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# NO<sub>x</sub> Emission from a Circulating Fluidized Bed Boiler Cofiring Coal and Corn Stalk Pellets

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**ABSTRACT:** The NO<sub>x</sub> emission from a circulating fluidized bed (CFB) boiler with 50 MW rated thermal output (50 MW<sub>th</sub>) during the cofiring of anthracite coal and pelletized corn stalk biomass was investigated. Cofiring could improve combustion efficiency and, thus, the thermal efficiency of the boiler. However, contrary to most results found in bubbling fluidized bed (BFB) combustors and the one found in a CFB boiler burning rice husk, NO<sub>x</sub> concentration in flue gas and fuel nitrogen conversion ratio increased with the biomass mass fraction. Several causes for such a contradiction were discussed and the configuration in combination with feedstock location of biomass was regarded as the most important one. In a CFB boiler, fuel is mostly burnt above the dense bed, opposite to that in a BFB boiler, and, thus, the average oxygen concentration above the dense bed, where biomass was fed, was higher. High bed temperature and high primary air ratio used for burning the anthracite coal increased NO<sub>x</sub> emission and enhanced the variation trend. In addition, the catalyst-contained ash also could promote NO<sub>x</sub> formation. The results indicated that biomass is not necessarily an effective cofiring fuel to reduce NO<sub>x</sub> emission in a CFB boiler and in order to reduce NO<sub>x</sub> emission, low primary air ratio, and locating the feedstock ports of biomass below the secondary air should be considered.

## 1. INTRODUCTION

Biomass fuels are renewable energy sources. The combustion of biomass fuels is regarded as CO<sub>2</sub>-neutral and can be taken as a primary measure to reduce greenhouse gases.<sup>1,2</sup> Because of the low heating value and low energy density, the collection, transport, and storage of biomass are costly. In order to overcome these problems, it is suggested that biomass, if applicable, can be crushed and then condensed into pellets. This treatment is of typical interest for corn stalk or straw utilization, which is rich in China and some other agricultural countries. The laboratory experiments showed that pelletized biomass is easy to ignite and burn with desired heat release density.<sup>3,4</sup> It is of great significance to find a way to optimally use this kind of fuel in industrial boilers.

It has been well-known that when biomass fuel is burned in a boiler, because of its high alkali metal content and low ash fusion temperature, corrosion and agglomeration problems are often encountered.<sup>3–5</sup> Therefore, cofiring of biomass with coal is often proposed.<sup>3–6</sup> It was found that as long as the mass fraction of biomass is less than a certain limit, cofiring can effectively increase the overall ash fusion temperature against burning pure biomass. In addition, because biomass has much greater volatile matter and char reactivity than coal, when it is cofired with coal, it can enhance the base fuel's ignition and burnout characteristics.<sup>6,7</sup> In recent years, it has been proved that circulating fluidized bed (CFB) combustion technology, which is widely applied in the utilization of low-grade coals, because of its the advantages of fuel flexibility and low pollutant emission, is effective for biomass cofiring.<sup>8–10</sup>

NO<sub>x</sub> and N<sub>2</sub>O emission from coal-biomass cofiring fluidized bed has attracted extensive research.<sup>2–5,8–19</sup> However, most research has focused on the formation and destruction mechanisms (e.g., refs 9, 10, and 13–15) or the experiments in the bubbling fluidized bed (BFB) (e.g., refs 7, 11, 16, and

17). Although some of the studies were conducted in the CFB, the installations used were mostly in laboratory or bench scale, and, so far, few studies have been conducted in industrial CFB boilers.<sup>2,3</sup> Moreover, because of the geographic availability, studies on the biomass cofiring in CFB boilers are diversified. In Europe and North America, several studies, even on industrial boiler scale, were conducted on cofiring wood-based and sewage sludge biomass,<sup>2,4,5,8,18</sup> but less on agricultural residues. However, China and other agricultural countries have much interest in the cofiring of corn stalk- and straw-based biomass.<sup>3,16,17,19</sup>

Nowadays, NO<sub>x</sub> emission from CFB boilers is stringently regulated. For example, in China, the standard for NO emission for a newly built CFB power plant is 100 mg m<sup>-3</sup> (standard state, 6% O<sub>2</sub>), although N<sub>2</sub>O is not regulated. Therefore, it is of particular interest to investigate NO<sub>x</sub> emission from cofiring the agricultural residues, of which the nitrogen content (fuel-N) can be higher than the base fuels and other biomass in a large-scale CFB. When these fuels are burned, it is expected that a great portion of the fuel-N could be converted to NO or N<sub>2</sub>O. Consequently, an experimental study was carried out on NO<sub>x</sub> emission during the cofiring of pelletized biomass and coal in an industrial CFB boiler, with a designated output of 50 MW<sub>th</sub> (here, the subscript "th" stands for thermal). Feasible methods to reduce NO<sub>x</sub> emission were discussed.

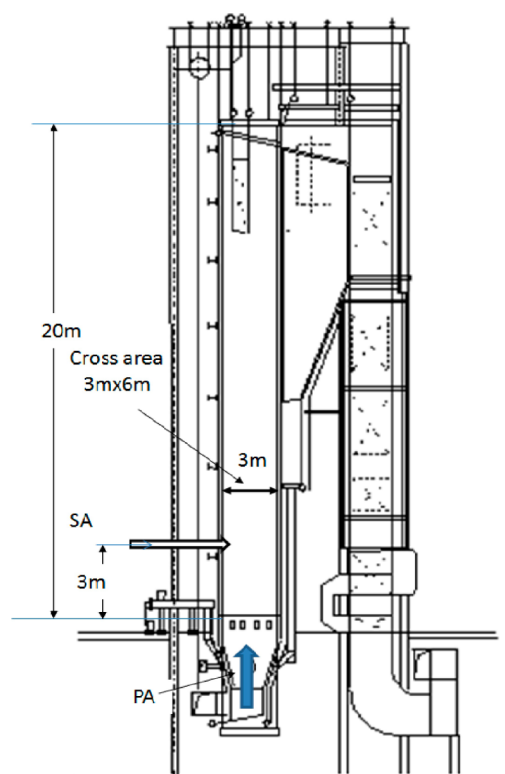
## 2. EXPERIMENTAL SECTION

**2.1. Test Facility.** The field test was carried out in a 50 MW<sub>th</sub> CFB boiler, the structure of which is shown in Figure 1. The boiler was a natural circulation type system with a single drum. The furnace,

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**Figure 1.** Schematic structure of the 50 MW<sub>th</sub> biomass-coal cofiring circulating fluidized bed (CFB) boiler.

cyclone, and vertical path of the boiler were made of the membrane tubes. The furnace was ~20 m in height and had a cross section of 3 m (width) × 6 m (depth). The furnace converged in the bottom section. The boiler was equipped with two water-cooled cyclones located on the same side. Each cyclone had a square cross-section, with an area of 3.0 m × 3.0 m. The cyclone leg was connected with a J-valve, by which the separated solid material was re-sent into the furnace. The  $d_{99}$  value, which represents the particle diameter at 99% separation efficiency of the cyclone, was 150–160  $\mu\text{m}$  and close to those of the cylindrical cyclone in the same capacity. The primary air was supplied from the bottom air distributor, and the secondary air was supplied from the ports located at 3 m above the air distributor. The bulk superficial velocity in the furnace was  $\sim 5 \text{ m s}^{-1}$ .

The boiler main steam pressure and temperature were 3.82 MPa and 723 K, respectively. The rated main steam flow rate was 75 tons per hour. The operational parameters, including the temperature, pressure, and air flow rate, were recorded and stored by the DCS (distributed control system). The test conditions are listed in Table 1. Three tests were conducted at 80% load and two were conducted at ~90% load. The coal and corn stalk pellets were mixed at the fuel yard and then loaded into a fuel silo located in front of the furnace and then fed into the furnace by a belt conveyer. The feed point was located ~4 m from the air distributor, slightly above the dense bed. The bed temperature was controlled between 1173 K and 1193 K, within a

**Table 1.** Test Conditions

mass mixing ratio, %	pelletized corn stalk	primary air ratio, %	bed temp, K	thermal output, MW <sub>th</sub>	O <sub>2</sub> in dry flue gas after cyclone, %
0	A	57.6	1184 ± 20	39.16	2.65
10	A	59.6	1175 ± 20	39.19	2.44
15	A	58.3	1174 ± 20	40.70	2.36
20	A	57.6	1189 ± 20	43.39	2.76
25	B	59.7	1194 ± 20	44.14	2.71

small range. The bed temperature was the average of values measured with K-type thermocouples at the points located at four different distances from the air distributor (0.56, 2.36, 13.26, and 19 m), as well as on both sides of the furnace. The primary air ratio varied between 57.6% and 59.7%, within a small range as well.<sup>3</sup> The primary air temperature was ~430 K, and the secondary air temperature was ~300 K.

The bed material mainly contained quartz sand and coal ash with bulk density of ~2400 kg m<sup>-3</sup>. Its particle size distribution is shown in Table 2, and the mean diameter was ~240  $\mu\text{m}$ . At ambient

**Table 2.** Particle Size Distribution of the Bed Material

size range $d_p$ , $\mu\text{m}$	percentage in mass $p_p$ , %
0–45	6.0
45–90	10.0
90–165	15.0
165–250	18.0
250–450	14.0
450–800	17.0
800–1200	9.0
>1200	12.0

temperature, the mean Archimedes number of the bed material was ~1330. During the operation, the bulk density in the free board section of the furnace was ~2–5 kg m<sup>-3</sup>.

The NO<sub>x</sub> concentrations and other flue gas components before the electrostatic precipitator (ESP) were measured under different mixing ratios, i.e., mass fractions of biomass ( $MR_b$ ) in the blended fuels with a Model G-40 portable gas analyzer. The measure point was located in the horizon part of the stack. The accuracy of the gas analyzer was approximately ±3 ppm, which is smaller than the amplitude caused by bed fluctuation. Corresponding samples of fly ash, bottom ash and circulating ash were collected and then analyzed in the laboratory. When circulating ash was collected, it was rapidly released and fully filled in a small container. The container then was immediately closed and cooled in the water to stop residue char combustion. The fly ash and bottom ash were collected in a standard way.

**2.2. Fuel Properties.** During the test, blended fuels of anthracite coal and pelletized biomass (corn stalk) in different  $MR_b$  values were used. Corn stalk pellets (CSPs) with different sizes, as shown in Figures 2a and 2b, were studied and are referenced as CSP-A and CSP-B hereafter. Both CSP-A and CSP-B had a cylindrical shape. CSP-A was ~40 mm in diameter and 60–100 mm in length, while CSP-B was ~6 mm in diameter and 20–40 mm in length. As listed in Table 3, besides the size, the biggest difference in the physical properties between CSP-A and CSP-B is density or voidage. The particle density of CSP-A is ~800 kg m<sup>-3</sup>, while that of CSP-B is ~1150 kg m<sup>-3</sup>. The nitrogen content is 0.9% in the anthracite coal and 1.5% in the corn stalk pellets.

### 3. RESULTS AND DISCUSSION

#### 3.1. Boiler Thermal Efficiency and NO<sub>x</sub> Emission.

Figure 3 depicts the variation of unburnt carbon content in the circulating ashes when  $MR_b = 0\%$ –25%. It clearly shows that, as  $MR_b$  increases, the unburnt carbon content decreases. Similar variation trends were found in the bottom ash and fly ash. The results indicated that higher boiler combustion efficiency could be achieved when the fuel contained a higher fraction of biomass.<sup>3,4</sup>

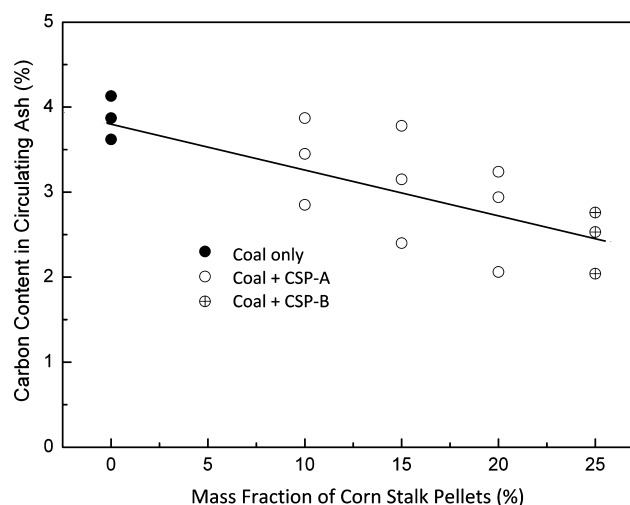
The variations of NO concentration in the flue gas, measured at the location before the ESP, with  $MR_b$  are shown in Figure 4. Consistent with the literature,<sup>10–14</sup> no NO<sub>2</sub> was appreciably found in all test cases and no N<sub>2</sub>O is expected to be remarkable, because of the rapid thermal and chemical decomposition at temperatures of ~1173 K.<sup>13–15</sup> Given that the bed temperature



**Figure 2.** Dimensions of the typical corn stalk pellets used: (a) CSP-A and (b) CSP-B. (Measurements are shown in units of centimeters.)

**Table 3. Properties of the Coal and Corn Stalk Pellets (CSPs)**

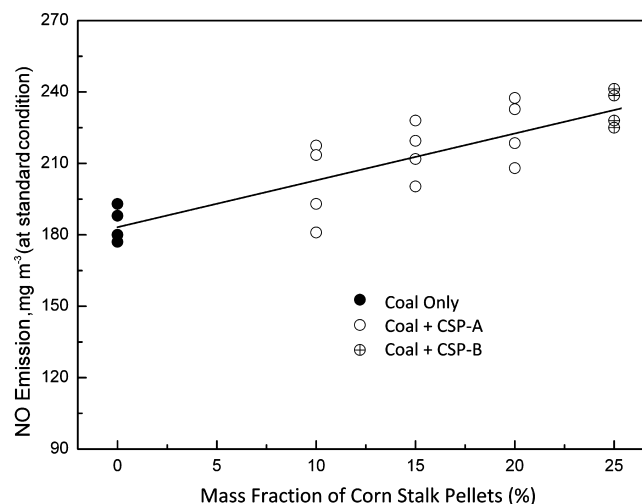
	unit	coal	CSP-A	CSP-B
particle density, $\rho_p$	$\text{kg m}^{-3}$	1380	800	1150
true density, $\rho_t$	$\text{kg m}^{-3}$	1490	1465.2	1411.7
voidage, $\epsilon$	%	7.4	46.1	28.2
proximate analysis (air-dry)				
fixed carbon, FC	%	47.6	6.9	8.7
volatile, $V_{ad}$	%	7.3	76.8	73.9
ash, $A_{ad}$	%	41.6	6.3	6.7
moisture, $M_{ad}$	%	3.8	10.0	10.6
lower heating value, LHV	$\text{MJ kg}^{-1}$	16.21	16.24	16.19
ultimate analysis (air-dry)				
$C_{ad}$	%	50.5	45.5	45.0
$H_{ad}$	%	0.9	5.5	5.5
$O_{ad}$	%	2.0	31.2	30.6
$N_{ad}$	%	0.9	1.5	1.5
$S_{ad}$	%	0.3	0.1	0.1



**Figure 3.** Variation of unburnt carbon content in circulating ash with mass fraction of corn stalk pellets in the fuel feedstock: (●) coal, (○) coal + CSP-A, and (⊕) coal + CSP-B.

difference could be neglected during the entire experiment, NO emission increased with  $MR_b$ .

As shown in Figures 3 and 4, the results of CSP-B cofiring fall on the variation trends of CSP-A cofiring. It was observed that the larger and looser CSP-A fragmented more seriously,



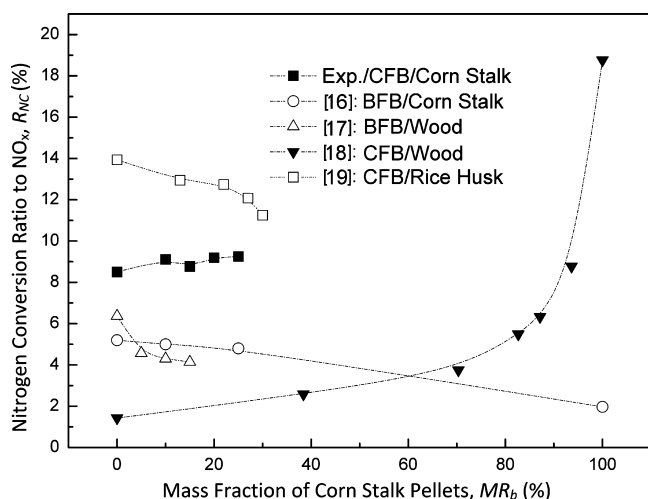
**Figure 4.** NO emission in the dry flue gas at different mass fraction of corn stalk pellets in the fuel feedstock (converted to 6%  $O_2$  base): (●) coal, (○) coal + CSP-A, and (⊕) coal + CSP-B.

while the smaller and denser CSP-B mixed better with the bed materials in the furnace. The bed temperature measured above the dense bed fluctuated more when CSP-A was burned, but as shown in Table 1, the variation range was mostly within  $\pm 20$  K. Approximately, under the present test conditions, the influence of physical properties (size and density) was not remarkable for these pellets, which were made of the same kind of corn stalk.

**3.2. Nitrogen Conversion into  $NO_x$ .** Since the cofiring changes the nitrogen content of the fuel feedstock, it is more reasonable to use the nitrogen conversion ratio (the mass ratio in percentage of the fuel nitrogen converted into over the total fuel nitrogen, denoted as  $R_{NC}$ ) instead of NO concentration in flue gas, to evaluate the effect of cofiring on NO emission. The total fuel-N in the present study was based on the measurement of elemental nitrogen in the fuel. The converted mass into NO was based on the NO concentration measured in the flue gas. The measurement error was within 5%, given that the NO concentration was not very small.

The variation of  $R_{NC}$  with  $MR_b$  in the blended fuels in the present study and the literature<sup>16–19</sup> are compared in Figure 5. Studies in a bubbling fluidized bed (BFB) combustor when lean coal or bituminous coal were burned as the base fuels, regardless of whether the nitrogen content of the biomass was higher or lower than that of the base fuel,  $NO_x$  decreased with the increasing biomass mass fraction.<sup>16,17</sup> The referenced





**Figure 5.** Variation of nitrogen conversion ratio with the mass fraction of corn stalk pellets in the fuel feedstock: (■) present study, (○) ref 16, (△) ref 17, (▼) ref 18, and (□) ref 19.

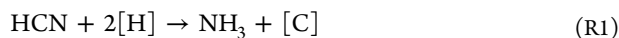
studies used sawdust,<sup>17</sup> wood chips,<sup>18</sup> or rice husk<sup>17,19</sup> as the cofired biomass. The coal-wood chips cofiring experiments were conducted in a 12 MW<sub>th</sub> CFB combustor at ~1123 K, and the base fuel was bituminous coal with a nitrogen content of 1.5% (in ash and moisture-free base) and the wood biomasses had nitrogen contents of ~0.1%–0.2%. The referenced CFB cases also include a bench-scale experimental study in which rice husk with nitrogen content of 0.55% was cofired with a bituminous coal with nitrogen content of 1.16% (dry base).<sup>19</sup>

As shown in Figure 5,  $R_{NC}$  increases with  $MR_b$  in the present study. This trend agrees with the trend determined by Leckner and Karlsson in a CFB boiler,<sup>18</sup> but it contradicts those obtained in BFB combustors and a bench-scale CFB.<sup>19</sup> This phenomenon is very important for the application of biomass and, thus, is further discussed.

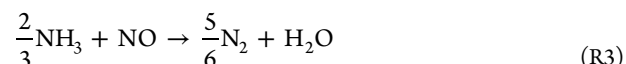
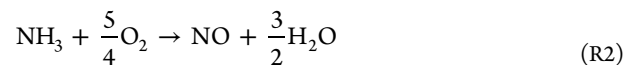
In this study, the biomass was corn stalk and its nitrogen content was higher than that of base fuel; thus, the average nitrogen content increased when more corn stalk was burned. However, found by the previous study,<sup>16</sup> regardless of whether the average nitrogen content is higher or lower than the base fuel, it does not change the variation trend of  $R_{NC}$  verse  $MR_b$ . Thus, the average nitrogen content might not be a prevailing reason for the above phenomenon, and, instead, the reasons should be attributed to the difference in boiler type and special operational conditions.

The most important reason could be attributed to the globally fuel-lean dense bed in the CFB boiler, against the globally fuel-rich dense bed in a BFB boiler.<sup>20,21</sup> In a BFB boiler, the fuel is mostly burned in the dense bed, while it is opposite in a CFB boiler. As a result, the average oxygen concentration in the upper furnace is higher in a CFB boiler. For both types of boiler, most of the fuel including biomass is normally fed into or slightly above the dense bed. This difference greatly affected the NO formation.

As the fuel is fed into the furnace, NO is from oxidation of HCN and NH<sub>3</sub>, which are produced mostly from the amino acid containing fuel-N in the pyrolysis process.<sup>22</sup> Furthermore, NH<sub>3</sub> can be regarded as the primary product of fuel-N, directly from pyrolysis or the secondary conversion from HCN through reaction R1.<sup>10–14,23–25</sup>

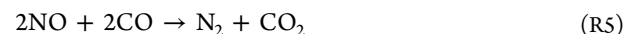


Depending on temperature and oxygen concentration, NH<sub>3</sub> could be partly oxidized to NO (reaction R2) or acts as a reducing agent to reduce NO into N<sub>2</sub> (reaction R3).



In a cofiring BFB boiler, because of the globally fuel-rich atmosphere in the dense bed zone, the increasing NH<sub>3</sub> concentration would be in favor of NO reduction (reaction R3). However, in a cofiring CFB boiler, the volatile nitrogen released will be mostly in an oxidizing atmosphere and then rapidly oxidized into NO through reaction R2.<sup>10,26</sup> The nitrogen retained in the char is of small fraction and mostly reduced to N<sub>2</sub> in the upper combustor regions by CO and char.<sup>13,27</sup> Therefore, with the increase of  $MR_b$ ,  $R_{NC}$  increases.

At the same time, NO reduction in the upper furnace for both types of boilers is also different. In a BFB boiler, a small amount of solid particles exist above the dense bed and most fuel is burned in the bottom furnace.<sup>28</sup> So does the volatile matter released from the biomass. As a result, reaction R2 is hindered, while reaction R3 (i.e., NO reduced into N<sub>2</sub>) is enhanced. Meanwhile, because the gas solid reaction is weaker, there could be more biomass char with porous structure to reduce NO into N<sub>2</sub> (reaction R4).



In a CFB boiler, combustion largely occurs in the upper furnace<sup>28</sup> and thus, a large amount of NO is reduced by CO and char via reactions R4 and R5.<sup>6,18</sup>

The reaction rates of reactions R4 and R5 are mainly determined by char and CO concentrations. As found by Leckner and Karlsson,<sup>18</sup> when coal only was burned, the carbon content in the furnace was ~10 times higher than that when biomass only was burned. Consistently, as shown in Figure 3, the unburnt carbon content in the circulating ashes decreases as the  $MR_b$  value increases, resulting in less NO reduction in the upper furnace in the CFB boiler.

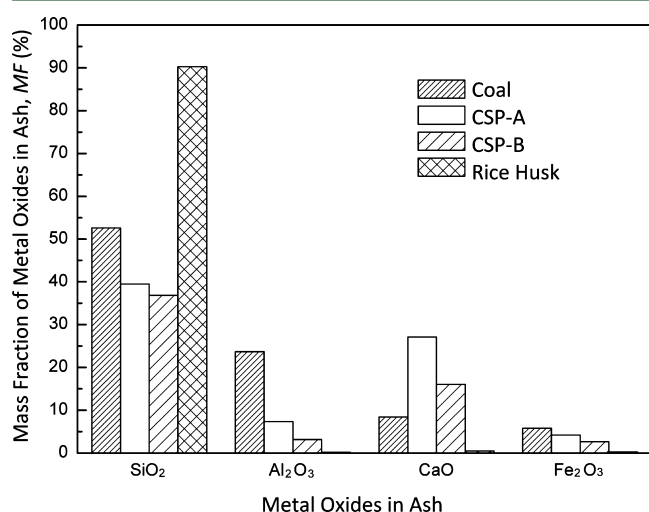
Anthracite base fuel and primary air ratio used in the CFB boiler operation was enhanced above the NO conversion trend. When anthracite coal was burned, the heat-release fraction became even larger in the upper furnace and the oxidizing atmosphere became more severe near the feedstock ports in the bottom furnace. As discussed above, according to reactions R1–R5, the oxidizing atmosphere was in favor of NO formation. In the same time, when anthracite coal is burned, the bed temperature used is usually higher than that of the other coals, and thus more NO could be formed.<sup>16–19</sup> Compared with the experiments reported in the previous study,<sup>18</sup> the bed temperature used this study was 1173–1193 K, which was 50–70 K higher, and the emitted NO concentration was ~3 times higher.

Air staging is also important to the NO formation and reduction.<sup>18</sup> In this study, biomass was fed with coal right above the dense bed. Because the bottom furnace wall was insulated with refractory material, to avoid overheating, more primary air than usual was introduced during operation. Thus, the amount of the secondary air injected above the dense bed became less.

As a result, the reducing atmosphere near the biomass feeding became weaker.

**3.3. Effect of Ash Composition on NO<sub>x</sub> Emission.** The present results also contradict the earlier reported results obtained from a CFB apparatus burning rice husk.<sup>19</sup> One reason was that the biomass used in their experiments was rice husk with a nitrogen content of 0.55%, which is lower than that of base fuel, which was 1.16%. The other reason, more importantly, might be the different ash composition of the biomass used in their study against the present one.

Some ash compositions may act as a catalyst in the formation of NO<sub>x</sub> precursors and NO formation.<sup>29–32</sup> It was found that the interaction of ash could increase the yield of NH<sub>3</sub> and decrease the yield of HCN.<sup>29</sup> The ash compositions of the coal and biomass used in present experiments and the similar rice husk as that used in the referred study were analyzed via XRD. The SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, CaO, and Fe<sub>2</sub>O<sub>3</sub> contents are shown in Figure 6. It can be seen that the ash of CSP-A and CSP-B are rich in



**Figure 6.** Composition of coal ash and biomass ash in mass percentage (%).

CaO, which is believed to be an active catalyst for NH<sub>3</sub> oxidation.<sup>14,15,30–32</sup> While the rice husk ash used in the previous study<sup>19</sup> was rich in SiO<sub>2</sub>, it lacked CaO, so the value of  $R_{NC}$  decreased as the rice husk mixing ratio decreased.

#### 4. CONCLUDING REMARKS

NO<sub>x</sub> emission from a circulating fluidized bed (CFB) boiler with 50 MW<sub>th</sub> cofiring anthracite coal with pelletized corn stalk biomass was studied. It was found that blending biomass in coal enhanced the overall combustion efficiency and, thus, gave a higher boiler thermal efficiency. Meanwhile, NO concentration in flue gas and nitrogen conversion ratio ( $R_{NC}$ ) increased with mass fraction of corn stalk pellets ( $MR_b$ ). The result was contradictory to that found in BFB (bubbling fluidized bed) boilers and that in a CFB boiler burning rice husk biomass.

Based on the analyses of possible chemical reactions, the main reason for such a variation trend was strongly attributed to the configuration in combination with feedstock port locations in the CFB boiler. The high primary air ratio and bed temperature used for the anthracite burning also increased NO<sub>x</sub> emission and enhanced the variation trend of  $R_{NC}$  with  $MR_b$ . The higher nitrogen content of biomass, compared to that of base fuel, and the catalytic effect of ash composition were

also in favor of NO<sub>x</sub> formation. The results indicated that the attempt to use corn stalk pellets to reduce NO<sub>x</sub> emission in a CFB boiler is not necessarily effective.

Based on the test results and discussion, for a cofiring CFB boiler, NO<sub>x</sub> emissions can be reduced by optimizing the oxygen concentration and temperature in the bottom furnace where the biomass was fed and burned. A small primary air ratio and low bed temperature in the bottom furnace should be used. Besides, biomass is suggested to be fed from separated feedstock ports below the secondary air or directly in the loop seal. In the future study, N<sub>2</sub>O emission during the cofiring of biomass and coal in a CFB boiler should also be given attention, although only NO<sub>x</sub> is regulated in the current emission control.

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##### Notes

The authors declare no competing financial interest.

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