Emissions Of Flue Gases In Staged Co-Firing Of Coal With Waste Woody Biomass Using Natural Gas -Reburning Technology

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ABSTRACT: In this paper results of research of co-firing coals from Middle Bosnian basin with waste woody biomass are presented. Tested fuels were subjected to pulverized combustion under various temperatures and various technical and technological conditions: different mass ratio of fuel components in the mixture, different coefficient of excess air for combustion, staged air supply into the reaction zone and application of natural gas as an additional fuel (reburning technology). The results are related to the emission of flue gas components (CO₂, CO, NO_x and SO₂) with respect to the technological conditions of combustion. In addition to the valorization of the influence of the primary fuel composition and process temperature on the emission values of the flue gas components, it has been shown that the application of primary measures in the combustion chamber results in lower or higher positive effects in terms of reducing the emissions of individual components – e.g. the NO_x emission is reduced from 837 mg/m_n³ in conventional combustion to 710 mg/m_n³ using air staging. It is also shown that when using natural gas as additional fuel there is an additional reduction in NOx emissions, and this is proportional to the amount of natural gas - e.g. for process temperature of 1350 °C and 10% of natural gas energy share co-firing with coal, a NO_x emission reduction of more than 250 mg/m_n³ was recorded, compared to emissions without additional fuel.

KEYWORDS: biomass, coal, combustion, natural gas, NO_x emission, OFA air

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I. INTRODUCTION

There are significant coal reserves in Bosnia and Herzegovina. The conversion of primary energy from this fossil fuel in thermal power plants produces about ³/₄ of the total electricity production. The balance and exploitation reserves of coal in Bosnia and Herzegovina are about $4.5 \cdot 10^9$ t, of which about 40% refers to brown coal and about 60% to lignite, [1]. However, the quality of Bosnian coal mines varies significantly from one mining basin to another, and even from one mine to another within the same mining basin. The basic characteristics of these coals are low heating value, high mineral mass and moisture content and poor reactivity, [2]. Consequently, various technical and technological problems arise in the combustion of these coals, including instability and discontinuation of the combustion process. The instability of the combustion process, as well as the level of emissions of polluting flue gas components into the environment - especially NO_x emissions - is directly related to the way and place of introduction of combustion air. Coals from Middle Bosnian mining basin (e.g. Kakanj, Breza and Zenica mines) are predominantly delivered to Thermal Power Plant Kakanj in which it is fired in boilers with pulverized combustion and with liquid slag discharge. It is well known that this combustion technology is characterized by high temperature in the furnace, especially in the melting chamber (e.g. 1300÷1550 °C), which results in very high emissions of polluting flue gas components: the CO₂ emission is at a level of 1140 kg/MWh, emission of NO_x is up to 900 mg/m_n³, while SO₂ emission is about 8000 mg/ m_n^3 due to the content of sulfur in coal mixtures of about 2.5%, [3].

Bosnia and Herzegovina also has significant energy potential from waste biomass derived from agricultural production and forestry, primary and secondary wood processing. It is estimated that the total annual technical energy potential of biomass remains in BiH is more than 33 PJ, which is equivalent to more than 3 million tons of BIH lignite, [4].

In principle, a significant reduction in CO_2 emissions from coal-fired power plants can be achieved by intro-ducing waste wood biomass as a renewable energy source into regular operation, while the reduction of NO_x emissions relative to conventional combustion systems can be sufficiently achieved by the use of burners of the newer generation (Low-NO_x Burner, LNB), staged air supply (OFA), staged primary fuel intake into the furnace and using additional fuel (e.g. natural gas, reburning technology). LNB burners work on the principle of a staged combustion air supply at the single burner level - these burners result in a significant reduction in NO_x emissions and as such today they are the essential equipment of newer boilers, which usually work with supercritical or ultra-supercritical steam parameters and high flexibility is required, [5]. In the paper [6], the results i.e. effects of the staged supply of combustion air on the NO_x emission (dried lignite was used as fuel), were presented. It has been shown that in this way there is a significant reduction in NO_x emission - the results are related to the investigation of the influence of the coefficient of excess air, the distribution of combustion air, and the distance of the position of introduction of the OFA air into the reaction zone relative to the burner. In the paper [7] it was stated that, with Low-NO_x burners, the NO_x emission can be reduced by a fifth, but that due to the high co-mbustion temperature this emission is still high (1036 mg/m_n³ in dry flue gases with 6% O₂), and for the further reduction of NO_x emissions, a zonal or staged combustion air supply (OFA) is recommended. Similar positive effects of the staged supply of combustion air on the reduction of NO_x emissions are presented in works [8] and [9] for coal combustion; as well as for co-firing coal with biomass, [10-14].

By subjecting coal blends, as well as mixtures of coal and waste woody biomass, to combustion at different te-mperatures with the application of staged combustion air supply as primary measure, and with the introduction of a portion of the OFA air at different distances from the primary burner and with the natural gas as third fuel, it is possible to determine the appropriate response of the combustion process through the measurement of process parameters and their analysis, i.e. it is possible to come to relevant conclusions about the impact of the location of the introduction of the OFA air and the application of natural gas to the process itself, and co-nsequently the effect on the emission of flue gas components into the environment (CO_2 , CO, NO_x , and SO_2). Based on these conclusions it is possible to quantify and sublimate the characteristics of co-firing coal with woody biomass and natural gas, including the advantages of conversion of primary energy from fuels under co-nditions of solid fuel pulverized combustion technology with a staged combustion air supply, [15].

II. FUEL TEST MATRIX, LABORATORY PLANT AND TEST REGIMES

Fuel Test Matrix: For the purpose of laboratory investigations of the influence of the staged supply of the combustion air into the furnace as well as the application of natural gas as additional fuel while co-firing coal with waste woody biomass on the emission of flue gas components, a matrix of basic test fuels was established, [15] - Table 1.:

Table 1. Basic properties of test fuel, [15]					
Fuel/label	U100	B100	U95B5	U93B7	U90B10
Proximate analysis	s, %				
Moisture	13.90	21.63	19.06	18.09	14.67
Ash	37.88	0.52	34.33	33.05	34.14
Volatiles	28.97	64.14	29.32	31.16	32.49
C _{fix}	19.25	13.72	17.28	18.59	18.70
Combustible	48.22	77.86	46.60	48.86	51.18
Ultimate analysis,	%				
Carbon	32.62	38.92	32.12	33.36	33.25
Hydrogen	2.60	4.95	2.20	2.52	2.84
Sulfur	2.06	0.14	1.57	1.59	1.87
Nitrogen	0.72	0.17	0.81	0.75	0.67
Oxygen	10.22	33.67	9.91	10.63	12.57
Heating values, kJ	/kg				
HHV	13351	15564	12651	13446	13572
LHV	12496	14081	11759	12510	12655

- The usual coal blend that has been fired in TPP Kakanj for the last few years fuel label: U100. This coal blend was produced by mixing coals delivered to the coal depot of TPP Kakanj from several mines (Kakanj, Breza, Zenica, Gracanica, Livno, Nova Bila, Banovići, etc.) in approximately the same percentage as they are delivered from the mentioned mines. For laboratory testing purposes a sample of this coal mixture (coal powder) is excluded directly behind the mills during real operation of the Unit 5 in TPP Kakanj.
- Wood biomass sawdust fuel label: B100. Sawdust is a mixture of beech and spruce formed during primary wood processing, in an approximate 50:50 weight ratio. The sawdust sample was excluded after delivery to the TE Kakanj depot.
- Mixture made of U100 coal blend and waste wood biomass B100 fuel label: U95B5 and U93B7. These mixtures consist of 5 and 7% wood biomass respectively and are excluded behind the mills.
- Mixture made of coal U100 and waste wood biomass B100 fuel label: U90B10. This coal mixture consists of 10% wood biomass and is formed after drying and grinding the components in a laboratory mill.

Laboratory plant: As part of the *Laboratory for Coal and Biomass Combustion* established at the University of Sarajevo - Faculty of Mechanical Engineering, Department of Energy, an experimental plant *Entrained flow tube reactor* was designed and installed, Fig. 1.



Figure 1. Principal scheme of the plant with indicated staged air supply and the application of natural gas (reburning technology)

The laboratory plant was designed in such a way as to enable the testing of a wider range of combustion chara-cteristics of different solid fuels under different ambient and technical-technological conditions. On the previo-usly given principle diagram of this plant, supporting systems for classic or conventional and graded or zonal combustion air supply to the reaction tube are indicated, including the possibility of using natural gas or biogas as an additional fuel when researching combustion characteristics under reburning technology conditions. In the lower part of the plant, solid samples of slag and ash are removed, and the composition of flue gases produced during combustion is analyzed - in this regard, it is recommended to additionally see [10-11] and especially [15]. The laboratory plant can work in a very wide interval of combustion temperature (practically from ambient temperature to max 1560 °C) and under the conditions of supply to the reaction tube of different amounts and distribution of basic solid fuel and air for combustion, including the possibility of researching reburning combustion technologies using the third/ additional fuel. Basically, the research provides output data related to the efficiency of conversion of primary energy from fuel in the established combustion process at the selected temperature, the intensity of deposition and the characteristics of ash deposits along the reaction zone, as well as the characteristics of slag and ash at the exit from the reactor, and the emission of corresponding components in the resulting flue gases into the environment - in this particular case, these are the components: CO₂, CO, O₂, NO, NO₂, NO_x=NO+NO₂ i SO₂, [15].

Test Regimes: The mentioned solid fuels (mixtures of coal and biomass) were subjected to a laboratory investigation of combustion under the conditions of staged supply of combustion air into the combustion chamber and with natural gas as additional fuel. Test regimes are defined and performed in the range of process temperatures corresponding to the temperatures from the real boiler operation in TPP Kakanj: $1350\div1450$ °C. A set of these research was aimed at gaining insight into the characteristics of co-firing coal with biomass, with a particular emphasis on emissions of flue gas components, primarily NO_x. Test regimes of the co-firing with natural gas as an additional fuel are carried out with 5 and 10% of the natural gas energy share. These tests of combustion with natural gas have been assigned the following markings: UPn and UB(95)Pn, where "n" represents the energy share of gas in total energy input supplied into the combustion chamber.

III. RESULTS OF THE RESEARCH: Emission of flue gas components

Co-firing of coal with biomass: For the same combustion temperature, NO_x emission in co-firing was found to be at the same emission level as for combustion of coal without biomass, and that there is practically no change in that emissions with the change in the fraction of wood biomass in the mixture. The average NO_x emission difference depending on the mode of supply of combustion air is 250 mg/m_n³: in the case of conventional air supply, the average NO_x emission is 942 mg/m_n³, while in the case of a stage combustion air supply, the emission is 692 mg/m_n³, Figure 2 - left. On the other hand, the sulfur content decreases proportionally with the increase of fraction of wood biomass in the coal mixture. Despite the fact that the results regarding SO₂ emission are dispersed, it can still be concluded that the SO₂ emission reduces proportionally with the increase in biomass content in the mixture. Additionally, based on the results of the measurements, it can be concluded that the SO₂ emission air supply - that emission is high and at a process temperature of 1350 °C it is approximately 5300 mg/m_n³, Figure 2 - left.

Total and net CO_2 emission due to the combustion of wood biomass as a renewable fuel in a coal mixture is shown in Figure 2 - right. Increasing the fraction of wood biomass in the mixture linearly reduces the net CO_2 emission, which at 10% of biomass in the mixture is 0.233 kg/m_n³. Somewhat more pronounced CO emissions from co-firing of coal with biomass, especially at 10% of the biomass content, compared to the combustion of coal, can be related to the granulation of wood biomass in the mixture that is not mechanically treated (not grinded in the laboratory mill) - a certain fraction of biomass particles was up to 4 mm, Figure 2 - right.

With the increase of combustion temperature, NO_x and SO_2 emissions are also increasing. For example, at temperature of 1450 °C the average NO_x emission is 800 mg/m_n³ and the SO_2 emission is about 5200 mg/m_n³. In these test regimes, the previously stated conclusions were confirmed that the NO_x emissions practically do not change with the increase in the fraction of wood biomass in the mixture, and that the SO_2 emission decreases proportionally by this increase of biomass fraction in the mixture.



Figure 2. Emission of NO_x and SO_2 (left) and CO_2 and CO (right) in the co-firing of coal with wood biomass at process temperature of 1350 °C

The effect of natural gas as reburning fuel on NO_x emission when combusting the basic test solid fuels at process temperature of 1350 °C is shown in Figure 3 - left. In addition, for given solid fuels, the NO_x emission values for the cases of 0%, 5% and 10% natural gas energy share in combustion are shown for comparison. In relation to emissions without the use of gas as additional fuel, a reduction in this emissions by more than 250 mg/m_n³ was recorded for the case of 10% of the gas energy share. For example, the NO_x emission for the U100 coal mixture was reduced to 489 mg/m_n³ or by about 27%, while when co-firing coal with biomass these emissions were reduced from 720 mg/m_n³ to 480 mg/m_n³, [15]. The efficiency of the use of natural gas in co-mbustion of the basic fuel as a function of the process temperature is presented in Figure 2 - right, with the results referring to energy share of the natural gas of 10% of the total energy input into the combustion chamber.



Figure 3. NO_x emission (left) and influence of temperature on NO_x emission (right) for fuels of different composition burning with natural gas

On the previous diagram (right) NO_xemission for combustion with reburning technology with natural gas is compared with the emissions measured during combustion of the coal mixture U100 without the application of natural gas. It can be noted that NO_x emission for U100 with reburning technology with 10% gas content me-asured by energy (UP) and at temperature of 1450 °C is 615 mg/m_n³, which is approximately 30% less than the emission without the use of natural gas. Also, when coal and biomass were co-fired under the same conditions (UB(95)P) and at temperature of 1450 °C, the emission was 602 mg/m_n³, which is practically the same 30% less than the emission when combusting only U100 (866 mg/m_n³), [15].

The effect of natural gas as reburning fuel on the SO₂ emission of combustion of basic test solid fuels at a pro-cess temperature of 1350 °C is shown in Figure 4 - left. In general, for combustion under these conditions, there is a slight decrease in the SO₂ emission. For example, the SO₂ emission of reburning with 10% of natural gas is in average by 325 mg/m_n³ lower than the SO₂ emission of combustion of coal blend U100. During these test regimes with natural gas, a very low level of CO emission was recorded, especially for combustion of basic fuels with smaller granulation and at a temperature above 1400 °C - the CO emission is practically negligible, below 5 mg/m_n³, Figure 4 - right.

The previous phenomenon can be explained by intense reaction, that is, by the combustion of natural gas in the reburning zone in which the products of incomplete combustion (such as CO) that were formed in the primary combustion zone in which the combustion air is lacking (λ <1, the sub-stoichiometric zone) burn additionally, [15].



Figure 4. Emission of SO₂ (left) and CO₂ and CO (right) for different fuel compositions with reburning technology with natural gas

Also, it is a known fact that the combustion of natural gas, in comparison with the combustion of liquid and especially solid fuels, is accompanied by the lowest CO_2 emissions. This was also reflected in the results of the test regimes for combustion of selected solid fuels with reburning technology with natural gas - see Figure 5. In this case, the CO_2 emission averaged about 0.231 kg/m_n³ and is lower by almost 0.020 kg/m_n³ in relation on emissions from the combustion of U100 coal mixture without the application of natural gas.

Comparative overview of the efficiency of primary measures in the combustion chamber on the NO_x emission: The effects of primary measures in the combustion chamber for fuels U100 and U95B5, for the process temperature range from 1350 to 1450 °C are presented in Figure 5 - left. There is a noticeable decrease in emissions at a temperature of 1450 °C, from 1154 to 615 mg/m_n³ or about 47% relative - these results refer to

 NO_x emissions for combustion of coal U100. The NO_x emission for co-firing coal with biomass (U95B5) with the application of different primary measures in the combustion chamber, including the efficiency of these measures by reducing this emissions relative to those in conventional combustion, is presented in Figure 5 - right.



Figure 5. Effects of application of primary measures in the combustion chamber on coal combustion in function of temperature (left) and for co-firing of coal with wood biomass at the same temperature (right)

From the results presented in the previous figure, it is concluded that the use of natural gas as an additional fuel is the most effective primary measure for the reduction of NO_x emissions. Also, in relation to other primary combustion measures, the impact of the combustion temperature on this measure is the smallest: the emission difference for process temperatures of 1450 °C and 1350 °C using reburning technology by introducing natural gas is on average 135 mg/m_n³, whereas this difference in the application of staging combustion air as primary measure is on an average of 180 mg/m_n³, [15].

IV. CONCLUSION

Based on the results of the research, the following conclusions can be drawn regarding the impact of the staged air intake into the combustion chamber and the use of natural gas as additional fuel on the emission of flue gas components in the co-firing of coal with waste woody biomass:

- NO_x emissions in co-firing coal with waste woody biomass is at the emission level at combustion of coal blend. There is practically no change in this emission with the change in the fraction of woody biomass in the mixture. The average difference in NO_x emissions depending on the mode of combustion air is 250 mg/m_n³: for the classical air intake the average emission is 942 mg/m_n³, while for the mode withair staging it is 692 mg/m_n³.
- The SO₂ emission is slightly reduced by increasing the fraction of woody biomassand practically does not depend on the mode of combustion air intake the emissions are high and generally over 5000 mg/m_n³.
- By increasing the fraction of woody biomass in the mixture, the CO_2 neet emission is proportionally reduced, which at the 10% biomass in the blend is 0.233 kg/m_n³.
- When co-firing coal-biomass mixture with natural gas as additional fuel (UB(95)P10) at temperature of 1450 °C, the measuredNO_x emission is 602 mg/m_n³, which is about 30% less than emission with coal U100 which is 866 mg/m_n³.
- When using the natural gas in reburning test regimes there is a slight decrease of SO_2 emission. Burning the coal U100 with 10% of natural gas, the SO_2 emission is on average about 325 mg/m_n³ is lower than the SO_2 emission when burning coalonly.
- In combustion regimes with natural gas, CO emission is very low, especially when at temperatures above 1400 °C, where CO emissions are practically negligible: below 5 mg/m_n³. In doing so, CO₂ emissions are lower by almost 0.020 kg/m_n³ compared to emissions when combusting U100 coal mixture without natural gas.
- By using natural gas in combustion of coal U100 at temperature of 1450 °C, the emission of NO_x decreased to 615 mg/m_n³ or by almost 50% compared to the emission of conventional combustion: 1154 mg/m_n³.
- When using natural gas as additional fuel, compared to other primary combustion measures, the impact of the process temperature on the NO_x emission is the smallest: the difference in emissions at temperatures of 1450 and 1350 °C is on average 135 mg/m_n³ the difference for the application of the combustion air staging as primary measure is on average about 180 mg/m_n³.
- Efficiency of primary measures by staging the base fuel and using natural gas as additional fuel weakens with rising process temperatures. Efficiency of the staging basic fuel at 1450 °C is 21% compared to 32% at 1350 °C, while the efficiency of natural gas utilization for those temperatures is 39% and 46% respectively.

In doing so, efficiency of the application of staging combustion air is practically the same for both observed combustion temperatures.

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