30–40 min with 13–18 min of blowing oxygen period. Off-gas composition concentrations frequently vary with blowing oxygen time. At present, the waste heat and off-gas are often recovered by non-combustion method. At the beginning and end of steelmaking, owing to high O_2 concentration in off-gas, there exists the explosion possibility of off-gas, and the concentrations of CO and O_2 cannot meet the requirement of off-gas recovery (Perlov and Nitskevich, 1965). Here in order to ensure the safety of off-gas recovery, a large amount of air is sucked in, and off-gas combusts in the cooling stack without recovery, namely precombustion and postcombustion. Therefore, a large quantity of "thermal" NO may be formed.

At present, the control of pollutant emissions during steelmaking mainly focuses on the dusts and SO_2 (Braginets and Prikhozhenko, 1967; Li et al., 2008); CO concentration at the stack outlet is always over emission standard in china (200 mg/m^3), and NO emission is still paid little attention. CO is a toxic gas, and it is dangerous to human health. CO and NO_x emissions of converter off-gas are, respectively, $1500 \sim 7960$ g/t (steel) and $5 \sim 20$ g/t (steel) in the European Union (Harald, 2001). In the meantime, CO is valuable fuel. Therefore, high emission of CO results in not only atmospheric pollutant, but also fuel loss. NO is a known precursor to the formation of ozone and acid rain, and it can react with volatile organic compounds to form photochemical smog (Li et al., 2008). In this article, CO and NO emissions are investigated during converter off-gas combustion by means of CFD.

CFD SIMULATIONS

Off-Gas Discharge Process

Figure 1 shows a sketch of the cooling stack, which comprises the following components: movable hood and four fixed sections. In the practical off-gas recovery process, to ensure the safety of off-gas recovery, the movable hood of the cooling duct is raised, the low-quality off-gas is combusted in the beginning of steelmaking, and the gas flue produced is used to scour the cooling stack of off-gas to prevent the contact of off-gas and air. When O_2 concentration is below 1%, the movable hood is lowered, and the off-gas is recovered. In the end stage of blowing oxygen, the movable hood is raised again, and the off-gas is combusted. In the cooling stack, high-temperature off-gas or flue gas is cooled to below 1000°C at the outlet of the cooling stack.

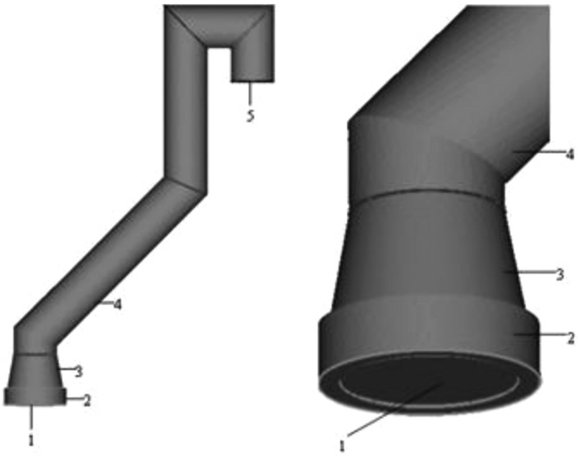


Figure 1 Sketch of the cooling stack. 1: Converter mouth; 2: the gap between converter mouth and moveable hood; 3: moveable hood; 4: cooling stack; 5: the outlet of the cooling stack.

Computation Domain and Mesh Generation

The simulating object is a 120-ton top-blown oxygen converter, whose cooling stack is 33.58 m long. In order to model the process accurately, the calculation domain should be chosen reasonably. For the present simulation, the modeled converter domain is the part above the converter mouth. In order to correctly describe the air suction into the converter from the atmosphere, a large cylinder representing the atmosphere is enclosed around the gap between the hood entrance and the converter mouth. The three-dimensional domain of the modeling system is illustrated in Figure 1. Based on the assumption that flow is symmetric along the plane of symmetry, the calculation was made using half of the domain to reduce computing time. The domain is modeled by a combination of (hexahedral) individually discretized numerical subgrids, as shown in Figure 2. The total grids contain approximately 46,000 cells.

Mathematical Models

This comprehensive model employs the k–ε model for gas turbulence, the eddy dissipation concept (EDC) combustion model for gas turbulent combustion, and the spherical harmonics approximation for radiative heat transfer.

Although the properties of off-gas vary with blowing oxygen time, their variations are small during the combustion residence time of off-gas in the cooling stack (1–2 s). Consequently, it is assumed that the processes of flow, heat transfer, and combustion are quasisteady states during combustion residence time of off-gas. Thus the entire off-gas combustion process in the whole blowing oxygen time is simulated by computing the combustion at different blowing oxygen time slices, which can not only reduce the computation time but also guarantee the computational accuracy.

The gas mixture in the modeling system includes six species: H_2O, O_2, N_2, CO, CO_2, and NO. The density of the mixture is defined by the incompressible ideal-gas

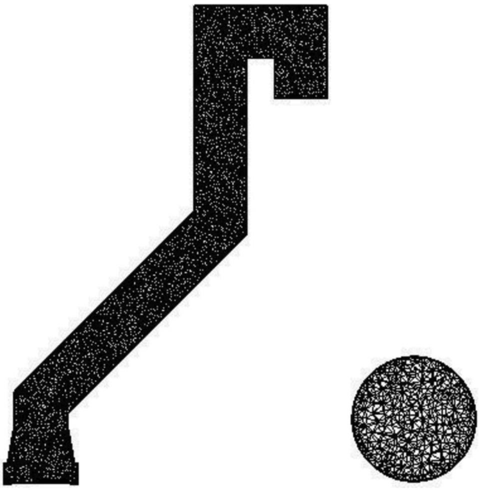


Figure 2 Scheme grids of the computing domain.

method, and mixing law is employed to express the specific heat capacity of the gas mixture. In the simulation, the thermal conductivity of each species is taken as a constant for simplification because heat conduction does not play an important role in the heat transfer over the full model domain. N_2 and O_2 can be considered optically transparent gases with regard to radiation. However, CO, H_2O, and CO_2 have radiation qualities and cannot be simply treated as gray gas. The weighted sum of gray gases model (WSGGM) is used for gray gas.

In the simulation, the only combustion reactions assumed to occur are as follows (Kobayashi et al., 2007; Tang et al., 2005):

$$\mathrm{CO} + \frac{1}{2}\mathrm{O_2} + \mathrm{H_2O} \rightarrow \mathrm{CO_2} + \mathrm{H_2O} \tag{1}$$

$$\mathrm{CO_2} \rightarrow \mathrm{CO} + \mathrm{O_2} \tag{2}$$

CO coming from the converter burns as it meets O_2 from the outside air. In this case, the combustion is said to be mixing-limited. The eddy-dissipation model is used to calculate the reaction rate, which is assumed to be controlled by the turbulence (Tang et al., 2005).

During fuel combustion, three principal NO_x forms are "thermal" NO_x, "prompt" NO_x, and "fuel" NO_x. The relative contribution of each of the total NO_x formed depends on the combustion process and fuel characteristics. Converter off-gas gas contains virtually no fuel nitrogen. For the high-temperature off-gas combustion in the cooling stack, the flame temperature may reach above 1500°C, and thus the NO_x form is thermal NO_x. Thermal NO_x is formed from the reaction of nitrogen and oxygen supplied by the combustion air stream, and it is highly dependent on temperature. During combustion, oxygen radicals are formed; these attack atmospheric nitrogen molecules to start the reactions that comprise the thermal NO_x formation mechanism (extended Zeldovich mechanism) (Miller and Bowman, 1989):

$$\mathrm{O} + \mathrm{N_2} \overset{k_{1f}\ k_{1r}}{\longleftrightarrow} \mathrm{NO} + \mathrm{N} \tag{3}$$

$$\mathrm{N} + \mathrm{O_2} \overset{k_{2f}\ k_{2r}}{\longleftrightarrow} \mathrm{NO} + \mathrm{O} \tag{4}$$

$$\mathrm{N} + \mathrm{OH} \overset{k_{3f}\ k_{3r}}{\longleftrightarrow} \mathrm{NO} + \mathrm{H} \tag{5}$$

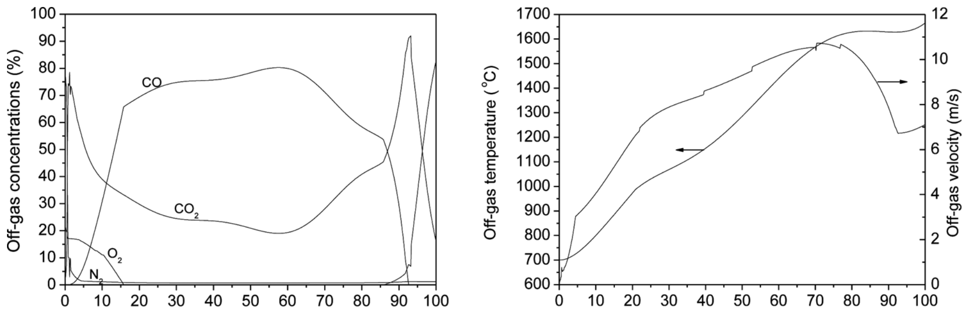
In the above expressions, k_{1f}, k_{2f}, and k_{3f} are the rate constants for the forward Eqs. (3)–(5), respectively, and k_{1r}, k_{2r}, and k_{3r} are the corresponding reverse rate constants. In Table 1, all of these rate constants have units of $\mathrm{cm}^3 \cdot (\mathrm{mol} \cdot \mathrm{s})^{-1}$. The simulation of NO_x is performed as the postprocessor after having obtained the temperature and species field.

Solving Process

The segregated solver is used for quasisteady models. Solution equations in the present CFD simulations include modeling of flow, turbulence, species transfer, heat transfer, and chemical reaction. Semi-implicit method for pressure-linked equations (SIMPLE) is applied to pressure–velocity coupling. Momentum, turbulence kinetic energy, turbulence dissipation rate, species, and energy equations used first-order upwind in the beginning. Based on the converged solution with the discretization scheme of first-order upwind, the second-order upwind scheme is employed afterward in order to reduce computing errors.

Definition of Initial and Boundary Conditions

During converter steelmaking, the temperature, velocity, and concentrations of converter off-gas at the inlet of cooling stack are as shown in Figure 3, and these parameters are obtained from basic oxygen furnace (BOF) simulation by a mathematical model (Li and Wei, 2011). The pressure of the gap between the converter



(a) (b)

Figure 3 The initial condition of converter off-gas at cooling stack inlet. (a) initial concentrations, and (b) initial velocity and temperature.

mouth and the moveable hood is set to 0 Pa. Water cooling tubes are installed on the cooling stack wall, and the temperature of the cooling stack wall is set to 180°C. In the range 0–17% and 83–100% of blowing oxygen times, converter off-gas with high O_2 concentration has the risk of exploding, and then off-gas is combusted with air sucked from the atmosphere through the raising moveable hood, where the height of the gap between the converter mouth and the moveable hood is 1000 mm. The temperature of the air sucked in is 35°C, and the relative humidity is 40%. In the range 17–83% of blowing time, converter off-gas is recovered by lowering the moveable hood, where the height of the gap between the converter mouth and the moveable hood is 0 mm.

RESULTS AND DISCUSSIONS

Model Validation

In the practical steelmaking process, the converter off-gas temperature is monitored. Figure 4 shows the comparison of the predicted and measured flue gas temperature at the outlet of cooling stack. The predicted values are in good agreement with measured values, which indicates that the model can reasonably describe the off-gas combustion and heat exchange process in the cooling stack.

CO Concentration During Converter Off-Gas Combustion in the Cooling Stack

Incomplete combustion of off-gas during pre- and postcombustion may cause not only energy loss, but also environmental pollution. As for CO and O_2 at the same simulation temperature points in Figure 4, the simulation results at the exit at the outlet of the cooling stack are shown in Figure 5. At the precombustion stage

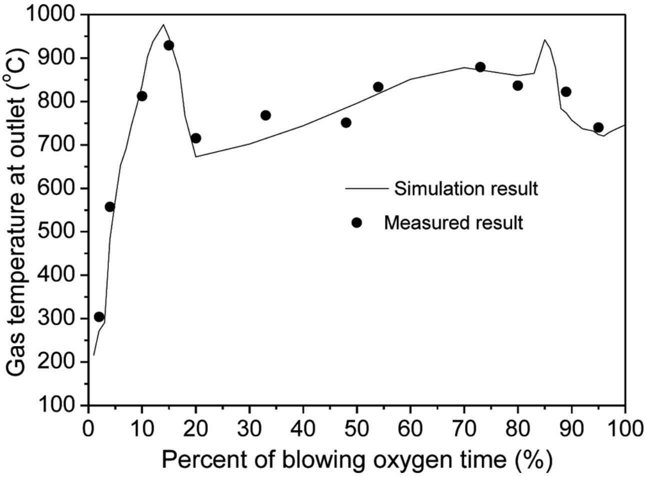


Figure 4 Comparison of the predicted and measured temperature of off-gas at the outlet of the cooling stack during blowing oxygen.

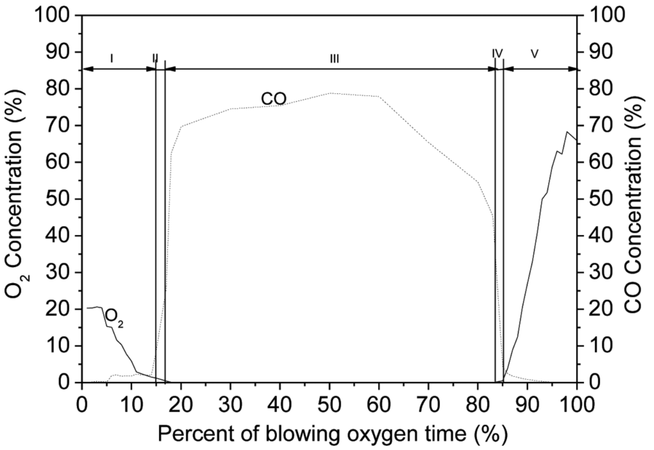


Figure 5 Concentrations of CO and O_2 at the outlet of cooling stack. I: precombustion; II: lowering moveable hood; III: off-gas recovery; IV: raising moveable hood; V: postcombustion.

(5–15% of blowing oxygen time), the concentration ranges of CO and O_2 are 1–2% and above 1.7%, respectively. At the postcombustion stage (85–95% of blowing oxygen time), the concentration ranges of CO and O_2 are 0.5–2.0% and above 4%, respectively. During pre- and postcombustion, the excess air ratio is larger than 1.05, and gas combustion should be complete when the combustion residence time is 1–2 s. However, simulation results indicate that off-gas combustion is incomplete, and CO concentration is above 0.5–2.0%.

During pre- and postcombustion of off-gas, the ambient-temperature air is sucked into the cooling stack through the gap between the converter mouth and the moveable stack raised, and converter off-gas mixes with the air to combust. The mixing of off-gas and air is crucial to the highly efficient combustion of off-gas. The simulating results indicate that the profiles of gas velocity at different blowing times are similar. The velocity profile at 14% of blowing time is given in Figure 6, which shows that a large amount of ambient air is sucked into the cooling stack, and there is little change of the profile velocity along the gas flow path, which is not conducive to the mixing of off-gas and air.

Turbulence kinetic energy (TKE) can reflect the intensity of turbulent fluctuation, and high turbulence kinetic energy means intensive mixing of gas flow. Generally, the TKE can be quantified by the mean of the turbulence normal stresses:

$$k = \frac{1}{2}\left(\overline{u'^2} + \overline{v'^2} + \overline{w'^2}\right) \tag{6}$$

Figure 7 shows the turbulence kinetic energy profiles of the central longitudinal section of the cooling stack during pre- and postcombustion of off-gas. The simulation results indicate that the turbulence kinetic energy is high only at the turning direction of the cooling stack. The gas disturbance in the cooling stack mainly depends on the direction change of the stack, and the disturbance is low, which is not conducive to off-gas combustion.

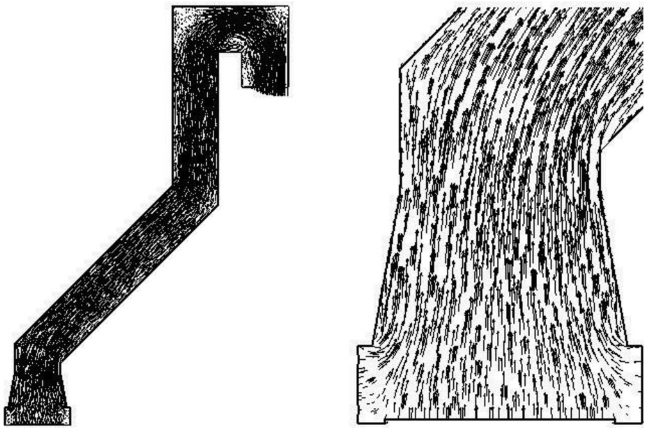
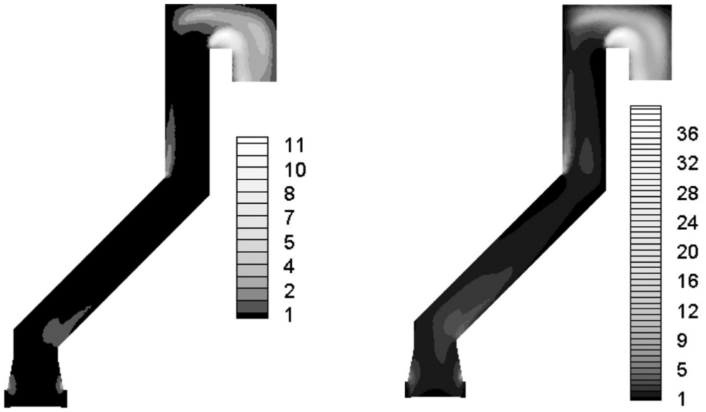


Figure 6 The profiles of gas velocity in the cooling stack at 14% of blowing oxygen time.

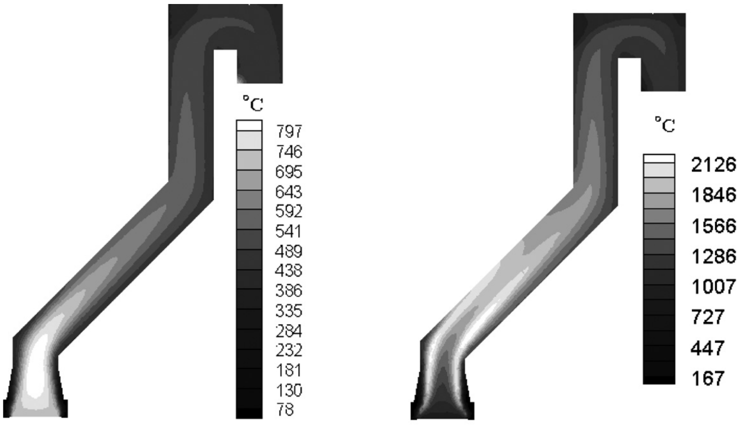
During carbon monoxide combustion, the combustion temperature of off-gas is important to combustion efficiency. Figure 8 shows the profiles of combustion temperature of the central longitudinal section of the cooling stack. At the early stage of precombustion (4% of blowing oxygen time), owing to the low temperature and low CO concentration of off-gas entering the cooling stack, the volume release heat rate during off-gas combustion in the cooling stack is low. The ambient-temperature air sucked in flows near the stack wall, and the mixing of air and off-gas is difficult. Therefore, off-gas combustion mainly occurs in the center of the cooling stack, as shown in Figure 8a.

At later stage of precombustion (14% of blowing oxygen time), the gas temperature can reach above 2000°C, as shown in Figure 8b. The high-temperature and high CO concentration of off-gas entering the cooling stack is conducive to



(a) (b)

Figure 7 The turbulence kinetic energy profiles of the central longitudinal section of cooling. (a) At 14% of blowing oxygen time, (b) at 87% of blowing oxygen time.



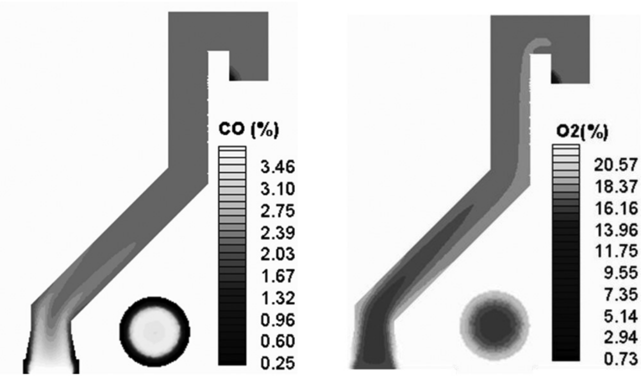
(a) (b)

Figure 8 The gas temperature profiles of the central longitudinal section of cooling stack. (a) At 4% of blowing oxygen time, (b) at 14% of blowing oxygen time.

off-gas combustion. However, when the combustion temperature is above 2000°C, CO_2 may decompose into CO and O_2 [see Eq. (2)], and CO and O_2 reach near reaction equilibrium, which makes the overall oxidation rate of CO decrease. Therefore, a combustion temperature that is too high results in high CO concentration.

During carbon monoxide combustion, the dependency of the oxidation rate on oxygen concentration is important to combustion efficiency (Roesler et al., 1994), which is enhanced with the increase of temperature.

Figure 9 shows the profiles of CO and O_2 concentrations at 4% of blowing oxygen time. Since a large amount of ambient-temperature air is sucked in, the combustion temperature is below 800°C (see Figure 8a). Although O_2 concentration is high in the cooling stack, the oxidation rate of carbon monoxide is independent of oxygen



(a) CO concentration (b) O_2 concentration

Figure 9 The profiles of CO and O_2 concentrations of the central longitudinal section and cross-section at 4% of blowing oxygen time. (a) CO concentration, (b) O_2 concentration.

concentration because of low gas temperature, and then off-gas combustion is prolonged. Therefore, CO emission concentration is high, and off-gas combustion is incomplete. In order to increase combustion effciciency, the amount of ambient-temperature air sucked in should be reduced.

Figure 10 shows the profiles of CO and O_2 concentrations at 14% of blowing oxygen time. Here, the gas temperature can reach above 2000°C (see Figure 8b) because of high-temperature off-gas entering, and thus the oxidation rate of carbon monoxide has a strong dependency on oxygen concentration. However, the simulating results indicate that the mixing of off-gas and air is poor, and CO and O_2 are separately concentrated in the stack center and the wall neighborhood. Therefore, the poor mixing of air and off-gas results in high CO concentration. In order to reduce CO emission, some methods should be taken to improve the mixing of off-gas and air and to reduce the combustion temperature.

Figure 11 shows the profiles of CO and O_2 concentrations at 87% of blowing oxygen time. The simulating results similarly indicate that the poor mixture of air and off-gas results in high CO concentration.

During pre- and postcombustion of off-gas, an improper air distribution mode of air sucked in and poor gas mixing may result in high CO emission. In order to increase the combustion efficiency of off-gas and to reduce CO emission, the air supply mode must be improved to effectively organize the mixing of off-gas and air in the cooling stack, and a large amount of excess air entering the cooling stack needs to be avoided. In different combustion stages, the air supply amount should be adjusted according to the off-gas flow rate.

NO Emission During Converter Off-Gas Combustion

Figure 12 shows the NO concentration at the outlet of the cooling stack. NO concentration can reach 150 ppmv at the precombustion stage of off-gas during

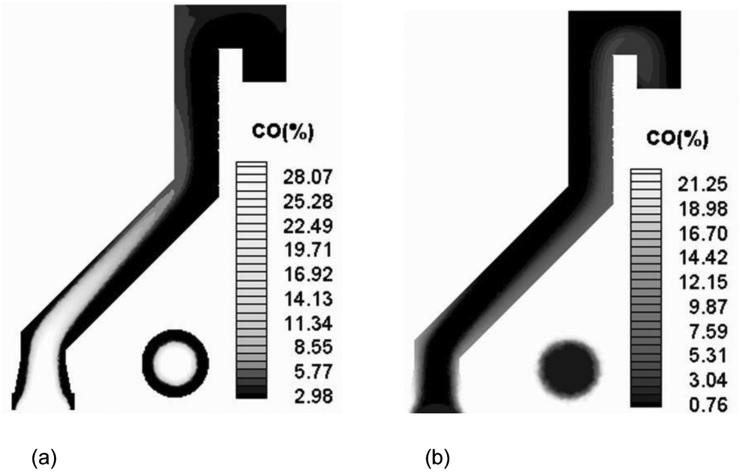


Figure 10 The profiles of CO and O_2 concentrations of the central longitudinal section and cross-section at 14% of blowing oxygen time. (a) CO concentration, (b) O_2 concentration.

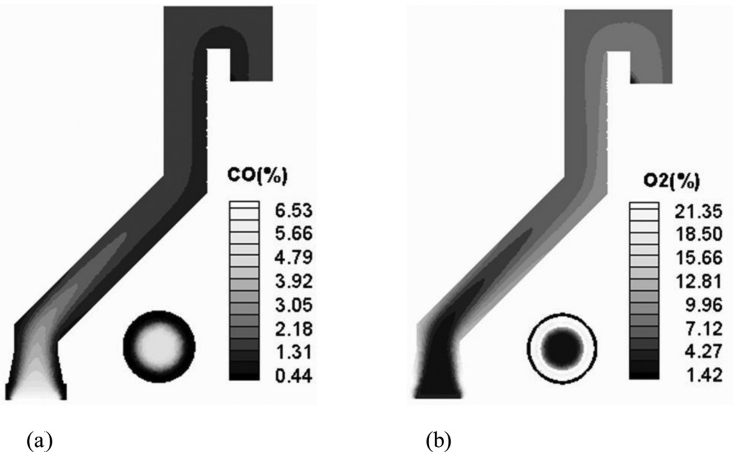


Figure 11 The profiles of CO and O_2 concentrations of the central longitudinal section and cross section at 87% of blowing oxygen time. (a) CO concentration, (b) O_2 concentration.

7–15% of blowing oxygen time and 10 ppmv at the postcombustion stage of off-gas during 83–87% of blowing oxygen time. The profiles of NO concentration at the above two combustion stages are provided in Figure 13.

At 14% of blowing oxygen time, compared with Figures 10b and 13a, the profile of NO concentration is similar to that of O_2 concentration. Thermal NO results from the oxidation of atmospheric nitrogen in high-temperature conditions, and the major factors are temperature, concentrations of oxygen and nitrogen, and residence time. In these four factors, temperature is the most important, and higher flame temperatures result in increased formation of thermal NO when the inlet temperature of off-gas is approximately up to 900°C, and then the combustion temperature can

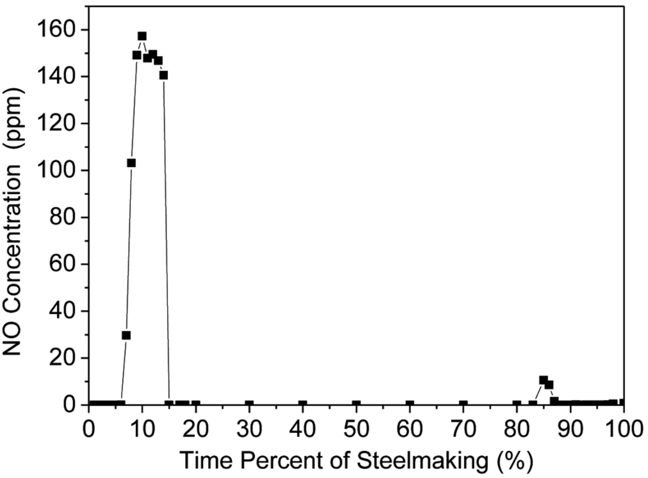


Figure 12 Concentrations of NO at the outlet of the cooling stack.

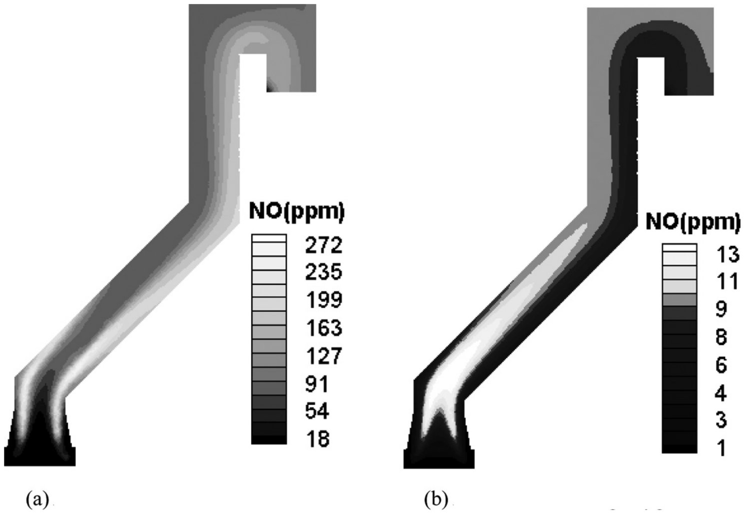


Figure 13 NO concentration profiles of the central longitudinal section at 14% and 87% of blowing oxygen times. (a) At 14% of blowing oxygen time, (b) at 87% of blowing oxygen time.

reach above 2000°C (see Figure 8b). At the moment, the concentrations of oxygen and nitrogen are crucial to NO formation, and higher concentrations of oxygen and nitrogen result in increased formation of thermal NO. At the later stage of pre-combustion (14% of blowing oxygen time), air flows along the cooling stack wall where oxygen concentration is high (see Figure 11b). As a result, a large amount of NO is formed along the stack wall.

Figure 13b shows the profile of NO concentration at 87% of blowing oxygen time, and NO concentration is relatively low (about 10 ppmv). Figure 11 shows the mixing of the off-gas, and the air sucked is poor. In this case, CO and O_2 are concentrated in the stack center and the wall neighborhood, respectively. Although O_2 concentration is high in the wall neighborhood, the rate of thermal-NO formation is still low because of low combustion temperature, and then a small quantity of NO is formed.

In practical steelmaking, off-gas entrains a large amount of fine dusts (FeO, Fe_2O_3, CaO, etc.). The amount of the dusts entrained by off-gas is about 80–150 g/m^3, and these metal oxides in dust have a catalytic effect on NO reduction when CO concentration is rich (14~84% of blowing oxygen time) (Hayhurst and Lawrence, 1997; Li, 2011).

As discussed above, the high combustion temperature of off-gas and the mixing of off-gas and air sucked in facilitate the formation of thermal-NO in the cooling stack during the pre- and postcombustion of off-gas. In order to reduce NO emissions, both the stoichiometric and temperature profiles in the cooling stack during off-gas combustion must be controlled. This control is achieved with design features that regulate the aerodynamic distribution and mixing of off-gas and air. In order to reduce thermal-NO formation, the air distribution mode should be improved to reduce combustion flame temperature by air stage.

CONCLUSIONS

In this article, CO and NO emissions are investigated during converter off-gas combustion by means of CFD. The simulating results indicate that CO concentration is above 0.5–2.0%. NO concentration can reach 150 ppmv at the precombustion stage of off-gas during 7–15% of blowing oxygen time, and 10 ppmv at the postcombustion stage of off-gas during 83–87% of blowing oxygen time.

During carbon monoxide combustion, the dependency of the oxidation rate on oxygen concentration is important to CO emissions and combustion efficiency. The improper air distribution mode of air sucked in and poor gas mixing result in high CO concentrations. In order to reduce CO emission, the air supply mode must be improved to effectively organize the mixing of off-gas and air in the cooling stack, and a large amount of excess air entering the cooling stack needs to be avoided. In different combustion stages, the air supply amount should be adjusted according to the off-gas flow rate.

The high combustion temperature of off-gas and the mixing of both off-gas and the air sucked in facilitate the formation of thermal-NO in the cooling stack during the pre- and postcombustion of off-gas. In order to reduce NO emissions, both the stoichiometric and temperature profiles of the combustion process must be controlled. At the same time, the air distribution mode should be improved to reduce combustion flame temperature by staged combustion using organized air introduction in the cooling stack during pre- and postcombustion of off-gas, and it can effectively strengthen the gas mixture.

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