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EFFECTS OF SECONDARY AIR ON SO_2 EMISSION IN CIRCULATING FLUIDIZED BEDS

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ABSTRACT

Coal is a major energy source, and on the other hand coal is a source of pollution. Some of the important environmental issues related to use of coal in energy production are the release of sulphur dioxide (SO_2). SO_2 is a precursor to acid rain, responsible for atmospheric pollution and damage to human health. Therefore, investigations and development of technologies promising low SO_2 emission remains of high interest. Among various kinds of desulfurization technologies, sulfur removal in furnace is very competitive for controlling the SO_2 emission derived from coal combustion, due to the low capital and operating costs. One of the advantages of the fluidized bed combustion technology of coal is in situ SO_2 capture by added CaO-based sorbents, usually uncalcined limestone ($CaCO_3$).

Fluidized beds are operated with excess air. In the case of circulating fluidized beds, air-staging further contributes to a more reducing environment in the bottom bed. Although only the bottom bed is predominantly reducing, these reducing conditions may penetrate to some extent all the way up to the cyclone in streamers of oxygen-deficient gas. As a consequence of the large impact of reducing conditions on the sulphur capture process, the total air ratio and the extent of air-staging become important. From this point of view, in this study, the effect of air-staging on sulphur capture is investigated in a circulating fluidized bed via previously developed 2D model.

Keywords: fluidized bed, combustion, SO_2 emission, sulphur capture, air staging

1. INTRODUCTION

Reducing SO_2 emission from power plants is one of the main issues for the environmental protection. During atmospheric fluidized bed coal combustion the SO_x emissions derive essentially from the coal-S, and can be kept low by using either low temperatures or low excess air levels, together with in situ SO_x adsorption. The limestone addition is a recognized successful practice used to capture the SO_2 during coal combustion. However, it is reported that the efficiency of SO_2 removal by limestone is influenced by the operational conditions [1-7].

Fluidized beds are operated with excess air. However, as a consequence of the combustion, zones with a deficit of oxygen will occur locally [9]. Firstly, such zones will appear in the close vicinity of burning or devolatilizing fuel particles. Secondly, these zones may be extended because of imperfect mixing. One important example is the by-pass of air through the bubbling bottom bed, resulting in inadequate exchange of the gas in the particle phase. Another example is inadequate fuel distribution in large combustion chambers [10]. Insufficient secondary air penetration may be an additional source for inadequate mixing. Air-staging, if used, also promotes the formation of locally reducing conditions.

Measurements in the beds of circulating and stationary fluidized beds with a zirconia-cell oxygen probe, show that the environment can be reducing 80% of the time. In the case of a stationary fluidized bed, reducing conditions predominated even under unstaged conditions at an overall air-ratio of 1.4

[5]. Reducing conditions in the bubbling bed of the stationary and circulating fluidized beds are explained by the by-pass of air according to the two-phase model and depletion of oxygen in the particle phase due to combustion [5]. In the case of circulating fluidized beds, air-staging further contributes to a more reducing environment in the bottom bed. Although only the bottom bed is predominantly reducing, these reducing conditions may penetrate to some extent all the way up to the cyclone in streamers of oxygen-deficient gas [5].

As a consequence of the large impact of reducing conditions on the sulphur capture process, the total air ratio and the extent of air-staging become important [11]. The shift between oxidizing and reducing conditions, experienced by the adsorbent particles, and in particular the cycle time during which the process occurs, appears to influence the efficiency of SO_2 removal by limestone [5-8]. The effect of air-staging on sulphur capture has been studied in a circulating fluidized bed [5]. In addition to normal air-staging with about 60% primary air, two extreme cases were studied: no air-staging where all the air was primary air, and intensified air-staging where the primary air was lowered to about 45% and the level of secondary air addition was raised to 5.5 m. At 850°C the sulphur retention dropped to 40% under intensified air-staging, compared to about 90% under normal and no air-staging. The temperature dependence is clearly related to the degree of air-staging, and at a temperature of 930°C the sulphur retention was negative both for normal and intensified staging, indicating a net release of sulphur from the sorbent. Zirconia-cell oxygen-probe measurements verify that the fraction of time under reducing conditions is strongly dependent on the air-staging conditions [5]. From this point of view, in this study, the effect of air-staging on sulphur capture is investigated in a circulating fluidized bed (CFB) via previously developed 1D model.

2. MODELING

The present CFB combustor model can be divided into three major parts: a sub-model of the gas-solid flow structure; a reaction kinetic model for local combustion and a convection/dispersion model with reaction. The latter formulates the mass balances for the gaseous species and the char at each control volume in the flow domain. Kinetic information for the reactions is supplied by the reaction kinetic sub-model, which contains description of devolatilization and char combustion, and emission formation and destruction respectively.

In the modeling, the CFB riser is analyzed in two regions. The bottom zone (dense bed) is considered as a bubbling fluidized bed in turbulent fluidization regime and is modeled in detail as two-phase flow. The flow domain is subdivided into a solid-rare bubble phase and a solid-laden emulsion phase. The upper zone (dilute region) is considered as core-annulus flow structure.

Developed model includes devolatilization, attrition and combustion of a char particle respectively which also simultaneously predicts the carbon concentration, O_2, CO, CO_2, NO, SO_2, V.M. (volatile matter) distributions, particle size distribution, solid mass flux, and bed temperature values along the bed height. Another advantage is that the model takes into account the NO and SO_2 reduction which are major environmental pollutants. The model which simultaneously predicts both hydrodynamic and combustion aspects has been validated against the data from the literature [12].

Oxides of sulfur produced in burning the coal may be retained in solid form by reaction with particles of limestone or dolomite which is directly fed to the CFBC together with the solid fuel. At the combustion temperatures, usually in the range of 800–900°C, the $CaCO_3$ calcines to CaO and CO_2. The porous alkaline solid, CaO, produced by the calcination of limestone reacts with SO_2:

$$CaO + SO_2 + \tfrac{1}{2}O_2 \rightarrow CaSO_4 \tag{1}$$

Based on the stoichiometry of the sulphur capture reaction with calcium oxide, a theoretical limestone feed of one mole calcium per mole of sulphur would be enough for complete sulphur capture. However, the molar volume of the reaction product $CaSO_4$ is about three times greater than the molar volume of CaO, therefore complete conversion of the adsorbent particle is impossible, because sulphation only proceeds at the outer shell of the CaO particle and formation of $CaSO_4$ causes pore mouth closure and reaction stops before all the CaO is consumed by the reaction [13]. This sulfation pattern is commonly referred to as the unreacted-core model [14]. The Ca utilization of limestone is known to be highly dependent on the flue gas temperature and particle size. Several researchers have found that increasing particle size reduces the utilization significantly, and that the sulfur capture capacity passes through a maximum at temperatures between about 800 and 850°C [15]. As a result, Ca/S mole ratio is usually chosen between two and four in a classical fluidized bed combustor. On the other hand, high SO_2 retention efficiencies were obtained for Ca/S mole ratios of less than two in a circulating fluidized bed combustor [12].

In CFBC the SO_2 generation and retention processes take place simultaneously in the bed. In the model, it is also assumed that the particle size of limestone particles change during the sulphation reaction and the attrition of limestone particles are taken into account. Moreover, the estimation of limestone particles is assumed instantaneous. The reaction rate of a limestone particle can be expressed as [16]:

$$k_L = \frac{\pi}{6} d_s^3 k_{vL} C_{SO_2} \qquad (2)$$

where, k_{vL} presents the overall volumetric reaction rate constant and is a rapidly decreasing function of limestone conversion, d_s is the limestone particle diameter and C_{SO_2} is the SO_2 concentration in the combustion gases. The reactivity of limestone is a function of conversion, temperature and particle size. Calcium sulphate formed owing to the sulphation of calcined limestone tends to block the pores formed during limestone calcinations, building an impervious layer on the particle surface and reducing the reactivity of limestone.

3. RESULTS AND DISCUSSION

Model predictions about the influence of the air-staging on SO_2 emissions are shown in Fig.1 which plots the variation of the SO_2 emissions along bed height for three different excess air values.

In CFB combustor, the SO_2 generation and retention processes take place simultaneously in the bed. As mentioned above, the SO_2 generation rate from the char depends on its combustion rate, which depends on the temperature, excess air, O_2 concentration, etc. [17].

Fig.1 plots the predicted SO_2 emissions as a function of the dimensionless bed height at particle diameter 0.0651 cm, at bed operational velocity 3.60 m/s with excess air values 0.2, 0.4 and 0.6. The efficiency of desulphurization decreases as the excess air ratio increases [17]. The figure shows that a higher excess air level gives higher emission levels of SO_2 because formation of SO_2 increases with the increase of oxygen concentration. At the bottom zone, increase of excess air leads to higher concentrations of O_2 and this result causes SO_2 formation from volatiles, but also by a faster combustion which liberates the fixed sulfur as SO_2. The results shown in Fig.1, are also verified by the experimental study of Adanez et al. [2]'s.

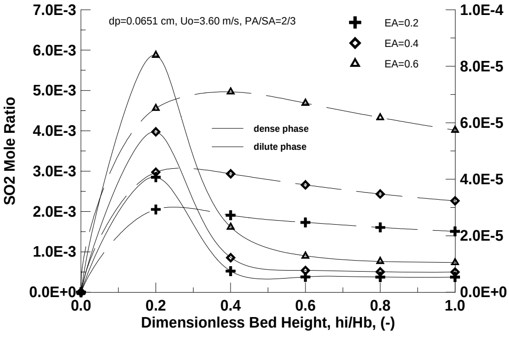

Fig.1. Effects of air-staging on SO_2 mole ratio at dilute and dense phase along the bed height.

4. CONCLUSION

In this study, the effect of air-staging on sulphur capture has been studied by previously developed a 1D model for a CFB combustor which integrates and simultaneously predicts the hydrodynamics and combustion aspects. As a result of this analysis, it is observed that: the higher excess air level gives higher emission levels of SO_2. The present study also proves that CFB combustion allows clean and efficient combustion of coal which is demonstrated by the fact that the model simulation results have low and acceptable level of emission pollutants.

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