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NO and SO₂ emissions from combustion of raw and torrefied biomasses and their blends with lignite.

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HIGHLIGHTS

- Torrefaction led to considerable decrease in nitrogen of poultry litter.
- NO emission from torrefied biomass was higher than that from raw biomass
- SO₂ emissions depend on basic mineral content rather than sulfur content in fuel
- Addition of torrefied biomass to lignite considerable decreased SO₂ emission.

ABSTRACT

The influence on NO_x and SO_x emissions of mixing torrefied and untreated olive tree prunings and poultry litter with a Turkish lignite was investigated using a bench scale single particle reactor. In addition, the emissions from the pure fuels were investigated. Time resolved as well as total emission amounts of SO₂ and NO for each fuel were measured at 900, 1000, and 1100 °C and 3 % O₂. The study revealed that the SO₂ emissions from blends are not proportional to the mixing ratio of the fuels and to the emissions properties of the respective fuels. No clear correlation was detected between the NO_x emissions and fuel nitrogen content.

INTRODUCTION

Intensive poultry production has led to stringent environmental legislations for poultry litter (Gerber et al., 2007) and to the requirement of alternative management options (biochemical and thermal processes). Combustion, as a thermal process, is the easiest way to convert the waste to a sterile material with a significant reduction in volume by 80–95% and to recover energy (Pandey et al., 2016). In combustion of poultry litter waste, the major environmental and operating problems are associated with its high moisture, nitrogen content and composition of its inorganic contents (Di Gregorio et al., 2014). One strategy in thermal conversion of poultry litter is co-combustion.

Co-combustion of biomass or waste with coal is a simple and economically feasible way of utilizing biomass and waste for replacement of fossil fuels. The co-combustion of lignocellulosic biomass is increasingly gaining importance as it represents a low cost, sustainable, and renewable energy option that can provide reduction in net CO₂, SO₂ and often NO_x emissions (Sahu et al., 2014). Lower SO₂ emissions can be expected due to lower sulfur content of the biomass and partly due to retention of sulfur in the ash as alkali sulfates. Zhang et al. (2013) observed significant reductions in SO₂ emission as the increase of tobacco stem content in blend containing high-sulfur bituminous coal due to low sulfur content of the biomass. Similar results were obtained for rice husk/coal blend (Huang et al., 2016), saw dust/coal blend (Kazagic and Smajevic, 2007) and agricultural wastes/coal blends (Narayanan and Natarajan, 2007). Despite its animalic origin, poultry litter waste is in many respects comparable with other biomass fuels. A number of co-combustion studies on poultry litter have been performed over the past decade. For instance, the influence of the combustion conditions on emission levels of pollutants such as SO₂ and NO_x during co-combustion of chicken litter and peat in fluidised bed combustion has been investigated by Henihan et al. (2003) and Abelha et al. (2003). Abelha et al. (2003) found that the main problem associated with the combustion of poultry litter was the high moisture content which influenced its feeding to the combustor. Li et al. (2008) investigated the effect of co-combustion of poultry litter and coal on emissions in a fluidized bed combustor. They observed that addition of poultry litter into blend reduced the levels of SO₂. However, the level of NO either increased or decreased depending on the percentage of poultry litter in the blends. Furthermore, thermogravimetric analysis (TGA) has been used in studying the combustion properties and thermal kinetics of poultry litter/coal blend (Yurdakul, 2016)

Although co-combustion is the least complicated and one of the most advantageous ways of utilizing biomass and waste in stationary energy conversion, there are technical challenges associated with co-combustion in existing coal power plants. These challenges include poor grindability and low energy density. In order to improve the fuel properties of biomass, torrefaction, i.e., mild heat treatment of biomass, may be used. Torrefaction of biomass can result in a less hydrophilic solid product with a higher heating value. In addition, the grindability may also improve (Van der Stelt et al., 2011; Gil et al., 2015). Torrefaction has attracted significant interest in recent years. Most of the research has focused on the effect of process conditions and feedstock type on the product properties. A number of kinetic studies have also been undertaken in order to understand the combustion behavior of torrefied

biomass. Nevertheless, only few studies on the emissions from combustion of torrefied biomass have been reported in the literature (Li et al., 2012; Ndibe et al., 2015a,b).

In this study, we investigate NO and SO₂ release from blends of lignite with four different biomasses: olive tree pruning and torrefied olive tree pruning, poultry litter and torrefied poultry litter. The study is carried out using single particle reactor where single fuel pellets with different biomass to lignite ratio are combusted in a controlled gas atmosphere. In addition, the sintering propensity of the ash residues are analyzed using Scanning Electron Microscopy.

MATERIALS and METHODS

2.1 Materials

The fuels used in this study included a Turkish lignite (from the Soma basin, Turkey), olive tree pruning, torrefied olive tree pruning, poultry litter and torrefied poultry litter. The torrefied fuels were produced in a vertical furnace at 300 °C for a residence time of 30 min. Detailed information about the torrefaction process can be found in a previous work (Toptas, et al., 2015). The fuel properties are listed in in Table 1. Prior to combustion experiments, the fuels were dried in an oven and subsequently ground to a particle size less than 2 mm. Pellets were prepared from pure fuels and blends. The pellets were pressed using a manually operated hydraulic press. The diameter of the pellets was 8 mm. Instead of identical heights of the pellets, all pellets had a mass of 0.2 g. The blends of lignite and torrefied biomass and lignite and untreated biomass were prepared by physical mixing. Four different blends of lignite and untreated biomass were prepared. The mass fraction lignite in these samples were 25 wt.%, 50 wt.% and 75 wt.%, referred to as 3/1, 1/1, 1/3. In the pellets containing lignite and torrefied biomass, the mass ratio was 50%.

2.2 Combustion Experiments

The combustion behavior of the fuels and their mixtures were investigated using a single particle reactor, which consists of a quartz tube reactor inserted in an electrically heated ceramic furnace. The main part of the gas enters through an inlet at the bottom of the reactor. A smaller portion of the inlet gas enters at the level where the fuel sample is inserted. This gas is needed to cool the sample before entering the reactor as well as to keep the viewing windows clean. A total flow rate of 220 l/h was used in all experiments. A mixture of 3 vol. % O₂ in N₂ was chosen to avoid very high particle temperatures in comparison to the reactor temperature because of exothermal oxidation reactions. The

combustion experiments were conducted at three different temperatures: 900 °C, 1000 °C and 1100 °C.

The flue gas from the reactor was analyzed with commercial analyzers for the measurement of O₂, CO, CO₂, SO₂ and NO. Based on the concentration profiles of the measured gases and a known total flow, the total amount of NO and SO₂ were calculated for the whole combustion time. A detailed description of the experimental procedures can be found in manuscript published by Giuntoli et al. (2010).

2.3 Characterization of fuels and ashes

Ultimate analysis was carried out using LECO CHNS 932 elemental analyzer according to ASTM D5291-96. The proximate analysis was done according to NREL/TP-510-42622 for ash analysis and ASTM D3175-89a for volatile matter. The higher heating values (HHV) of the biochars were calculated according to the formula proposed by Channiwala and Parikh (2002). The metal analysis of ashes in feedstocks was analyzed by Atomic Absorption Spectrometry (AAS) following acid digestion. In addition, the residues from each combustion were analyzed by a scanning electron microscopy (SEM) equipped with an energy dispersive X-ray spectrometry (EDX).

RESULTS AND DISCUSSIONS

3.1 Fuel analysis

The volatile content of the olive pruning and the poultry litter is significantly higher than that of the lignite of the torrefied fuels. However, when torrefied, the volatile content of olive pruning and the poultry litter becomes similar to that of the lignite. In case of poultry litter, torrefaction decreased the nitrogen content considerably. In case of olive tree pruning, on the other hand, the nitrogen content was slightly increased. In partitioning of nitrogen between the volatiles and char during torrefaction, the nitrogen precursor in biomass is one of the parameter (Darvell et al. 2012). A major nitrogen containing compound in poultry litter is urea. Consequently, poultry litter releases nitrogen compounds at low temperatures due to the low decomposition temperature of urea (Whitely et al., 2006). Previous studies (Kim et al. 2009; Agblevor et al., 2010) related to pyrolysis of poultry litter also showed that most of nitrogen in poultry litter was released during the pyrolysis. Nevertheless, it should be noted that torrefied poultry litter still contain a considerable amount of nitrogen. On the other hand, sulfur behaved differently: most of the sulfur remained in the torrefied fuels.

3.2 Ash sintering behavior

To test the ash sintering tendency, the residues from the combustion of all fuels at different temperatures were analyzed by different temperatures. The holding time varied

between 10 and 35 minutes, partly because of differences in char reactivity. Figure 1 displays the morphology of the residues from the combustion of poultry litter, olive pruning and lignite at different temperatures. Although, residues was powder structure as visually, the figures reveals that for biomass fuels, ash sintering started at 900 °C. The spherical shape and smooth surface imply they have been experienced a molten stage. At 1100 °C, the poultry litter residues presents a quite homogenous structure, probably due to total melting of alkali compounds (Fernández et al., 2012). As expected, the ash from lignite shows different morphology. Especially the content of Si and Al are higher in this case.

SEM-EDX was used to analyze the composition of the ash residue. The result from this analysis, presented as oxides is shown in Figure 2. The analysis revealed significant amounts of K and Ca and minor amount of Si in the residue from combustion of olive pruning and torrefied olive pruning. For poultry litter and torrefied poultry litter, the combustion residue contain significant amount P besides K and Ca. This indicates the presence of alkali phosphate besides alkali oxides, explaining the observed ash sintering.

3.3 Gaseous emissions

To determine the time required for complete combustion for each fuel, a series of experiments with varying combustion time (15, 20, 25 and 30 min) and temperatures (900-1100 °C) were carried out. Based on the carbon oxides (CO+CO₂) levels, 15 min, 20 min, 30 min reaction times for olive pruning, poultry litter, biochars / lignites, respectively, were taken as combustion times in the following experiments .

SO₂ Emission

Figure 3 shows SO₂ release during the first 200 seconds. It can be seen that SO₂ is mainly released during the devolatilization stage. Total emissions of SO₂ varied significantly between the different fuels. One of the benefits of biomass combustion is reduced SO₂ formation through a decrease in fuel bound sulfur (Africa, 2000). It is interesting, that although biomass is seen as viable option to decrease SO₂ emission, the highest SO₂ emissions were measured during combustion of olive pruning, although this fuel has the lowest S content (Figure 4). There was no clear relation between the sulfur content of the fuel and the SO₂ emissions. This indicates that SO₂ emission depends on the amount and the composition of the mineral content of the fuel rather than on the sulfur content. It should be noted that SO₂ emissions from torrefied biomasses are lower than that from that from raw biomasses. This shows that torrefaction led to decrease in the combustible sulfur content of biomasses. Combining the results from the sulfur contents and the char yields, we can say that 50% and 10 % of total sulfur in poultry litter and olive pruning, respectively, were released

during torrefaction. The fact that the combustion residue from torrefied biomasses contained higher sulfur than that from raw biomass (Figure 1) shows that most of sulfur in torrefied biomasses were in un-combustible form.

The experiments showed an impact of temperature on the SO₂ release. The SO₂ release is lower at 900 °C compared to at the higher temperatures. This may be due to a decrease in self-desulfurisation with rising temperature (Heschel et al., 1999). Similarly, Dayton et al. (1999), who investigated the release of sulfur during combustion of switchgrass, observed that SO₂ release is temperature dependent.

A comparison between the measured and anticipated SO₂ emissions from combustion of blends at 1000 °C is shown in Figure 5. The anticipated emission is based on a purely additive behavior of the fuels. The figure reveals that the SO₂ emissions cannot be predicted based on the results obtained using pure fuel. For the blend containing 25 % and 75 % poultry litter, SO₂ emission increased compared with case of the lignite only, whereas blending poultry litter with the lignite at the ratio of 50% led to considerable decrease in SO₂ emission. As seen from Figure 5, in the case of the blend containing 25 % poultry litter, the experimentally obtained total SO₂ emission was much higher than the anticipated one. In contrast to poultry litter-lignite blends, adding olive pruning to lignite at the ratio of 25% decreased the SO₂ emission compared with the lignite only case. In this case, the experimentally obtained total SO₂ emission was much lower than the anticipated one. Increasing the fraction of olive three pruning in blend resulted an increase in the total SO₂ emission. A similar behavior was observed by Wang et al., (2011) in their study of blends of wheat straw and coal. In contrast to the blends containing untreated biomasses, the blending of lignite with torrefied biomass significantly decreased the SO₂ emission compared with the lignite case. In addition, the experimentally observed emissions were lower than those anticipated based on a purely additive behavior. Consequently, in the co-combustion experiments, using torrefied biomass showed a larger reduction in SO₂ emission than using untreated biomass. To summarize, in spite of the higher sulfur content of the torrefied fuel, the blend of the torrefied fuel and the lignite released less SO₂ than the blend of the untreated fuel and the lignite. This may be related to differences in the ash compositions and the higher ash content of the torrefied biomass compared to the untreated biomass.

NO Release

The formation of NO is a complex process depending on fuel type and combustion conditions, such as temperature, air ratio, heating rate, particle size, etc. In solid fuel combustion systems, at temperatures below 1500 °C, the most important NO formation

mechanism is that of oxidizing fuel bound nitrogen. In this study, it is expected that measured NO is from the fuel-N, because the combustion temperatures were too low for formation from molecular nitrogen in the inlet gas (Glarborg et al., 2003; Roy et al., 2013).

Figure 6 shows the total NO emission obtained in the single particle experiments. The total emissions of NO differed significantly from fuel to fuel. The NO emission is not purely depending on nitrogen content in fuel. For example, the nitrogen content of olive pruning was slightly lower than that of the lignite, but the total NO an emission from olive pruning was higher than from the lignite. One of the probable reason is the NO destruction via DeNOx process ($4\text{NO} + 4\text{NH}_3 + \text{O}_2 \rightarrow 4\text{N}_2 + 6\text{H}_2\text{O}$). It has been reported that the pyrolysis of amine type nitrogen containing species in biomass would provide a possible source for NH_3 (Tchapda and Pisupati, 2014). Winter et al., (1999) found that the fuel nitrogen conversion decreased with increasing nitrogen content and contributed this to the DeNOx process. Moreover, also Giuntoli et al. (2010) found that a high fuel-N content could enhance De-NOx reaction. Similar result was also reported by Billen et al. (2015). . One of the most important finding in this study is that although the fuel bound nitrogen content in torrefied biomass (especially torrefied poultry litter) was lower than in raw biomass, the NO emission from the combustion tests of torrefied biomass was higher. A possible reason is the fact that the fuel-N functionality in fuels were different. The amine type functional groups in biomass (especially poultry litter) seems to be destructed during torrefaction.

The temperature influence on the NO emissions also varied with fuel. In the case of lignite, NO emission decreased with increasing combustion temperature. This result may be reasonable because of the increasing of the NO destruction reactions with NH_3 . However, in case of biomass and torrefied biomasses, there is no linear correlation with temperature and the minimum NO emission was observed during combustion at 1000 °C.

A comparison between the measured and anticipated NO emissions from combustion of blends at 1000 °C is shown in Figure 7. In case of blends, the NO emissions increased as the poultry litter ratio increased. This is reasonable because the NO emissions from poultry litter alone is higher than that of lignite alone. Consequently, NO emissions during co-combustion of lignite with poultry litter might be expected to be higher than the lignite only case. However, the NO emissions decreased slightly as the olive pruning ratio increased. It is noticeable that experimental NO emission values from both raw and torrefied biomass containing blends were lower than the anticipated ones. A probably reasons is the reactions between volatiles from biomass and from lignite, leading to the suppression of NO formation.

CONCLUSION

Gas emissions and combustion behavior of individual pellets of blends of lignite with olive tree pruning, torrefied olive tree pruning, poultry litter and torrefied poultry litter were investigated. The results were compared to those obtained using non-blended fuels. In contrast to previous studies, no clear relation between the sulfur and nitrogen contents of the fuel and the NO/SO₂ emissions was observed. Although the torrefied biomasses had higher sulfur content, SO₂ emissions from the torrefied biomasses were lower than those of the untreated biomasses. However, fuel bound nitrogen content in the torrefied poultry litter was lower than in the untreated poultry litter, NO emission from combustion of torrefied poultry litter was higher. Co-combustion of lignite and torrefied biomasses had a beneficial impact on SO₂ release. In contrast, no benefit of co-combustion on the NO release could be observed for blends of torrefied biomass with lignite. The results reported that torrefaction provide a technical option for high substitutions of biomass in the co-firing system..

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