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Efekti primjene prirodnog gasa pri sagorijevanju čvrstih goriva u letu - *reburning* tehnologija

Effects of Natural Gas Application in Combustion of Pulverized Solid Fuels - Reburning Technology

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Rezime **-** U sadašnjem vremenu je, istovremeno projicirano i za blisko buduće vremensko razdoblje, na globalnom nivou veoma izražena energetska tranzicija koja u osnovi podrazumijeva fazno i značajno smanjenje primjene fosilnih goriva, naročito uglja u energetici i industriji, sve do konačnog prestanka te primjene. Na tom putu energetske tranzicije, sve značajniju ulogu generalno dobijaju obnovljivi izvori energije te alternativna goriva, pri čemu su, paralelno tome, i zahtjevi prema energetskim sistemima u pogledu povećanja energetske efikasnosti i okolinske prihvatljivosti neizostavni kriteriji. S tim u vezi se naročit napor usmjerava na proširenje portfolija primarnih goriva u postojećim velikim energetskim sistemima kakve su npr. termoelektrane na ugalj. Stoga se danas u velikim kotlovima, s ciljem i smanjenja emisije NO_x, skoro pa neizostavno primjenjuje stepenovani privod vazduha za sagorijevanje kao primarna mjera u ložištu (*air staging*). Uz prethodnu mjeru, veoma brojni su primjeri i stepenovanog privoda osnovnog goriva u zonu sagorijevanja (*fuel staging*), te ipak manji broj primjera primjene trećeg ili dodatnog goriva, kakva je npr. primjena biogasa ili prirodnog gasa pri sagorijevanju osnovnog čvrstog goriva u letu (*reburning technology*). U opštem slučaju, ovim mjerama istovremeno se povećava efikasnost konverzije primarne energije iz goriva i smanjuje emisija nepoželjnih komponenti u okolinu. Do koje mjere su izraženi prethodni efekti pri primjeni prirodnog gasa kod sagorijevanja bosanskohercegovačkog uglja i otpadne drvne biomase, pokaza-no je u ovom radu kroz rezultate obavljenih istraživanja: pri takvom sagorijevanju dolazi do dodatnog smanjenja emisije NO_x i to proporcionalno udjelu gasa - pri procesnoj temperaturi 1350 °C i pri 10% energetskog udjela gasa pri sagorijevanju sa ugljem, u odnosu na emisiju bez primjene dodatnog goriva, evidentirano je smanjenje emisije NO_x za više od 250 mg/m_n^3.

Ključne riječi - ugalj, drvna biomasa, prirodni gas, sagorijevanje, emisija NO_x.

Abstract **-** The energy transition basically implies a phased and significant reduction in the use of fossil fuels, until the final cessation of use in the near future. Renewable energy sources and alternative fuels are an increasingly important part of the energy transition. At the same time, it is necessary that energy systems increase energy efficiency and environmental acceptability. In this regard, great efforts are being made to expand the portfolio of primary fuels in existing large energy systems, e.g. thermal power plants. Therefore, today in large boilers, in order to reduce NO_x emission, staged air supply for combustion is inevitably used as the primary measure in the furnace (*air staging*). In addition to the previous measure, there are many examples of staged fuel supply to the combustion zone *(fuel staging)*, and still a small number of examples of the use of third or additional fuel, e.g. application of biogas or natural gas in combustion of pulverized solid fuels *(reburning technology)*. General, these measures simultaneously increase the efficiency of primary energy conversion from fuel and reduce the emission of undesirable components into the environment. The results of this paper show the extent to which the effects of the use of natural gas in the co-firing of Bosnian coal and waste wood biomass are expressed. In this process there is an additional reduction of NO_x in proportion to the share of gas - at a process temperature of 1350 °C and at 10% of the energy content of gas in combustion with coal, compared to emissions without additional fuel, recorded reduction of NO_x emissions by more than 250 mg/m_n^3.

Index Terms - Coal, Wood biomass, Natural gas, Combustion, NO_x emissions

I INTRODUCTION

The introduction of renewable fuels into regular operation, with the aim of decarbonisation or reduction of CO_2 emissions, such as waste wood biomass, is a complex and quite demanding pro-cess, not only due to the necessary technical interventions at an existing plant, but also unknowns to which such activities can result in the given concept of the furnace, i.e. the boiler. These unknowns are particularly pronounced in the combustion of mixtures of different fuels, including the behaviour of the mineral part of the fuel in this process, and the impact of this process on the environment, i.e. the emission of polluting components of flue gases. Some of these potential problems were detected in the re-search presented in [1]. Despite the previous, due to the necessity and importance of implementing the energy transition towards decarbonisation in the world, so far there are a large number of examples of energy plants adapted to the simultaneous use of different fuels (fossil, renewable and even alternative) in regular operation. Thus, for example, co-firing of coal and waste wood biomass or biomass residues from agricultural activity by the middle of the last decade put into regular operation in over 230 thermal power plants across the European Union (EU), [2]. On the other hand, a significant reduction in NOx emissions, compared to conventional combustion systems, can be achieved by using newer generation burners (vortex or flow Low NOx burners - LNB), graduated air combustion air supply (air staging, Over Fire Air - OFA), gradual introduction of basic fuel into the furnace (fuel staging), installation of double-row or multi-row burners observed by the height of the furnace and the use of additional fuels and their introduction into the furnace subsequently (eg. natural gas, biogas) - reburning technology. LNB burners work on the principle of zone or staged supply of combustion air observed at the level of a single burner - these burners result in a significant reduction in NOx emissions and as such are practically indispensable equipment in the design and construction of new energy boilers. Also, these burners are widely used for existing solid fuel boilers as a replacement for conventional flow-type burners, [3]. The scientific paper [4] presents the results and effects of staged air supply on NOx emissions - dried lignite was used as fuel. It has been shown that in this way there is a significant reduction in NOx emissions - the results relate to the study of the influence of excess air coefficient, combustion air distribution and the distance of the OFA portion of air into the reaction zone relative to the burner. Similar to the previous one, reference [5] states that, with Low-NOx burners, NOx emissions can be reduced by one-fifth, but that, due to the high combustion temperature, these emissions are still high (1036 mg/mn3 at 6% O2 in dry flue gases), and for further reduction of NOx emissions, a zone or staged air supply - OFA is recommended. Also, the positive effect of staged air supply on NOx emission reduction is presented in references [6-7] where the results refer to coal combustion, as well as in references [8-12] for the case of co-firing coal and bio-mass. The paper [13] presents interesting results of research on coal combustion as a basic fuel with subsequent reburning of different types of waste wood biomass with an emphasis on the application of such a measure in the furnace to NOx emissions.

By subjecting coal blends, as well as mixtures of coal and waste woody biomass, to combustion at different temperatures 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 consequently 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 conditions of solid fuel pulverized combustion technology with a staged combustion air supply, [2]. In this case, the total combustion air is divided into four streams or portions: primary, secondary, tertiary and OFA - see the picture of the experimental plant given behind.

II Experimental Set-up

Experimental line: Automatic Controlled Tube Reactor experimental plant has been installed at Laboratory for Coal and Biomass Combustion (University of Sarajevo - Faculty of Mechanical Engineering, Department of Energy). This plant enables testing of combustion characteristics of different fuels under different ambient and technological conditions. Principal schema of the upper part of the plant with designated ancillary systems for classical and stepped fuel supply (basic and additional fuel) and zone or stepped supply of combustion air to the reaction pipe is given in Figure 1. - see also [2] and [14-16].

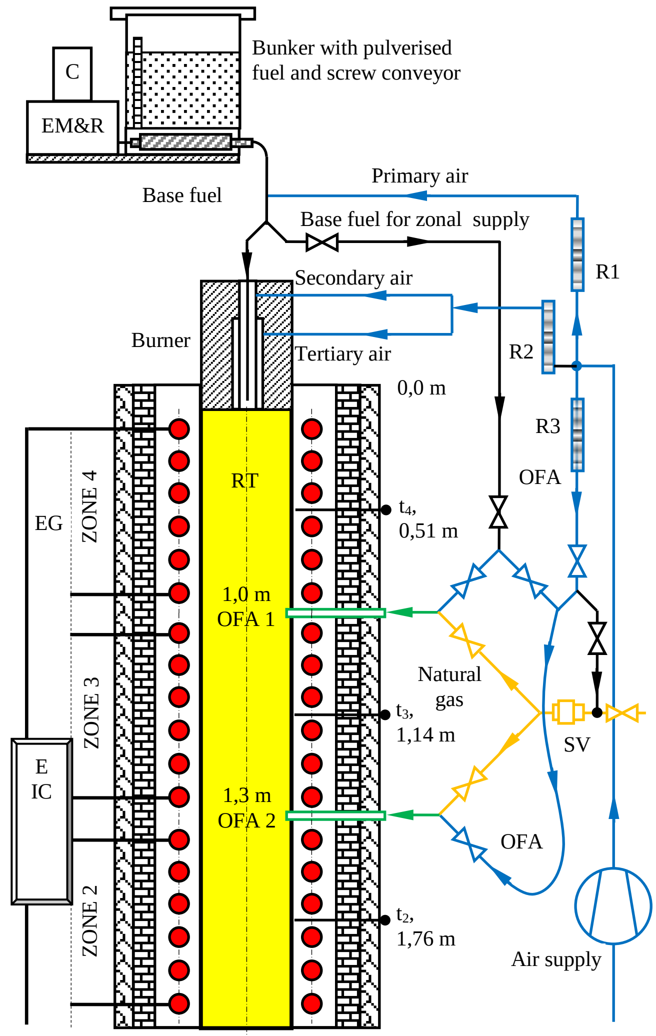

Figure 1. Principal scheme of the experimental plant: Upper part of the experimental plant with designated systems for classical and stepped fuel supply (basic and additional fuel) and zone or stepped combustion air supply to the reaction tube (RT) - *air staging, fuel staging, reburning* technology

Briefly, the plant is designed to operate in a wide temperature range (from ambient temperature to 1560 °C) and in conditions of different amount and distribution of basic fuel and air combustion, including the possibility of testing *reburning* combustion technologies using additional fuel, such as natural gas. Basically, the research provides data related to combustion efficiency, deposition intensity and characteristics of deposits from the reaction zone as well as slag and ash at the reactor outlet, and emissions of flue gas components: O_2, CO, CO_2, NO, NO_2, NO_x i SO_2, [2].

Mixtures of solid fuels for research: For the purpose of experimental research on the effects of natural gas on the combustion characteristics of pulverized solid fuels (coal mixtures or mixtures coal and waste woody biomass) in terms of emissions of flue gas components into the environment, especially nitrogen oxides (NO_x=NO+NO_2), the following matrix of basic test mixtures of solid fuels was formed, [2] - Table 1:

- The coal mixtures 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 be-hind the mills during real operation of the Unit 5 in TPP Kakanj.
- Mixtures of previous test fuel U100 and waste wood bio-mass (sawdust) **B**100 - fuel code: U95**B**5, mixture in which the mass fraction of coal U100 95% and biomass **B**100 5%. The sawdust is a mixture of beech and spruce formed during the primary processing of wood, in an approximate ratio of 1: 1 by weight. This test fuel (U95**B**5) was also excluded in the operation behind the mills of block 5 in TPP Kakanj.
- A mixture of brown coal from the mines Kakanj, Breza and Zenica in a mass ratio of 70:20:10, respectively - fuel designation: K70B20Z10. This coal mixture was formed after drying and grinding the components in a laboratory mill.

Table 1. Basic properties of test fuel, [15]

Fuel/Label	U100	U95**B**5	K70B20Z10
Moisture, %	13.90	19.06	10.71
Ash, %	37.88	34.33	40.84
Volatiles, %	28.97	29.32	27.71
C_{fix}, %	19.25	17.28	20.73
Combustible, %	48.22	46.60	48.44
Carbon, %	36.62	32.12	34.48
Hydrogen, %	2.60	2.20	2.33
Sulfur, %	2.06	1.57	2.41
Nitrogen, %	0.72	0.81	0.75
Oxygen, %	10.22	9.91	8.48
HHV, kJ/kg	13351	12651	13898
LHV, kJ/kg	12496	11759	13171

Basic settings of test regimes: The previously mentioned test solid fuels (mixtures of coal and a mixture of coal and wood bio-mass) 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 - pulverized-fuel technology with slag tap furnace: 1350÷1450 °C. The set of research aimed to gain as much knowledge about the characteristics of co-firing coal and biomass in the conditions of *reburning* technology with natural gas with a particular emphasis on emissions of flue gas components, primarily NO_x. Test regimes of co-firing with natural gas as an additional fuel, were performed with 5% and 10% energy content of gas. These test tests for natural gas combustion were assigned fuel designations: UPn, U**B**(95)Pn i KBZ70Pn, where "n" represents the energy share of natural gas in the total energy input into the furnace through fuel. In all test regimes, the total coefficient of excess air was λ=1.15 while the distribution of that total air was with the ratio λ_1/λ=0.95/1.15 - a substoichiometric amount of air is supplied to the burner, λ_1=0.95.

III Research Results With Discussion

The effects of the use of natural gas as a fuel reburning on NO_x emissions during the pulverized combustion of these test solid fuels and at a combustion process temperature of 1350 °C are shown in the following Figure 2. At the same time, for the given solid fuels, the NO_x emission values for the case of 0%, 5% and 10% of the share of natural gas in combustion are also given - observed by the share in the total energy input into the furnace. In general, in relation to NO_x emissions without the use of gas as an additional fuel (0%e), a decrease in that emission was recorded. For example, NO_x emissions from the combustion of U100 fuel when reburning with 5%e gas are lower by 120 mg/m_n^3 or 18%, while the same emission at 10%e energy content of gas is lower by more than 170 mg/m_n^3 or by about 27%. The situation is similar with the co-firing of coal and biomass with natural gas – fuel U**B**(95)Pn: at 10%e gas content, NO_x emissions are lower by approx. 29%. It is important to note that NO_x emissions from co-firing coal and biomass are practically at the level of emissions from coal combustion - compare NO_x emissions for fuels U100 and U95B5 in Figure 2 and then related emissions from firing of the same fuels but with natural gas (fuels marked UPn and U**B** (95) Pn).

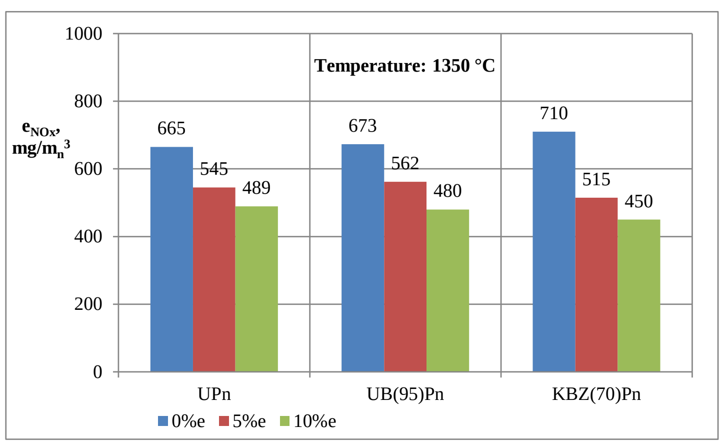

Figure 2. NO_x emissions at a combustion process temperature of 1350 °C for fuels of different composition and when using natural gas as an additional fuel - *reburning* technology

Similarly, the effects of natural gas on NO_x emissions in combustion of pulverized solid fuels but as a function of process temperature are presented in the following Figure 3, with all results referring to the case when the energy content of combustion gas is 10%. This diagram also shows the NO_x emission values when burning basic test fuels (without natural gas) at 1350 °C - compare the values with Figure 2, but also the emission values at 1400°C and 1450 °C for U100 fuel. Thus, it can be seen in the diagram that the NO_x emission for reburning technology with natural gas in the entire considered temperature range is compared with the emission measured during the combustion of U100 coal mixture without the use of natural gas. For example, it is noticeable that the NO_x emission for U100 fuel with reburning technology with 10%e gas content (UP10) and at temperature of 1450 °C is 615 mg/m_n^3, which is approximately 30% lower emission compared to the comparative emission without application natural gas. Also, the co-firing of coal and waste woody biomass in these conditions (UB(95)P) and at a temperature of 1450 °C measured emissions of 602 mg/m_n^3, which is practically the same 30% less than the emissions from combustion only coal marked U100 (866 mg/ /m_n^3), [2].

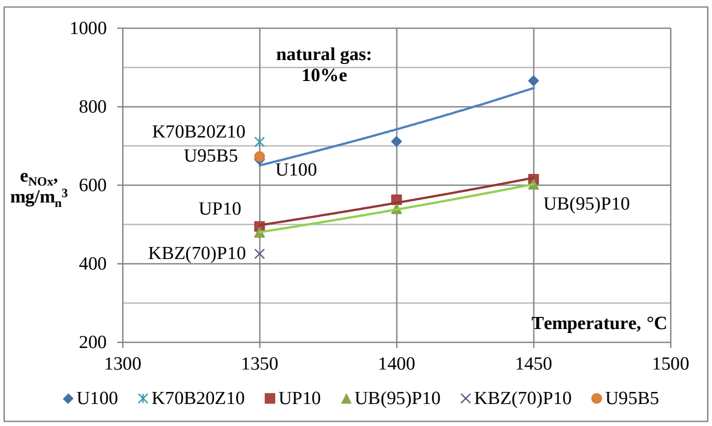

Figure 3. Effects of natural gas application on NO_x emissions at variable process temperature and co-firing of solid fuels of different composition - *reburning* technology

Other effects of natural gas application on the combustion characteristics of the subject fuels were investigated during these test regimes. For example, the impact of natural gas as a fuel reburning on SO2 emissions during the combustion of basic test solid fuels at a combustion process temperature of 1350 °C was qualitatively detected - see the results of the research in the following Figure 4.

In general, combustion in such conditions and with regard to the level of that emission, there is a practically insignificant reduction in SO2 emissions. For example SO2 emissions from reburning with 10%e of natural gas are on average about 325 mg/mn3 lower than SO2 emissions from the combustion of U100 coal mixtures.

At the same time, in all these test regimes of co-firing with natural gas, a very low level of incomplete combustion was recorded, validated through CO emissions, especially in combustion of basic fuels of smaller granulation and combustion process temperature above 1400°C - in these cases CO emissions are practically negligible, below 5 mg/mn3 - see the results in Figure 5.

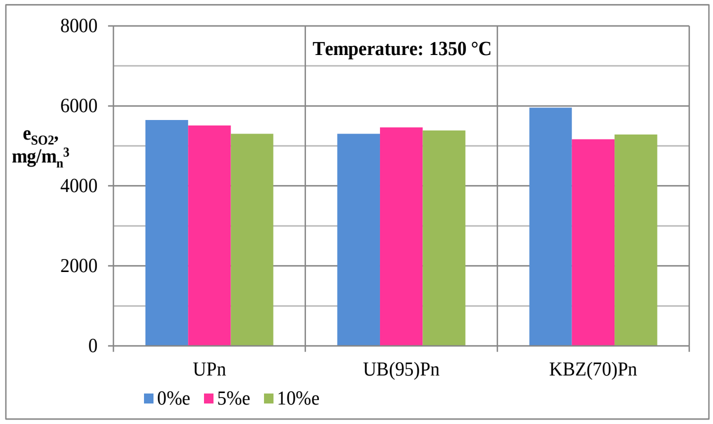

Figure 4. SO_2 emissions at a combustion process temperature of 1350 °C for fuels of different composition and when using natural gas as an additional fuel - *reburning* technology

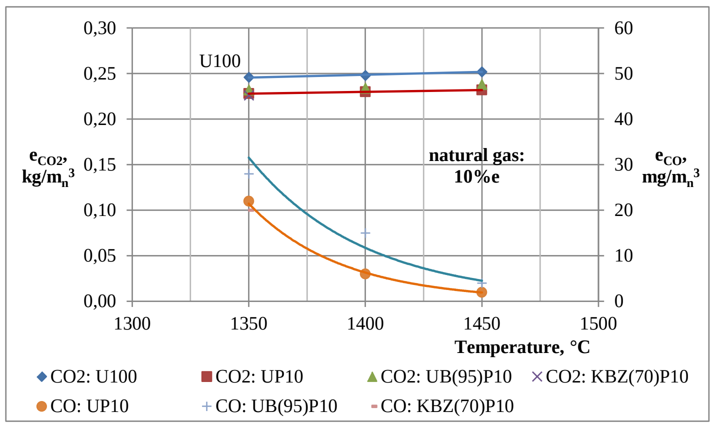

Figure 5. Effects of natural gas application on CO_2 and CO emissions at variable process temperature and co-firing of solid fuels of different composition - *reburning* technology

The above phenomenon can be explained by a very violent reaction, i.e. the combustion of natural gas in the reaction zone of re-burning (reburning zone) in which the local temperature rises, which further burns C_{fix} in solid fuel particles and incomplete combustion products (such as CO and/or C_mH_n) which occurred in the primary combustion zone in which there is a lack of combustion air (from the burner outlet to the place of introduction of OFA air portion is $\lambda<1$, substoichiometric zone), [2]. It is also known that the combustion of natural gas, in relation to the combustion of liquid and especially solid fuels, is accompanied by the lowest CO_2 emissions. This was reflected in previous results in the set of subject test regimes for combustion of selected solid fuels with reburning technology with natural gas - see Figure 5 and compare CO_2 emission values during reburning with gas (e.g. fuel UP10, UB(95)P10) in relation to CO_2 emissions generated during the co-firing of coal mixtures without the use of natural gas - fuel marked U100. During reburning, CO_2 emissions average about 0.231 kg/m_n^3 and are lower by almost 0.020 kg/m_n^3 compared to emissions from the co-firing of U100 coal mixture without the use of natural gas.

In parallel with the previous determination of the values and emission levels of essential components in flue gases, during the test regimes the quantified behaviour of the mineral part from fuel (ash) in the given ambient and technical-technological settings of combustion of pulverized solid fuels (coal, woody biomass) and when using natural gas as an additional fuel for subsequent co-firing - *reburning* technology. In this regard, for the purpose of visual and appropriate chemical analysis, samples of ash deposits were collected from the reaction tube, which, during combustion, are collected on uncooled ceramic tablets and samples of slag and fly ash from the bottom of the reactor. In this regard, as an example, the following Figure 6 shows the exempted deposit from tablets and slag from the bottom of the furnace in the co-firing of coal and waste woody biomass U95**B**5 in terms of reburning technology with 5% natural gas and process temperature of 1400 °C - the fuel designation for this test mode is U**B**(95)P5.

Figure 6. Basic test mode settings: UB(95)P5, 1400 °C, λ1/λ=0.95/1.15; ash deposit on ceramic tablet - left, slag from reactor bottom/furnace - right

Compared to other solid samples of coal combustion products and waste wood biomass, with a different share of biomass in the mixture and at the same process temperature, in which, as in this case, are also quite similar larger molten deposits, and it can be concluded that intensification of slagging at this combustion temperature should not be associated with the content of woody biomass in the mixture. In this case, too, the reburning technology has the same form and structure of deposits as in the test regimes without the use of natural gas. Namely, the initial formation of larger molten deposits at a combustion temperature of 1400 °C can further escalate because new layers accumulate on these initial molten deposits and a stepped structure of hard molten deposits is formed. The tablet also contains a part of the deposit that is not fused - see previous Figure 6. Therefore, comparing the condition and structure of solid samples of combustion products for different fuels and for different temperature and technical-technological conditions of combustion, it can be concluded that natural gas does not worsen the state of the combustion process from the aspect of the behaviour of solid fuel ash in that process.

IV Conclusion

The purpose of this paper is to present the effects of simultaneous use of natural gas in the process of combustion of pulverized solid fuels, and to present the results of research on applied reburning technology at the experimental plant. Based on the obtained and partially presented research results, the following concrete conclusions can be made regarding the effects of natural gas as an additional fuel on the emission of flue gas components during co-firing of various mixtures of coal and waste woody biomass:

- When reburning co-firing with natural gas as an additional fuel, the NO_x emission is reduced - this reduction is proportional to the share of natural gas that is subsequently introduced into the reaction zone. For example, at a process temperature of 1350 °C and at 10% energy content of gas, in relation to emissions without additional fuel, a reduction in NO_x emissions by more than 250 mg/m_n^3 was recorded - NO_x emissions for coal mixtures K70B20Z10 reduced from 710 to 450 mg/m_n^3 or about 37%.
- When co-firing coal-biomass mixture with natural gas as additional fuel (U**B**(95)P10) at temperature of 1450 °C, the measured NO_x emission is 602 mg/m_n^3, which is about 30% less than emission with coal U100 which is 866 mg/m_n^3. At a combustion temperature of 1350 °C, a NO_x emission of 425 mg/m_n^3 was measured for the K70B20Z10 coal mixture, which is about 40% less than the comparative emission.
- When using the natural gas in reburning test regimes there is a slight decrease of SO_2 emission. Burning the coal U100 with 10%e of natural gas, the SO_2 emission is on average about 325 mg/m_n^3 is lower than the SO_2 emission when co-firing mixture coal only.
- 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_n^3. In doing so, CO_2 emissions are lower by almost 0.020 kg/m_n^3 compared to emissions when combusting U100 coal mixture without natural gas.
- Primary measures in the furnace, staged air supply and reburning technologies with natural gas, do not have a negative impact on the process of formation and structure of deposits in the boiler and therefore do not worsen the tendency of base fuel ash, coal or coal mixture with waste woody biomass boiler heating surfaces.

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