

Effects of Co-firing Bosnian Coals with Wooden Biomass to the Emissions of NO_x and SO₂

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Abstract

The co-firing using sawdust in combination with Bosnian brown coal and lignite blend, with coal/biomass mixtures at 93:7%wt and 80:20%wt, were tested. The tests performed by a 20 kW entrained flow reactor varying the temperature between 880 °C and 1550 °C and the excess air ratio between 0.95 and 1.4. It was found that content of nitrogen in the fuel was not decisive factor effecting on NO_x emission, however, it was not possible to identify clearly the influence of the biomass content in the blend on NO_x emissions. Reduction of SO₂ emission was detected for all co-firing regimes, with identification of a synergy effect for a brown coal-wooden biomass co-firing.

Introduction

The main reasons for using biomass as co-fuel is its dual role in greenhouse gas mitigation, as a substitute for fossil fuels (bio-energy) and as a carbon sink [1]. This is an increasingly important factor, since the countries of the extended European Union (EU) began trading in emissions in 2005. Fuels derived from biomass also contain less sulfur or trace elements. As set out in the European Commission's White Paper on Energy for the Future: Renewable sources of energy [2], in the short-to-medium term, the co-combustion of biomass and bio-waste in coal-fired power plants is one of the most straightforward biomass applications. Current research on co-combustion is focused on controlling combustion behavior, emissions, corrosion, agglomeration, and fouling-related problems. Biomass for combustion in industrial-scale combustors must meet a number of criteria, including availability throughout the year to ensure security of supply, high density to minimize transportation costs, sufficiently high heating value, and acceptable price [1]. Wood residue meets these requirements, [3].

Paper presents research into effects of cofiring Bosnian coals, brown coal and lignite, with wooden biomass to the emissions of NO_x and SO₂. For a basic coal, emissions in co-firing regimes could be investigated as function of process temperature, excess air ratio and air distribution, and content of wooden biomass in the co-firing mixture.

Specific Objectives

Reactor configuration

For purpose of the research, 20 kW lab-scale furnace electrically heated entrained PF flow reactor is used for the tests, Figure 1. The main feature of the experimental facility is ability to change the process temperature at desire in the range from the ambient to 1560 °C. It is provided by SiC-type electric heaters controlled by a

central PLC and thiristor units connected at PLC digital outputs for each heating zones of the reactor, [3].

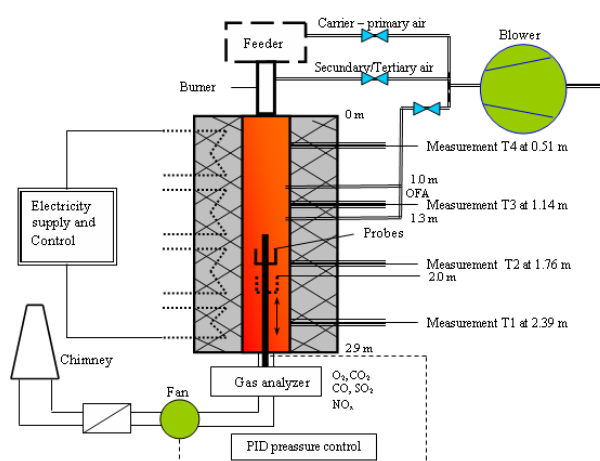


Figure 1. Schematic layout of the reactor used for the co-firing tests

Various types of coal and biomass can be used, processed to appropriate particle size by a laboratory hammer mill. The pulverized fuel particles were fed into the reactor by means of a volumetric-type feeder, equipped with a speed controller, allowing mass flow in the range of 0.25 – 5.25 kg/h. The air for combustion from the blower was split into carrier or primary air, secondary air, tertiary air, and over fire air (OFA). The first three air portions were introduced into the reactor over a burner placed at the top of the reactor, so that the air-fuel particle mixture flowed downward. The final air portion or OFA was used for burning out, i.e. to investigate the air stage combustion, simulating the OFA system used in large boilers. It was consequently introduced directly into the reaction tube (see Figure 1). The excess air ratio was adjusted by controlling the air flow in each air line at a constant fuel flow.

In this way, NO_x and SO₂ emission are measured under different conditions in the reactor, varying the

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process temperature, excess air ratio, and air distribution.

Fuel test matrix

The co-firing tests used spruce sawdust in combination with Kakanj brown coal and a lignite blend consisted of Dubrave lignite and Sikulje lignite. Coal/biomass mixtures at 93:7%wt and 80:20%wt were tested. The resultant co-firing fuel test matrix is given in Table 1. Table 2 presents proximate and ultimate analyses of the fuels tested. Comparison of the results in

Table 2 for the coal and biomass suggest that the main difference between them relates to the ash content, as the coal types tested have very high ash content compared to the sawdust. This has a significant influence on the combustible content and calorific value of the fuels. On the other hand, the biomass sample has a very high volatile content, which may be expected to accelerate the combustion process, while the coal types have a much higher fixed carbon concentration. Finally, the wood sample has significantly lower sulfur content, [3,4].

Table 1. Fuel test matrix

No.	Fuel	Fuel type	Symbol	% Weight	%Thermal input
1	Dubrave-Šikulje	Lignite	L	100	100
2	Kakanj	Brown coal	K	100	100
3	Blend of Kakanj with Spruce	Co-fuel	K93S7	93:7	89:11
4	Blend of Kakanj with Spruce	Co-fuel	K80S20	80:20	72:28
5	Blend of Dubrave/Sikulje with Spruce	Co-fuel	L93S7	93:7	88:12
6	Blend of Dubrave/Sikulje with pruce	Co-fuel	L80S20	80:20	70:30

Table 2. Proximate and ultimate analysis of the fuels tested

Fuel	L	K	S	L93S7	L80S20	K93S7	K80S20
<i>Proximate analysis, %, as-received</i>							
Moisture	34.22	11.30	11.20	32.60	29.61	11.29	11.28
Ash	23.62	41.43	0.26	21.99	18.95	38.55	33.20
Volatiles	24.32	25.88	75.48	27.90	34.55	29.35	35.80
Fixed C	17.42	21.39	13.06	17.11	16.55	20.81	19.72
Combustible	42.16	47.27	88.54	45.41	51.44	50.16	55.52
<i>Ultimate analysis, %, as-received</i>							
Carbon	29.58	35.11	38.91	30.23	31.44	35.38	35.87
Hydrogen	2.49	2.78	7.36	2.83	3.47	3.10	3.70
Sulfur	0.73	2.28	0.33	0.71	0.65	2.14	1.89
Nitrogen	0.48	0.96	0.33	0.47	0.45	0.92	0.83
Oxygen	9.29	7.03	41.61	11.55	15.75	9.45	13.95
<i>Heating value, kJ/kg, as-received</i>							
Gross	10889	13490	17386	11344	12188	13763	14269
Net	8588	12657	15612	10009	10793	13022	13700

Test runs

In each co-firing run, sawdust was mixed with coal and the fuel blend was pre-dried at approximately $w = 0\%$ and then supplied into the feeder tank. Table 3 presents the particle size distribution of the fuels tested. Fuel thermal load was kept at approximately 5 kW_{th} in all runs. Process temperature varied from 880 to 1550 °C and excess air ration from 0.95 to 1.4. Depending on the fuel and excess air used, the total airflow rate was between 4.29 and $6.60 \text{ m}_n^3/\text{h}$. The primary (carrier) air flow rate was set at $1.50 \text{ m}_n^3/\text{h}$ for all runs, with the rest of the air divided into secondary and tertiary portions, at a ratio of 2.6:1.

NO, NO₂, SO₂ and CO emissions were measured in ppm by a TESTO 350 instrument with an integrated TESTO 339 dry unit, and emissions were then converted at $[\text{mg}/\text{m}_n^3, \text{dry}, 6\% \text{ O}_2]$. Measurements were repeated

several times during each test (90 minutes lasting) to eliminate the influence of accidental phenomena. Measurement error was estimated at 18 ppm (2.8%) for the NO emissions and at 59 ppm (2.6%) for the SO₂ emissions. Flue gas temperature was measured at the point in the partially insulated outlet tube where the gas sample was taken for emission measurement. Depending on the test run, gas temperature was between 50 and 140 °C. The processes in the flue gas line were frozen; there was no either post combustion or air suction from the reactor to the TESTO instrument.

There was sufficient combustion efficiency under all co-firing regimes, with burning out in ash deposits at 96.5 to 99.5% for brown coal–sawdust co-firing, and 99% for lignite-sawdust co-firing.

Table 3. Particle size distribution for the fuels tested – rest on the sieve in %wt

Fuel	Total %	200 μm	100 μm	90 μm	80 μm	71 μm	63 μm	45 μm	passed
K	100	18.2	28.0	-	17.4	-	-	-	36.4
S	100	67.5	-	-	26.7	2.0	1.0	1.2	1.6
K93S7	100	21.1	-	-	34.2	4.7	22.3	3.0	14.7
K80S20	100	29.6	-	-	31.3	5.7	16.9	4.0	12.5
L93S7	100	49.5	-	-	25.5	3.7	12.5	2.0	6.8
L80S20	100	49.0	-	-	27.9	3.1	8.6	2.9	8.5

Results and Discussion

NO_x emission

Figure 2 presents the NO_x emissions for the brown coal-biomass blends tested.

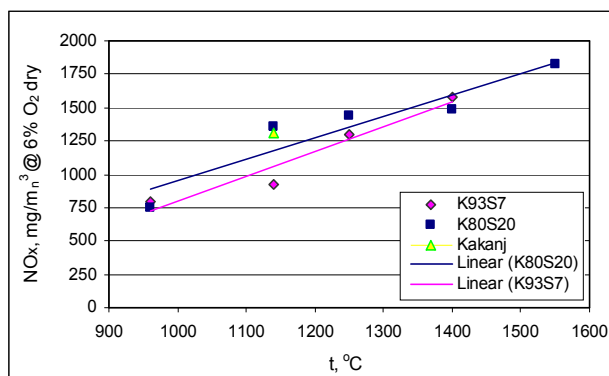


Figure 2. NO_x emissions for the Kakanj coal-spruce sawdust co-firing at $\lambda = 1.2$

The results are in line with the Zelkowski-model of flame temperature impact on NO_x formation, as reported in [5]. Very high NO_x emissions are also noted at 1550 °C (1800 mg/m^3 at 6% O_2 dry), due to *thermal NO* formed at temperatures above 1400 °C, according to the Zeldovich-mechanism. Both of the coal-biomass blends tested showed a 50% reduction in NO_x , as the process temperature fell from 1400 °C to 960 °C (reducing from 1600 to 800 mg/m^3 normalized to 6% O_2 dry, at excess air ratio $\lambda = 1.2$). There was, however, an unexpected difference in NO_x emissions for the coal-biomass blends at 1140 °C. The deviation was ascribed to the difference in particle size of the tested blends, as well as to the difference in volatile content of the fuels. No clear relation could be detected between the NO_x emissions for the different brown coal-biomass blends, despite differences in nitrogen content (see Table 2).

Measurements, moreover, confirmed that nitrogen content was not a decisive factor for NO_x emissions for the lignite-sawdust co-firing at the same temperature, either. Figure 3 shows comparison of NO_x emissions for the lignite blend of Dubrave and Sikulje (L), the mixture of L and sawdust at 93:7 %wt (L93S7) and the mixture of L and sawdust at 80:20 %wt (L80S20), at excess air ratio $\lambda = 1.2$. Principally, the highest emission was measured during test of L93S7 coal/biomass blend (with N = 0.47% and Volatiles of 27.9%), which is slightly higher compared to both lignite alone L (with N

= 0.48% and Volatiles of 24.32%) and L80S20 coal/biomass mixture (with N = 0.45% and Volatiles of 34.55%).

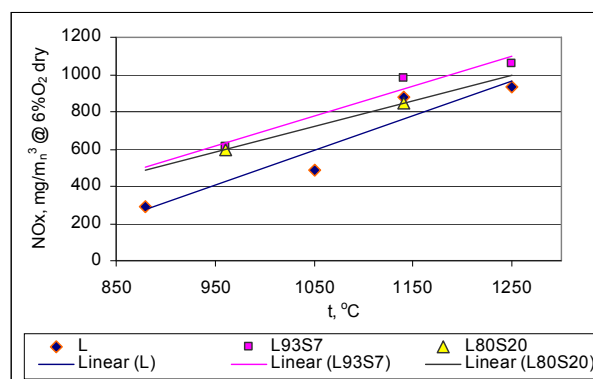


Figure 3. NO_x emissions for the Lignite coal-spruce sawdust co-firing at $\lambda = 1.2$

Explanation of such trend of NO_x emission during the lignite-sawdust co-firing could be supported by the following facts and theses: With adding more biomass to the basic lignite, nitrogen content in the coal/biomass mixture decreases and Volatiles content increases. Both contribute to the NO_x emission decreasing, as reported in [5]. For this reason, NO_x emission for the L80S20 blend is lower than for other two fuels. From the other side, however, more spruce sawdust in the coal/biomass mixture possibly contributed to the local separation of the slighter wooden biomass particles in the combustion zone of the reactor, which were held longer, against coal particles, in the high temperature zone. This effect increased NO_x emission during coal/biomass blends, resulting in higher NO_x emission for L93S7 mixture against the lignite alone. These results are in line with those reported in [6, 7].

By comparison of the NO_x emission values between the brown coal-spruce sawdust co-firing, Figure 2, and the lignite-spruce sawdust co-firing, Figure 3, it can be noticed that the brown coal-spruce sawdust co-firing produced in between 25 and 30 % higher NO_x emission than the lignite-spruce sawdust co-firing, under the same conditions. This finding was ascribed to the almost double higher content of nitrogen in the Kakanj-spruce mixtures against the Lignite-spruce mixtures, Table 2.

As regards the impact of the excess air ratio on the emissions, it was found to exert a strong influence on

the NO_x emissions both for the basic coals tested and for the co-firing fuels tested, see Figure 4.

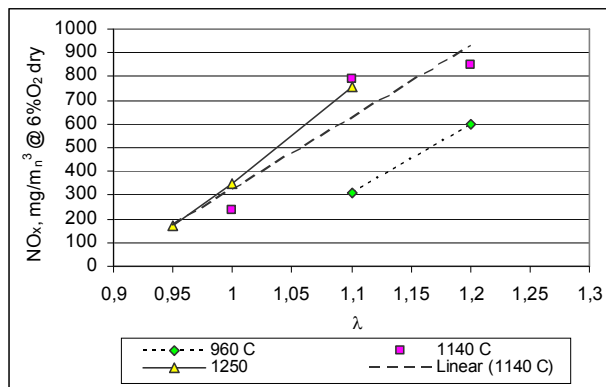


Figure 4. NO_x emissions as a function of excess air ratio and process temperature for the L80S20 mixture

In the co-firing regime of L80S20 mixture at 960 °C and with excess air ratio of $\lambda = 1.1$, NO_x emission is approximately 300 mg/m^3 at 6% O_2 dry. Also, acceptable NO_x emission values are for the L80S20 co-firing regime for stoichiometric $\lambda = 1.0$; 240 mg/m^3 at 6% O_2 dry at 1140 °C and 350 mg/m^3 at 6% O_2 dry at 1250 °C was measured. For excess air ratio under stoichiometric zone ($\lambda = 0.95$), NO_x emission decreased below 200 mg/m^3 at 6% O_2 dry, but accompanied with very high CO emission of 1850 mg/m^3 at 6% O_2 dry in this regime. It should be emphasized here that there was no air staging applied that explained very high CO emission measured in this regime.

SO₂ emission and sulfur capture rate

Considerably less SO_2 was measured for the brown coal-biomass blends than for the coal alone. Thus, at 1140 °C, there was 15% less SO_2 for the K93S7 blend and 28% less for the K80S20 blend than for Kakanj coal alone, see Figure 5, [3].

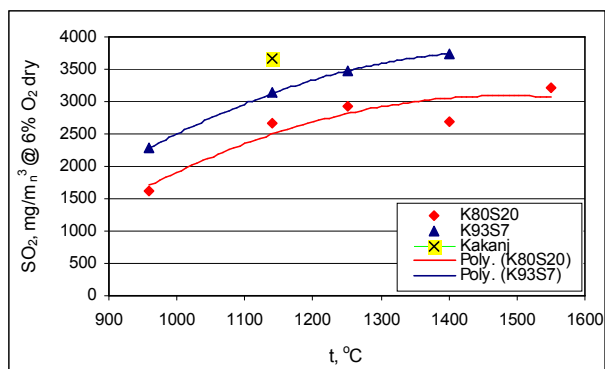


Figure 5. SO_2 emissions for the Kakanj coal - spruce sawdust co-firing, $\lambda = 1.2$

The main reason is less sulfur in the coal/wooden biomass mixture against the coal alone; see Table 2, and also, increasing Ca/S molar ratio with adding wooden biomass to the coal/biomass mixture.

With regard to the lignite-wooden biomass co-firing, the lowest SO_2 emission was measured for the L80S20 mixture ($S = 0.65\%$, $\text{Ca}/S_{\text{mol}} = 0.863$), Figure 6. Approximately 20% higher SO_2 emission was measured for the L93S7 mixture ($S = 0.71\%$, $\text{Ca}/S_{\text{mol}} = 0.897$). However, it is interesting that for the basic fuel – lignite L ($S = 0.73\%$, $\text{Ca}/S_{\text{mol}} = 0.899$), emission of SO_2 at lower process temperatures (880 and 960 °C) is lower against lignite-spruce sawdust mixtures. This can be ascribed to the more convenient Ca/S molar ratio for the lignite alone, which makes sulfur bonding to the calcium from the ash better at lower temperatures.

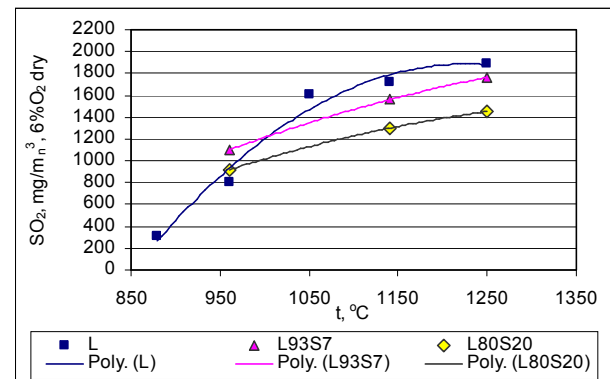


Figure 6. SO_2 emissions for the Lignite - spruce sawdust co-firing, $\lambda = 1.1$

By comparison the results of SO_2 emissions between brown coal/biomass co-firing, Figure 5, and lignite/biomass co-firing, Figure 6, it can be noticed approximately 50% higher SO_2 emission for the brown coal/biomass mixtures in all co-firing, although sulfur content is three times greater than in the lignite/biomass mixtures, Table 2. The reason is more convenient conditions for the sulfur bonding in the case of the brown coal/wooden biomass mixtures. This thesis is also supported by the results of sulfur capture rate (S_b) for the co-firing fuel test matrix, Figure 7 and Figure 8, showing higher values for the brown coal/wooden biomass mixtures. Sulfur capture rate is set here as rate of bonding of SO_2 generated immediately in the furnace with the basic oxides from the ash, predominantly CaO .

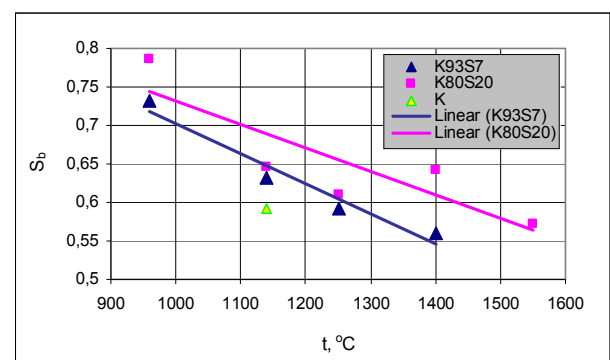


Figure 7. Sulfur capture rate as a function of process temperature for the Kakanj-spruce sawdust co-firing, $\lambda = 1.2$

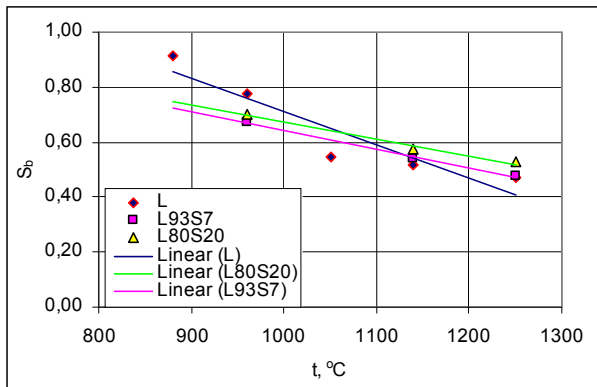


Figure 8. Sulfur capture rate as a function of process temperature for the Lignite-spruce sawdust co-firing, $\lambda = 1.1$

It can be noticed also from the figures that sulfur capture rate decreased for all tested fuels, as the temperature raised. The reason is the well-known phenomenon that sulfur bonds better to CaO particles at lower temperatures [3, 4].

Finally, an increase in the sulfur capture rate was noted for the Kakanj coal-spruce sawdust co-firing for all temperature regimes, as the Ca/S molar ratio rose – i.e. as biomass spruce sawdust was added to the Kakanj coal, Figure 9. This consequently represents a synergy effect relating to SO₂ emission for the co-firing of brown coal with wood biomass, [3].

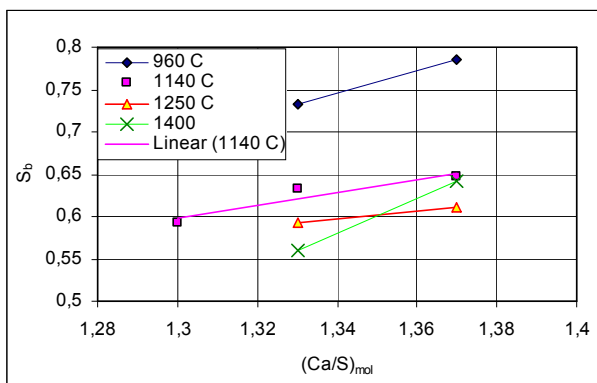


Figure 9. Sulfur capture rate as a function of $(Ca/S)_{mol}$ and process temperature for the Kakanj-spruce sawdust co-firing, $\lambda = 1.2$, $(Ca/S)_{mol K} = 1.30$, $(Ca/S)_{mol K93S7} = 1.33$, $(Ca/S)_{mol K80S20} = 1.37$

This additional SO₂ reduction effect could not be identified for the lignite-spruce sawdust co-firing. Actually, Figure 10 shows that as biomass spruce sawdust was added to the Kakanj coal, accompanied in this case with decreasing Ca/S molar ratio, effect of better bonding of sulfur was noticed only for higher temperatures; 1140 and 1250 °C, while at temperature 960 °C this effect failed. This interesting finding at higher temperatures was ascribed to the lower content of silica oxide in ash of the spruce sawdust (SiO₂ = 21.3%) compared to the lignite (SiO₂ = 49.88%), mitigating

inhibition acting of SiO₂ to the sulfur capture at higher temperatures for lignite/spruce co-firing.

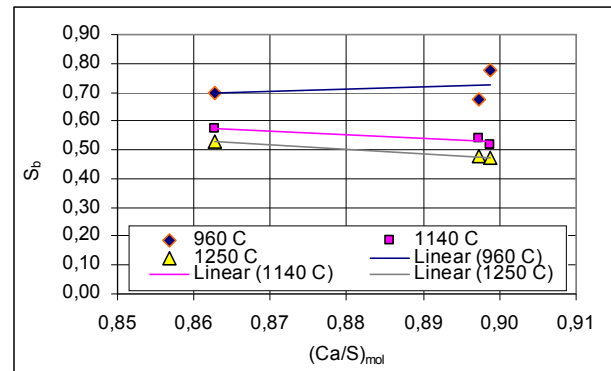


Figure 10. Sulfur capture rate as a function of $(Ca/S)_{mol}$ and process temperature for the Lignite-spruce sawdust co-firing, $\lambda = 1.2$, $(Ca/S)_{mol D\delta S} = 0.899$, $(Ca/S)_{mol D\delta S93S7} = 0.897$, $(Ca/S)_{mol D\delta S80S20} = 0.863$

Conclusions

The paper reports on research into emissions of NO_x and SO₂ during the co-firing of Bosnian coals with wooden biomass. As well as reduced CO₂ emissions, the results provide grounds for optimism regarding the reduction of SO₂ emission. The measurement of SO₂ emissions during the co-firing tests showed considerable decrease of SO₂ emission when the wooden biomass is fired with the coals. In addition to the reduction of SO₂ due to the lower sulfur content in the coal/biomass blends tested, the brown coal-sawdust co-firing generated a further reduction due to the higher sulfur capture rate than for coal alone in all temperature regimes tested.

From the other side, much lower NO_x emissions were measured in all co-firing regimes at the lower process temperatures and the lower excess air ratio used. It was found that content of nitrogen in the fuel was not decisive factor effecting on NO_x emission. Thus, volatiles content and characteristics of the particles and their mixing in the fuel blend, effecting to residence time in combustion zone, were identified as important factors which influenced to the NO_x emission. It was not, however, possible to identify clearly the influence of the biomass content in the co-firing blend on NO_x emissions during the tests performed.

Acknowledgements

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