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Daood, S.S., Javed, M.T., Gibbs, B.M. et al. (1 more author) (2013) NOx control in coal combustion by combining biomass co-firing, oxygen enrichment and SNCR. Fuel, 105. 283 - 292. ISSN 0016-2361

https://doi.org/10.1016/j.fuel.2012.06.087

NO_x Control in coal combustion by combining biomass co-firing, oxygen enrichment and SNCR

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Abstract:

There has been renewed interest in evaluating the effect of biomass co-firing on the multi-pollutant control system such as Selective Non-catalytic Reduction (SNCR) and oxygen enrichment. Emissions savings have been attained by combining SNCR and biomass-coal co-combustion under various oxygen enriched staged air levels. Biomasses with higher tendency of generating CO produced better reduction in NO_x emission with and without using SNCR. NO reduction of around 80% were attained using SNCR for 15% and 50% blending ratios of biomasses at 21% overall O_2 concentration for unstaged combustion. Whereas, a range of 40%-80% NO reductions were attained for RC2 (a Russian Coal) and 15% co-fired biomasses with 3.1%-5.5 % overall O_2 concentration at 22%-31% levels of flame staging. Moreover, it was found that better NO_x removal efficiency was attained for higher NO_x emission baselines under both oxygen enriched and normal firing conditions. However, SNCR NO_x control for both coal or coal-biomass blends was observed to produce higher NO_x reductions during O_2 enrichment, believed to be due to the self-sustained NO_x reduction reactions. Hence, NO_x control by SNCR, oxygen enriched co-firing in the furnaces would result in lower NO_x emissions and higher carbon dioxide concentration for efficient scrubbing with better carbon burnouts.

Key words: NOx, SNCR, biomass cofiring, coal

1.Introduction

Some conventional coal fired power stations of Europe are under threat of closure, due to enforcement in 2020 (previously 2016), of the Large Combustion Plant Directive (LCPD) [1]. This is due to the economics of implementing control technologies to reduce the emission of NO_x to lower than 200mg/Nm^3. Moreover, UK is also expected to fail in meeting the NO_x emissions ceiling target set by the European National Emissions Ceiling Directive (ENECD) [2]. Hence, the ENECD is reviewing to produce new emission ceilings targets for 2020 [3]. The studies related to hybrids of different existing technologies such as co-combustion, oxy fuel combustion, SNCR and selective catalytic reduction (SCR) can potentially meet the emissions reduction goals efficiently and economically[4-5]. In this paper, the effect of firing coal and coal-biomass blends in normal air and oxygen enriched conditions were studied, using ammonia as a reducing agent for control of NO_x.

A number of comprehensive reviews and research articles have already been published signifying the developments and findings related to co-combustion, oxy-fuel and SNCR application, for control of combustion generated nitrogen oxides (NO_x) [6-9].The effect of oxygen enrichment for NO_x emission from coal and coal-biomass combustion under deeply staged configurations has already been discussed in detail [10-11]. This study extends these concepts for exploitation under SNCR conditions in a 20 kW combustion test facility of University of Leeds (UK). Initially the effect of addition of biomass blends in coal with and without SNCR is studied. Moreover, deep staged oxygen enriched conditions were also applied with and without SNCR, in order to give a comparison between the two operating configurations

The impact of O_2 concentration and other species on the SNCR performance have been reported separately or in combination. The chemical kinetic modelling using sodium carbonate under 4% O_2 concentration showed promoted SNCR. The increased concentration of sodium salts was also found to enhance the performance of SNCR process with widened temperature range at 3.9% O_2 concentration for the performed experiments [12]. Similarly, NO reduction efficiency at 1.7% O_2 concentration was found lowered than at 3.8% O_2 concentration in a separate study [13]. Moreover, an existence of conversion temperature point (CTP) has also been discussed, on the two sides of which O_2 performs differently. It was found that below 1000^{o}C, higher NO reduction was reported for higher O_2 concentration due to formation of more radicals to drive the sustained chain reactions by increasing the rate of $H + O_2 \Leftrightarrow OH + O$ and $O + H_2O \Leftrightarrow OH + OH$ reactions [14]. However, there is a need to do more investigations especially on the effect of O_2 enrichment on the performance of SNCR process for coal-biomass co-fired pulverised combustors due to limited available published literature on such hybrid configurations.

Moreover, it is believed that if the biomass-coal co-fired power generation units are equipped with retrofit able oxygen enhanced combustion (OEC) and SNCR processes, the results can easily far outweigh the benefits of SCR with better carbon burnouts, plant efficiencies and emission levels [10-11, 15-20].

2.Experimental

The experimental detail of the test facility has already been discussed in previous published articles [10-11, 19, 21], especially detailing the method of calculations regarding oxygen enrichment[11]. All tests were carried out in a 20 kW (thermal) down-fired, refractory-lined, furnace (Fig. 1). The height of the furnace is 3.5 m and constructed of nine modular sections of varying lengths with an internal diameter of 200 mm. Located along the length of the furnace are a number of utility ports for the injection of oxygen enriched over-fire air (OFA), gas sampling and temperature measurements. The location of thermocouples and ports are also elaborated in Table 1. Optimisation of the SNCR process (discussed in detail in 4.2.1), in order to inject ammonia at the correct temperature for effective reduction resulted in the selection of port 6. A water-cooled injection probe was used in all the tests in order to avoid thermal decomposition of ammonia before entering in to the furnace.

The measurements of CO_2, O_2, NO_x, CO and SO_x were taken using standard gas analysers and recorded on data logger. The readings were averaged over a typical period where the levels were uniform especially when the respective temperatures were observed steady. Standard deviation values were calculated in order to carefully process the data (Table 1).

In OEAS combustion tests, all the oxidants and fuel were delivered into the furnace using the same configurations as those used in coal-air combustion firing tests. The continuous coal or coal-biomass feeding during changing over from air combustion to oxygen enrichment minimizes any errors resulting from coal or coal-biomass feeding rate variations between the two combustion configurations. The reported combustion gas temperatures were also averaged over a period of time when their values were observed to be steady.

3. Fuel Characterization

Table 2 contains the proximate and ultimate analyses along with pyrolysis gaseous products measured using thermogravimetric analyser (Shimadzu TGA-50), CE Instruments Flash EA1112 series and pyro-probe attached with an on line gas Chromatograph, respectively. The different nature of biomass from coal is not only apparent from the values of O/C, H/C, and GCV but also from the flash pyrolysis products (CH_4, CO, H_2) emitted from the raw samples. The produced species at $1200^{o}C$ clearly depict the differences between coal (RC1, RC2) and the biomass, emitting relatively high amounts of CO and low amounts of CH_4 and H_2. It is believed that for different configurations of fuel / air staging the evolved species especially CO, play a major role in reducing the NO emissions through the reactions beneficial towards enhanced NO reduction [22-23].

Comparison of the different fuels revealed a number of differences between the coal and biomass. The major difference is attributable to the volatile matter and fixed carbon contents of the fuels. The ash content in RC1 and RC2 on the other hand, is much higher than that in the biomass samples excluding SB2. This influences the combustible content and calorific value of the fuels. Biomass fuels produced lower char yields and bulk densities due to their higher volatile matter content and higher char surface areas as evident from Table 3. Biomass samples contain higher proportion of oxygen, hydrogen and less carbon, effectively reducing the heat content within these fuels as is obvious from Table 2. This is attributable to less energy contained in carbon-oxygen and carbon-hydrogen bonds as compared to carbon-carbon bonds [24]. Hence, RC1 was observed to contain approximately 52%, 56%, 57% more heat content compared to CS, SB3 and SM, whereas 134% and 65% higher compared to SB2 and W respectively. Similarly this difference of heat content amongst the biomass samples and RC2 was slightly more due to its higher FC as compared to RC1. The higher VM in the biomass is expected to accelerate the combustion process. Amongst the coal samples the RC2 contained about 2% higher VM & FC but 3% less ash matter. This difference in the VM may suggest a slight difference in the reactivity of RC2 from RC1. Table 3 represents particle size distribution, weight equivalent share corresponding to thermal share of biomasses with RC1 and RC2. Moreover, the calculated ratios of volatile matter (VM) to fixed carbon (FC) are presented in both Table 3.

4.Results and Discussion

4.1. Unstaged co-combustion results without SNCR

Unstaged co-fired results without SNCR produced substantial NO_x reduction especially with increasing biomass input shares up to a VM/FC ratio of 1.8 [26]. Fig. 2 does show an ongoing decrease of NO emissions but the impact certainly reduces beyond VM/FC ratio of about 1.8 for almost all the biomass fuels. Under unstaged co-combustion conditions without SNCR despite the equal or higher fuel-N content of the biomass relative to the coals, co-fired coal-biomass fuel blends emitted lower NO levels especially beyond 15% blends; this is evident from SM-RC1 and SM-RC2 co-combustion results beyond 15% input share as shown in Table 4. Similarly, coconut shell (fuel-N content of 1.2%) though greater than Bituminous coal (fuel-N of 0.91%) resulted in lower NO emissions. Moreover, lowered NO emissions were found with high fuel-N carrying biomass [27-28]. Hence it is believed that the lower NO emissions can be achieved even with high fuel-N content carrying biomass when combusted with coal or as replacement of coal. It is believed that with addition of biomass, gas phase combustion reactions become dominant due to faster devolatilization, liberating biomass fuel-N as NH_3 which can either convert to NO or act as reducing agent in further reactions with NO to form N_2. Nonetheless, NO, after formation, may also be recycled through hydrocarbon radicals to cyanide or reduced to N_2 by surface reactions on char classed as heterogeneous NO reduction reactions [29-31]. Table 4 summarizes the effect of combustion of different blending ratios of biomasses with both RC1 and RC2 on % fuel mix-

N, VM/FC and NO emissions. The NO reductions were observed to be higher for biomasses with higher CO emissions emitted during the prolysis gaseous product analysis (Table 2, 4). Hence it can also be concluded that for different configurations of fuel / air staging the evolved species especially CO, play a major role in reducing the NO emissions through the net gain in OH radical pool beneficial for NO reduction. Moreover, RC2, despite having higher FC than RC1 produces lower NO emissions especially without SNCR. This is believed to be due to difference in the reactivity of RC2 from RC1. Hence, it can be summarized that addition of biomass does not impede NO_x reduction.

4.2. Unstaged co-combustion results with SNCR

4.2.1. Injection of Ammonia for Selective Non Catalytic Reduction (SNCR)

The experimental results reported here include the optimization of location for ammonia injection, nitrogen stoichiometric molar ratio (NSR) and the utilization of optimized parameters directed towards the 15% thermal blends of biomass-coal.

It is apparent that the effectiveness of injected ammonia depends upon how efficient the mixing takes place inside the furnace. In order to enhance the mixing of ammonia with NO present in the flue gas stream, different flow rates of the carrier medium (i.e. nitrogen) were tested as illustrated in Fig. 3. The concentration of NO emissions after ammonia injection for the NSR = 1.5 (where NSR is defined as the molar ratio of ammonia to NO) dropped from 201ppmv to 131ppmv as the carrier gas flow rate was increased from 0 to 20 litres per minute. The effectiveness of the inert nitrogen carrier was not significant beyond 20 litres per minute as evident from Fig. 3. Hence 20 l/min flow rate of nitrogen was selected as a carrier flow for SNCR experiments.

The effect of varying the sample port position for NH_3 injection on NO reduction and temperature in case of RC1 combustion is shown in Fig. 4 for NSR=1.5. The temperature range with in which ammonia is most likely to react, causing optimum net reduction of NO is usually defined as a temperature window. Usually 800°C is selected as the lower limit of temperature below which the reaction between the injected NH_3 and NO_x is too slow to cause an appreciated NO reduction. Thus un-reacted ammonia leaves the furnace adversely affecting the ammonia utilization efficiency. On the other hand at higher temperatures (>1200°C), NH_3 tends to oxidise to form NO rather than reducing it to N_2 [9]. However the effect of increasing temperature on NO emission for NSR = 1.5 using ammonia as SNCR reagent has also been indicated [32].

Within the range of injection temperatures, the optimum NH_3 position resulting in 84% NO reduction is 230cm away from the burner (i.e. port 6). When the NH_3 was injected further downstream in furnace the lower gas temperature is believed to have slowed the rate of reactions beneficial for NO reduction. The temperature range of 1025^oC to 950^oC is estimated, from this work, to be the optimum temperature window within which the homogenous gaseous phase reactions between ammonia and NO have given highest NO reduction. This temperature range is in agreement with the optimum NO reductions quoted in the literature review [9]. The residence time for the optimum temperature window, based on the plug flow reactor assumptions, is 0.2s (Equation 1).

$$\tau = \frac{(L)}{(Q / A)} \qquad (1)$$

Where τ is the residence time of ammonia in seconds (within the optimum temperature window), L is the length of the furnace across which the optimum temperature window exists (i.e. = 0.15m), Q is the volumetric flow rate of the flue gases in m^3/s and A is the area of flow of the furnace in m^2. The NH_3 utilization efficiency (ηNH_3) (i.e. the amount of ammonia added that reduces NO to N_2) reported in Fig. 5 is calculated via the Equation 2. The injected ammonia flow rate (i.e. ammonia used to reduce NO) is calculated by Equation 3.

$$\eta NH_3 = \frac{(NO_{initial} - NO_{final})}{(NO_{initial} \times NSR)} * 100 \qquad (2)$$

$$AFR = (NO_{initial} \times 10^{-6}) \times Q \times NSR \qquad (3)$$

$NO_{initial}$ and NO_{final} are NO emissions before and after ammonia injection, respectively, AFR is ammonia flow rate in millilitres per minute (ml/min) and Q is the volumetric flow rate of air in ml/min.

The effect of varying the NSR on the NO reduction efficiency and ammonia utilization efficiency is also illustrated in Fig. 5. The effectiveness of added ammonia is limited beyond a certain value of NSR. Initially the molar ratio of ammonia to NO (i.e. NSR) is beneficial to achieve higher NO reductions but beyond 1.5 to 3 no significant reduction in NO occurred.

Moreover, the ammonia utilization efficiency continued to decline beyond the 1.5 value of NSR. This over all suggests 1.5 to be the optimum molar ratio of ammonia to NO which can be used for the optimized injection location (i.e. port 6).

Fig. 6 shows the effect of addition of ammonia with 15% and 50% thermal based blends of biomass in coal. The optimized values of NSR= 1.5, port position 6 and 20 l/min nitrogen carrier are used for co-combustion experiments. The results indicate the maximum benefit of NO reduction for SB2 and SM. This is probably due to higher initial NO emissions for 15% blending ratios (i.e. 813ppmv and 842ppmv) in the cases of SB2 and SM as compared to 669ppmv, 635ppmv and 709ppmv of CS, SB3 and W biomass blends respectively. The highest initial NO emission of SB2 and SM resulted in 85% and 86% NO reductions as compared to 81%, 79% and 82% for CS, SB3 and W respectively. The ammonia utilization efficiency for the 15% blending ratios varied within the range of 52% to 57%. On the other hand, the 50% blending ratios resulted in 83% and 84% NO reductions for the SB2 and SM as compared to 76%, 71% and 74% for CS, SB3 and W co-fired coal-biomass blends, respectively. These NO reductions for CS, SB3 and W are less than the 15% thermal blends. This is because of lower initial levels of NO emissions giving in lower NO removal efficiency. The lower initial NO emission levels observed were 546ppmv, 449ppmv and 491ppmv for CS, SB3 and W, respectively.

Higher NO removal efficiency corresponding to the higher initial NO emissions for varying retention times; and higher impact of SNCR has been reported for both cases of fuel lean and fuel rich SNCR at higher initial NO emissions [33]. The utilization of ammonia injection, in case of 15% blends of biomass with coal, has proven to be better due to higher NO removal and NH_3 utilization efficiencies.

4.2.2. Impact of SNCR on NO reduction in OEC conditions

OEC tests performed on the same test facility has proven to produce better carbon burnouts published elsewhere [10-11]. The application of SNCR NO control has been shown to be applicable to O_2 enriched combustion of RC2 and RC2-SM and RC2-CS (Fig. 7a-b). The technique resulted in substantial reductions (i.e. 77%-80%) under unstaged combustion conditions from 21% to fullest 100% oxygen concentration in the secondary air (Fig. 7a). Moreover, SNCR NO control technique proved to be particularly effective at the low level of staging (22%), Fig. 7a (where O_2 enrichment tends to increase NO emissions for RC2), achieving about 64% to 83% reduction in NO emissions. At higher staging level (31%), Fig. 7b, where NO emissions are lower (<200ppm), reductions of about 50% were observed.

Table 5 summarizes the SNCR NO emission results and corresponding NH_3 utilization efficiencies.

The decreasing NO emission profiles with increasing secondary and over fire air oxygen concentrations were attained for both 22% and 31% levels of staging. However, the impact on NO reduction was more significant in the case of a lower level of staging because of the initial levels of NO emission were higher. Moreover, the following reasons are associated with the SNCR NO control under oxygen enriched conditions;

1- The reduction in the volume of flue gases will enhance the concentration of reacting species (i.e. NO, O_2 and NH_2) resulting in enhanced rates of reduction reactions.

2- The availability of concentrated oxygen in the reduced flue gas volume may help to further sustain the NO reduction reactions, because of the net gain in the OH radical pool via the self sustained reactions referred in literature [31-34].

3- It can also be anticipated that mixing of ammonia, as a reducing agent, with the flue gas species under reduced flows will be more efficient as compared to normal air firing condition.

Few researchers have highlighted the presence of oxygen as being essential for the initiation of SNCR reactions. In the SNCR de NO_x process the reactions for NO reduction by ammonia, in the presence of oxygen and the oxidation of ammonia are competitive. In a recent study, it was shown that due to the presence of increased levels of oxygen the effective temperature window for NO destruction widened and shifted to lower temperature resulting in diminished levels of NO [35]. Similarly, a monotonic increase of NO reduction with oxygen concentration near 900^oC was achieved; whereas, NO reduction appeared to be independent of oxygen at higher temperatures [36]. Additionally, the presence of excess oxygen has been reported to decrease the ammonia slip [37]. It has also been reported that in the absence of O_2, the hydrogen atoms react with ammonia at slower reaction rates for NO removal [31]. Less variation of the temperature measurements in the region where ammonia was injected, has been observed (i.e. $1000\text{-}1080^oC$) for different oxygen enriched conditions. The results obtained here in were found comparable to the work done by other researchers as shown in Table 6.

Fig 8-9 shows the impact of SNCR on NO emissions for 15% co-fired conditions. The results indicate the maximum benefit of NO reduction in case of RC2-SM co-firing for both staging levels. This is again believed to be due to the higher initial NO emissions for 15%

blending ratios of SM with RC2 compared to RC2-CS along with the earlier enlisted reasons. Notably, the SNCR NO control technique resulted in a range of about 40% to 80% reduction in NO emissions for co-fired conditions at a 22% level of staging. At higher staging levels (31%), Fig. 9, reductions of about 40-70% were observed. In general, a decrease in NO emission with increasing secondary and over fire air oxygen concentrations was again attained for both 22% and 31% levels of staging. Hence, NO control proved to be beneficial for both coal firing and coal-biomass co-firing conditions.

5. Conclusions

SNCR Co-combustion tests with and without SNCR resulted in higher NO reduction, especially for the biomasses emitting higher evolved species like CO, which indeed plays a major role in reducing the NO emissions. A temperature range of 1025°C to 950°C is estimated to be the optimum temperature window, within which the homogenous gaseous phase reactions give highest NO reduction with SNCR NO controlling process. The utilization of ammonia injection, in case of 15% blends of biomass with coal, has proven to be better due to higher NO removal and NH_3 utilization efficiencies, in comparison to 50% biomass blending ratio. In order to understand the effectiveness of the SNCR NO control technique, comprehensive tests were also performed under oxygen enriched firing conditions, including oxygen enriched air-staging. The technique proved to be effective at 0.9 SR1 (22% staging level) with results in the range of about 64% to 83% reduction in NO emissions for RC2, and 40% to 80% reductions in the cases of co-combustion. Whereas at 31% staging levels (SR1=0.8), where NO emissions were lower, reductions of about 50% and 40%-70% were observed for RC2 combustion and co-combustion conditions, respectively. This suggested that the availability of concentrated oxygen in the reduced gas volume might have helped to result in the net gain in the OH radical pool in order to further sustain the NO reduction reactions. Moreover, the reduced gas volumes will be beneficial towards enhanced mixing of ammonia since the mixing of ammonia reductant is very crucial at boiler scale. Hence, in the case of oxygen enrichment of the burner air only, where NO emissions are likely to increase many fold, the SNCR NO control technology should be applied while maintaining the advantages of higher thermal efficiency, highly concentrated streams of CO_2 and better carbon burnouts.

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