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The chemical and physical effects of CO_2 on the homogeneous and heterogeneous ignition of the coal particle in O_2/CO_2 atmospheres

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Abstract

Oxy-fuel combustion technology is deemed a promising technology to the reduction of CO_2 emissions during fossil fuel combustion process and has been comprehensively investigated. Many researchers found that the ignition of pulverized coal delayed in O_2/CO_2 atmospheres compared to in O_2/N_2 atmospheres with an identical mole fraction of O_2 and attributed the ignition delay to the higher volume specific heat of CO_2 compared with N_2. This paper studied in detail the chemical effects and physical effects (specific heat, radiation characteristics and mass diffusivity) of CO_2 on both coal homogeneous and heterogeneous ignitions in O_2/CO_2 atmospheres. Jovanovic's ignition experiments (Jovanovic et al., 2011, 2012) were adopted to verify the simulated results. The artificial materials X, Y, Z and W were introduced to isolate the effects of chemical properties, specific heat, radiation characteristics and mass diffusivity of CO_2 during the homogeneous and heterogeneous ignition process of the coal particle, respectively (as can be seen in Table 3). The results showed that in the homogeneous ignition mechanism of coal in O_2/CO_2 atmospheres, the reasons for the ignition delay compared to the corresponding O_2/N_2 atmosphere were higher specific heat and chemical properties of CO_2. The chemical properties of CO_2 resulted in the ignition delay during the homogeneous ignition of pulverized coal in O_2/CO_2 atmospheres. High concentration of CO_2 resulted in the increase in the amount of heat absorption in the chemistry reaction system, especially R99 ($CO_2 + H \Leftrightarrow OH + CO$), which was an exothermic reaction in the X case, whereas an endothermic reaction in the CO_2 case. Higher specific heat, higher thermal radiation and lower mass diffusion coefficient of CO_2 played important roles in the

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ignition delay in the heterogeneous ignition of pulverized coal in O_2/CO_2 atmospheres compared to O_2/N_2 atmospheres.
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1. Introduction

Oxy-fuel combustion, in which the combustion of fuel occurs in an atmosphere of oxygen and recycled flue gas (RFG), i.e., O_2/CO_2, can produce a high CO_2 concentration in exhaust gas, which is almost sequestration-ready. It is almost feasibly by using existing technologies, reducing the risks for the implementation of new technologies. Therefore, it has been deemed a promising technology to obtain the reduction of CO_2 emissions in coal-fired power plants [1--6].

In oxy-fuel combustion, CO_2 substitutes for N_2 in the combustion atmosphere to moderate the high temperature produced by the combustion of fuel in pure O_2. Because the physical and chemical properties of CO_2 are very different from those of N_2, the characteristics and emissions of oxy-fuel combustion are significantly difference from air combustion [7--14]. Many studies on the ignition of pulverized coal in O_2/CO_2 atmospheres had been conducted by a thermogravimetric analyzer (TGA) [7--9], drop tube furnace (DTF) [7,10,11] and entrained flow reactor (EFR) [9--11], and the researchers had founded that the ignition of pulverized coal delayed in O_2/CO_2 atmospheres compared to in O_2/N_2 atmospheres with identical mole fraction of O_2. Many researchers attributed the ignition delay to the higher volume specific heat of CO_2 compared with N_2 [15--17]. The investigation performed by Masayuki et al. [18] showed that the flame propagation velocity of pulverized coal cloud in O_2/CO_2 mixture gas decreased about 1/3–1/5 of that in O_2/N_2 mixture gas at the same oxygen concentration, and they attributed these to the higher specific heat, smaller mass diffusivity of CO_2 as well as it absorbs radiation heat. However, they did not analyze the respective effects of these three physical properties on the flame propagation velocity in oxy-fuel combustion.

In fact, CO_2 is not passive diluents like N_2, rather, it can take part in some combustion reactions. Hirotatsu et al. [19] studied the NO_X formation in O_2/CO_2 and O_2/N_2 combustion by experiments and numerical simulations. He found that a high concentration of CO_2 greatly improved the reaction rate of the reaction $CO_2 + H \Leftrightarrow CO + OH$, which was closely associated to the generation of OH and O radicals that were beneficial of the oxidation of NH_3 and HCN. Song et al. [20] compared the temperature profiles of CH_4 combustion in air and in O_2/CO_2 atmospheres using numerical simulation in a counter-flow flame, and their results indicated that the chemical effects of CO_2 on the combustion temperatures were significant and even comparable with the physical effects. Consequently, the chemical properties of CO_2 can impact the ignition process of coal in O_2/CO_2 atmospheres. However, a few studies have been conducted on the chemical effects of CO_2 on the coal ignition in O_2/CO_2 atmospheres.

Moreover, there are three mechanisms of the ignition of pulverized coal [21,22]: homogeneous ignition mechanism, heterogeneous ignition mechanism and hetero-homogeneous ignition mechanism. For a homogeneous ignition mechanism, the primary step consists of the initial ignition of volatiles accumulated around the coal particle. Subsequently, the homogeneous combustion of volatiles ignites the char particle [23--26]. Heterogeneous ignition can occur via the direct attack of oxygen on the coal or char particle [22,27]. However, the effects of CO_2 on the coal homogeneous and heterogeneous ignition in O_2/CO_2 atmospheres have not been studied in detailed.

In this study, numerical simulation was used to analyze the effects of the physical and chemical properties of CO_2 on the homogeneous ignition and heterogeneous ignition processes of coal in O_2/CO_2 atmospheres. The modified CFD model proposed by Zou et al. [28] was used. And the artificial materials X, Y and Z, whose physical properties can be set according to needs and chemical properties are inert, were used to isolate the physical properties, such as the volume specific heat, mass diffusivity and radiation. The detail gaseous fuel reaction mechanism, GRI 3.0, was adopted to analyze the chemical effects of CO_2 on the homogeneous ignition of coal, and the CO_2 gasification was considered.

2. Modeling approach and validation of the CFD Model

2.1. Coal combustion models

Numerical simulations of the ignition process of coal in the study were carried out using the commercial CFD code ANSYS Fluent version 12. The coal combustion models had been described in detail in the previous paper [28].

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