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Homogeneous and heterogeneous contributions of CO_2 and recycled NO to NO emission difference between air and oxy-coal combustion

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ABSTRACT

High concentration of CO_2 and recycled NO-containing flue gas are known to affect the total NO emission during oxy-coal combustion through both homogeneous and heterogeneous reactions. To evaluate each contribution, this work investigated the NO emission behavior in air and oxy-coal combustion atmosphere (O_2/CO_2 and O_2/recycled flue gas) with three different oxygen concentrations (21%, 27% and 32%). Three typical Chinese coals and their derived chars were employed and all of the experiments were conducted in a drop tube furnace at a temperature of 1373 K. The results show that the conversion ratio of coal nitrogen (coal-N) to NO increased with decreasing coal rank in all atmospheres. The NO emission rate in oxy-coal was lower than that in air combustion. The heterogeneous effect of CO_2 mitigated NO emission in oxy-coal combustion, whereas homogeneous contribution of CO_2 may have increased NO formation due to a great amount of coal-N was released in the form of volatile nitrogen (volatile-N) at a high concentration of CO_2. The reduction of recycled NO was the major reason for the lower NO emission in oxy-coal combustion, but the main removal route differed. The reduction of recycled NO through homogeneous reactions was more important for both the lignite and the high volatile content bituminous coal, whereas the heterogeneous reactions were more important for the anthracite NO removal.

1. Introduction

Coal-fired power plants provide more than 70% of the electricity in China [1] but cause serious environmental issues such as global warming. Oxy-coal combustion technology, which uses pure O_2 and recycled flue gas (RFG) (mainly composed of CO_2) as a substitute for air, has been proven to be a promising way to control CO_2 emission from coal fired plants. The CO_2 may account for 95% (vol/vol) of flue gas during oxy-coal combustion, which afterwards can be captured economically and effectively [2–4]. It is worth noting that the pollutant emissions, e.g., NO_x, in oxy-coal combustion may differ from those in air combustion because recycled flue gas is used instead of N_2. Compared with air combustion, the NO emission in oxy-coal combustion with flue gas recycling is affected by the high concentration of CO_2 and recycled NO in the furnace [5,6].

Compared with the case of air combustion, the amount and composition of volatile nitrogen (volatile-N) would change in oxy-coal atmosphere due to the high concentration of CO_2. According to Sowa et al.'s experimental results ranged from 1600 to 1900 K, an American sub-bituminous coal released more volatile-N at a resident time of 15 ms in pure CO_2 than that in N_2 [7]. Duan et al. [8,9] found that during long residence time devolatilization (>150 s), CO_2 enhanced the conversion rate of coal nitrogen (coal-N) to volatile-N by releasing more HCN and less NH_3. Various possible reaction schematics are summarized in Table 1 from the reported literatures [5,8,10–14]. In regards to the influence of homogeneous impact of CO_2, Eq. (1) is thought to be the key factor because it changes the compositions of the O/H/OH radical pool. As a result, these composition changes could weaken NO formation from volatile-N [11,12]. The heterogeneous reactions between char nitrogen (char-N) and CO_2 could generate N_2 [14] or HCN [8] rather than NO in oxidation atmosphere. These reactions may decrease the amount of direct NO generation from char-N. It is not difficult to find that CO_2 could influence coal-N transformation through both homogeneous and heterogeneous reactions. However, the specific contribution of them is unknown.

Furthermore, recycled NO could also be reduced in the furnace by reacting with different reactants through homogeneous and heterogeneous reactions [5,13] (Table 1). NO reacts with hydrocarbons (CH_i), which are released at the beginning of devolatilization to form cyanide and amine intermediates (XN) through Eq. (2). NO converts into N_2 by reacting with XN as well, which is shown in

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