Laboratory Research On The Combustion Of Brown Coal And Woody Biomass In An Oxygen-Enriched Atmosphere – Contribution To The Development Of Ccs Technologies

Kenan Kadić¹, Nihad Hodzic²

 ¹ Elektroprivreda BiH d.d. - Sarajevo Power utility, Power plant Kakanj - Kakanj 72240 Kakanj, Bosnia and Herzegovina
² University of Sarajevo - Faculty of Mechanical Engineering, Energy Department Vilsonovo setaliste 9, 71000 Sarajevo, Bosnia and Herzegovina

ABSTRACT: The increase in temperature and the increasingly pronounced change in the Earth's climate must be mitigated by decarbonizing the energy sector. The inevitable gradual reduction in the consumption of fossil fuels means the need for the use of renewable fuels. The aim of these laboratory researches is to obtain new knowledge about the characteristics of brown coal combustion with different renewable fuels, with the motive of scientific contribution to the energy transition process. Different mixtures of brown coal, waste woody biomass and Miscanthus energy crop were subjected to combustion with variable process conditions. In addition to changing the fuel composition, the tests included a significant change in process temperature (1250 - 1450 $^{\circ}C$) and air staging. With these combustion conditions, the composition of flue gases was analyzed, that is, the pollutant emissions and the efficiency of combustion were determined. The characteristics of deposits of slag and ash were also analyzed in terms of identification of slagging/fouling of the boiler heating surfaces. Despite the insufficient quality of grinding, it has been shown that even with co-firing at a temperature of 1250 °C, it is possible to establish efficient process with an acceptable content of unburnt carbon in the slag (unburnt carbon content, UBC<4%) as well as CO emissions ($e_{CO} \le 340 \text{ mg/m}_n^3$ at 6% O_2 dry), a relatively low emission of nitrogen oxides $e_{NOX} \leq 670 \text{ mg/m}_n^3$ at 6% O_2 dry. Also, the process proved to be well controlled from the aspect of possible ash slagging in the furnace. Co-firing at 1450 °C is accompanied by favorable characteristics: the process of conversion of primary energy from fuel is even more efficient (UBC $\approx 0\%$, $e_{co} < 150 \text{ mg/m}_n^3$ at 6% O_2 dry), NO emission is below 740 mg/m_n³ at 6% O₂ dry, while the ash deposit is in a molten state. In both combustion settings, the net CO_2 emission decreases in proportion to the proportion of biomass in the mixture, while the SO₂ emission is always high.

KEYWORDS: coal, woody biomass, oxygen-enriched combustion, emissions.

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I. INTRODUCTION

In recent decades, the efforts of experts, scientists and the general public in the fight against global warming and climate change as a consequence of the enormous anthropological emission of greenhouse gases, primarily carbon dioxide (CO₂), have become more and more pronounced. Evidence of this is the continuous setting of increasingly ambitious goals for limiting that emission at the global level. The agreement signed in Paris in 2015 by 195 countries of the world set the goal of keeping the increase in the average temperature on the planet at a level of less than 2 °C compared to the pre-industrial state and investing additional efforts to limit this increase to a maximum of 1.5 °C by 2050 [1]. The challenge of solving the problem of climate change involves reducing the emission of greenhouse gases as much as possible. This is actually the need for a major reduction in the generation and emission of greenhouse gases into the atmosphere originating from human activities. It is basically possible to achieve this by substituting fossil fuels with renewable or alternative fuels, i.e. decarbonisation. Namely, it is necessary in the projected period of time to significantly reduce, reduce to zero or, in an even better variant, the negative emission of CO₂ as the dominant gas that causes the greenhouse effect. In particular, the contribution of CO₂ to that effect has been more and more pronounced in recent decades, so that it currently accounts for $\frac{3}{4}$ of the total contribution of all greenhouse gases [2].

Historically, that CO_2 emission, which resulted primarily from the burning of fossil fuels, has increased dramatically since the beginning of the industrial revolution, and especially in the last 70 years, Fig. 1. The total CO_2 emission in 2020 amounted to over 34,000 million tons. Although it is at a very high level, it is still predicted that this is the time of maximum values of this emission, and that in the coming decades the CO_2

emission will at least stagnate. At the same time, it is expected that the emission reduction in developed economies will compensate for the increase in CO_2 emissions in developing countries. The majority of global greenhouse gas emissions come from a relatively small number of countries. China, the United States of America (USA) and the European Union (EU) are the top three emitters on an absolute basis. The emission of greenhouse gases, expressed per one inhabitant, is the highest in the USA and Russia. Europe is where the industrial revolution began, which led to the extensive use of fossil fuels. Although in 2020 the EU participated with only 11% in global energy consumption and with 9% of global CO_2 emissions, the share of the EU in the total accumulated emissions observed in the period from 1950 to today is estimated at about 23% [3,4].



rigure 1. Instolled growth of CO₂ emissions in the world [5].

In addition to reducing the use of fossil fuels, as the primary measure to reduce CO_2 emissions, the decarbonisation strategy also includes the development and application of new technical and technological solutions in the field of energy production. It has long been clear that reducing the use of fossil fuels is nowhere near enough to achieve the set goals. This is precisely why there is increasing emphasis and pressure for more massive investments in the development and implementation of new technical and technological solutions that will result in a reduction of CO_2 emissions. These solutions, in the general case, primarily require extensive, professionally and financially demanding research and significantly more massive affirmation and implementation. Collectively, these solutions imply the application of clean energy technologies, namely those energy technologies that result in minimal or zero, even negative emissions of CO_2 and other greenhouse gases. As such, these technologies do not involve the transformation of primary energy from fossil fuels (coal, oil and natural gas) unless accompanied by CO_2 capture and storage systems (CCS - Carbon Capture and Storage). In addition to nature-based solutions such as reducing large-scale logging and deforestation and related agroforestry technologies that involve using waste biomass as fuel and growing crops to increase soil carbon sequestration, there are also solutions that include renewable energy sources, storage and smart networks, hydrogen and CO_2 capture and storage technologies [5].

Oxy-combustion technology, in which the outgoing flue gases are recycled back into the incoming oxidant stream, is considered an energy-efficient CCS technology [6]. Further development of this technology expects economically viable applications. And while flue gas recycling technology is still developing, oxygenenriched combustion (OEC) technology can also be applied to existing plants [7]. By adding pure oxygen to the combustion air stream, the concentration of oxygen increases in the oxidant, and this is the essence of OEC technology, which is applied for the first time in the high-temperature metallurgical industry [8]. The advantages of OEC combustion technology are increased productivity and energy efficiency along with reduced exhaust gas volume and is considered one of the most promising technologies for improving CO₂ capture, especially for existing units [9,10]. Extensive research has been conducted and presented in the field of combustion in an oxygen-enriched atmosphere [11,12,13,14]. However, there is a limited number of experimental researches in this area, which again include a very wide and diverse circle of possible technological research conditions, which largely and for this reason draw different conclusions and final evaluations. In this regard, the research results presented in this paper refer to the co-firing of a mixture of Bosnian brown coal and waste woody biomass as a function of changing the proportion of oxygen in the oxidant at different process temperatures. At the same time, the results, analysis and discussion of those obtained research results are exclusively related to the technical and static characteristics of the process.

II. RESEARCH SETTINGS

II-1. Test fuels

A mixture of brown coals (K100 in Table 1) was extracted from the thermal power plant Kakanj (TE) where these brown coals originated from the central Bosnian mining basin. In the last few months, in the period preceding the exclusion of samples of these coal mixtures, the delivery of coal from certain mines to TE Kakanj was quite stable in terms of quantity, so that the average percentage ratio of brown coal in the mixture: Kakanj-Breza-Zenica-Han Bila=64-22-8-6. This coal mixture was removed directly from the plant, during the operation of unit 5, 110 MW_e and in a powder form, after partial drying and grinding in mills. Waste woody biomass (sawdust, B100) is a mixture resulting from the cutting of beech and spruce wood in sawmill in a ratio of 1:1 by mass. The K90B10 and K80B20 fuels represent a mixture of coal K100 and biomass B100 formed under laboratory conditions, with a mass fraction of biomass in the mixture of 10 and 20%, respectively.

Sample	Table 1. Fuel and Ultimate analysis, %					HV,	Proximate analysis, %			
	С	Н	Ν	S	0	kJ/kg	FC	V	А	М
K100	28.17	2.60	1.08	1.17	11.76	11119	17.34	27.49	46.15	9.08
B100	32.75	3.99	0.29	0.00	27.82	11111	8.26	56.61	0.21	31.42
K90B10	28.63	2.74	1.00	1.05	13.37	11118	-	-	41.56	11.31
K80B20	29.09	2.88	0.92	0.94	14.97	11117	-	-	36.96	13.55

FC - Fixed Carbon, V - Volatile matter, A - Ash, M - Moisture.

II-2. Laboratory plant

Electrically heated pulverised-fuel entrained flow furnace (see Fig. 1) is designed to enable the examination of a wide range of combustion characteristics of different pulverized solid fuels under different temperature, ambient and technical-technological conditions. These conditions primarily include the possible application of various solid fuels, including renewable fuels (woody biomass, biomass from agriculture, fast-growing energy crops - e.g. Miscanthus), alternative fuels (fuels originating from municipal waste, RDF - *Refuse Derived Fuel*, SRF - *Solid Recovered Fuel*), changing the coefficient of excess air/oxygen and the method of air supply to the combustion zone, application of a third or additional fuel (e.g. natural gas - reburning technology), application of additional pure oxygen ($O_2=21\div100\%$) or some other inert gas as part of the oxidant (reactant) - it is possible to investigate combustion characteristics at temperatures from a wide range, practically from the ambient temperature up to 1560 °C. Basically, with a concretely established research program, it is possible to obtain output results related to the efficiency of primary energy conversion from fuel in the combustion process at constant or time-varying temperature, deposition intensity and characteristics of ash deposits as well the characteristics of slag and ash at the exit from the reactor, and the pollutant emissions [15,16].

II-3. Settings of the test

The test fuels were subjected to an experimental in-vestigation of the static characteristics of combustion in an oxygen-enriched atmosphere. At the same time, the installation of a separate supply of pure oxygen, purity $O_2>99.95\%$, enables the performance of tests with different oxygen content in the oxidant.

According to the total content of oxygen in the oxidant, tests with air, that is, with an approximate oxygen content of 21% in the oxidant, and tests with enriched oxygen content, specifically with 27 and 35% O₂ in the oxidant, are defined. In addition to the composition of the oxidant, the tests are also defined by different temperatures, in the range from 950 to 1250 °C, which in a wider interval correspond to the pulverized fuel combustion technology with dry bottom furnace. In all combustion tests, staged supply of oxidant was carried out in the ratio $\lambda_1/\lambda=0.95/1.15$. Also in all tests, flue gas composition was continuously analyzed, and the mean value of pollutant emission was determined: NO_x, SO₂, CO₂ and CO - the emission of these components was expressed in relation to the reference content at O₂=6% dry. The set of subject research aimed to obtain as much comparative knowledge as possible about the characteristics of the combustion of brown coal and biomass, as well as mixtures of these fuels under the conditions of combustion technology in an oxygen-enriched atmosphere, with a special emphasis on the pollutant emission, including accented emission of CO₂ as the gas that dominantly causes climate change at the global level.



Figure 2. University of Sarajevo - Faculty of Mechanical Engineering, Principal scheme of the lab-scale furnace: Electrically heated pulverised-fuel entrained flow furnace, (AUCR).

III. RESULTS AND DISCUSSION

In this chapter, the results of laboratory research related to the emission of key components of flue gases (NO_x, SO₂, CO₂ and CO) are given, depending on the content of O₂ in the oxidant and the process temperature at appropriate other technological settings of combustion of test fuels. Fig. 3 presents the results for NO_x emissions. It is evident that this emission is significantly lower for biomass (fuel B100), compared to coal K100, and also the mixture of coal and biomass K90B10 and K80B20.



Figure 3. NO_x emission during the oxygen-enriched combustion of brown coal and woody biomass for different process temperatures.

For example, the NO_x emission for B100 during combustion with air (O₂=21%) and a temperature of 950 °C is 162 mg/m_n³, while under the same combustion conditions, the emission for brown coal K100 is 412 mg/m_n³, and for a mixture of coal and K80B20 biomass is 410 mg/m_n³, all at O₂=6% dry. There is also a visible trend of NO_x emission growth, for all test fuels and regardless of process temperature, with an increase in the proportion of O₂ in the oxidant. It is known that NO_x emission during the combustion of biomass B100 in the air stream (O₂=21%) at a temperature of 1250 °C is 203 mg/m_n³ at O₂=6% dry, while at the same process temperature and content of O₂=27% in the oxidant it is NO_x emission 210 mg/m_n³ at O₂=6% dry. Similar to temperature as a process parameter, the change in the content of O₂ in the oxidant affects the rate of NO_x generation - the rate of each NO_x component increases as an effect of the increase in local temperature with the increase of oxygen in the oxidant. Specifically, when burning B100 at a temperature of 1250 °C in an oxidant stream with O₂=35%, that NO_x emission is at the level of 338 mg/m_n³ at O₂=6% dry - an increase of 135 mg/m_n³ compared to the emission during combustion with air. For the other test fuels, given that the average NO_x emission is significantly higher, the increase in NO_x emission as a function of the increase in oxygen in the oxidant is even more pronounced, see Fig. 3.

The emission of SO₂ during the combustion of woody biomass B100 is equal to 0 mg/m_n^3 , regardless of the combustion temperature and the content of O₂ in the oxidant, see Fig. 4. For fuels that contain sulfur in their composition, it is known that SO₂ emissions significantly depend on the process temperature. This was confirmed in this case as well, additionally including the increase in SO₂ emissions with the increase in oxygen in the combustion atmosphere. From the results of the research, it is evident that the emission of SO₂ during the combustion of brown coal and the co-firing of the same brown coal and woody biomass in the air stream at the observed temperature is quite high and practically at the same level or slightly lower in co-firing tests with a significant proportion of biomass in the mixture (>10 %). Also, it turns out that in the co-firing tests of brown coal and wood biomass, there is a significant reduction in SO₂ emissions with an increase in oxygen in the oxidant - compared to emissions during the combustion of brown coal under the same other conditions.





Here, as expected, the highest SO₂ emissions were recorded during the combustion of the K100 brown coal mixture at the highest process temperature of 1250 °C. In addition, this emission increases significantly in conditions of combustion in an oxygen-enriched stream, primarily due to the effect of local temperature increase. For example, SO₂ emission at 1250 °C and O₂=21% is 5362 mg/m_n³ increased to 8633 mg/m_n³ at 27% O₂ in the oxidant, i.e. to even 13659 mg/m_n³ at 35% O₂ in the oxidant – all results at O₂=6% dry, see Fig 4. Furthermore, the SO₂ emission during the co-firing of the K80B20 and O₂=35% mixture is at the level of 8000 mg/m_n³ at O₂=6% dry, which is 40% lower than the K100 brown coal combustion under the same conditions.

The results from Fig. 5 show an apparent and significant increase in CO_2 emissions with an increase in the proportion of oxygen in the oxidant. This is a consequence of a significantly different composition of flue gases - an increased share of flue gas components that are produced under these conditions, and due to a decrease in the share of N₂ in the oxidant. It should be emphasized here that this is the main reason for the application of combustion technology in an oxygen-enriched atmosphere [17,18,19]. In addition, if we compare the results presented in Fig. 5, for different process temperatures, it is clear that the process temperature has no significant effect on the CO_2 emission, regardless of the oxidant composition. Also, it is visible that the CO_2 emission is slightly lower for the biomass combustion test compared to the coal combustion test and the cofiring tests of fuel mixtures, and that this trend of difference in CO_2 emission, which is up to 20%, is maintained even with the increase in the proportion of oxygen in the oxidant and the increase temperature.



Figure 5. CO₂ emission during the oxygen-enriched combustion of brown coal and woody biomass for different process temperatures.

For example, in both temperature test when combustion biomass B100 with air, the average CO_2 emission value of 0.215 kg/m_n³ at $O_2=6\%$ dry was recorded, while for brown coal K100 the emission was 0.250 kg/m_n³ at $O_2=6\%$ dry. When combustion these fuels in an oxidizer stream with 35% O_2 , the average CO_2 emission for B100 is 0.512 kg/m_n³, while for K100 the recorded emission is 0.550 kg/m_n³, all at $O_2=6\%$ dry.

Analogous to the previous one, Fig. 6 presents the results related to the CO emission resulting from the combustion of the test fuels for the test conditions. Although this emission is the lowest for biomass combustion, it is interesting to note that the average CO emission is higher for co-firing tests of coal and biomass compared to the emission for coal combustion. This phenomenon, the increase in CO emissions with the increase in the proportion of biomass in the mixture with coal, is valid regardless of the process temperature and the composition of the oxidant, Fig. 6, and can be related to the different granulation of component fuels in the mixture and the content of volatiles and total combustible substances. In addition, it is a general and expected conclusion that regardless of the fuel composition, CO emission decreases with an increase in the process temperature and an increase in the proportion of oxygen in the oxidant. A very significant decrease in CO emissions was recorded with an increase in the proportion of oxygen in the oxidant. This effect is clearly expressed for combustion tests with a 27% share of O_2 in the combustion atmosphere, and especially for tests with a share of O_2 of 35%, where practically negligible CO emission values were recorded for all tests, Fig. 6. Specifically, if the combustion of B100 fuel with air is considered in this context, the CO emission at a temperature of 1250 °C is 94 mg/m_n³, while this emission during combustion with 27% O_2 in the oxidant is 74 mg/m_n³, and with 35% O_2 in the oxidant only 12 mg/m_n³, all at $O_2=6\%$ dry.



Figure 6. CO emission during the oxygen-enriched combustion of brown coal and woody biomass for different process temperatures.

For more results and conclusions about the combustion of lignite and brown coal from mines in Bosnia and Herzegovina with different types of biomass (waste woody biomass, beech and spruce sawdust, and Miscanthus as a fast-growing energy crop) in different ratios, process temperatures and technical technological settings of combustion (e.g. air staging, fuel staging and reburning technology) [20,21,22,23,24,25]. These results, in addition to the emission of undesirable and harmful substances into the environment, also include results related to the characteristics of component fuels and fuel mixtures, the quality of mechanical fuel preparation, the efficiency of the process, the intensity and characteristics of the deposited ash in the furnace, and a more detailed analysis of the overcomposition of the ash components from the fuel in relation to the composition of slag and ash at the exit from the furnace.

IV. CONCLUSION

From the research results obtained and presented, the conclusion is that it is possible to establish highly efficient co-firing of brown coal and waste woody biomass in oxygen enriched stream, and in a wider range of process temperatures. Combustion of these fuels at the selected test temperatures in a wider range corresponds to the pulverized fuel combustion technology with dry bottom furnace, which is currently dominantly applied, but also projected for use in thermal power plants on solid fuels. In this regard, with an increase in the proportion of oxygen in the combustion atmosfere, the composition of flue gases changes significantly, i.e. the pollutant emission: CO_2 , CO_2 , NO_x and SO_2 . In this context, it is concluded that it is possible to achieve the desired effect of an apparent increase in the concentration of CO_2 in flue gases, which is the primary reason for the application of this technology for the further capture and storage of this greenhouse gas. Therefore, the CO_2 emission generally increases with the increase in the oxygen content in the oxidant, and this applies to all fuels and without significant influence of the process temperature. The increase in this emission is best reflected in the comparison of the combustion test with 35% O_2 in the oxidant compared to the combustion tests with air, where the apparent CO_2 emission increased by an average of two times. The efficiency of the process, viewed through CO emissions, is at an extremely high level. For example, in combustion tests with $35\% O_2$ in the oxidant, that CO emission averages below 20 mg/m $_n^3$ at O₂=6% dry. The emission of SO₂ during the combustion of biomass is equal to 0 mg/m_n³. Also, the NO_x emission is significantly lower compared to the emission resulting from the combustion of brown coal and the co-firing of brown coal and biomass. The presence of biomass in the mixture with coal proportionally and to a lesser extent contributes to the reduction of SO₂ emissions. The emission of SO_2 , in addition to the increase in temperature, also increases significantly with the increase in the proportion of O_2 in the oxidant, regardless of the fuel composition. The inevitability of the use of flue gas desulfurization plants is recognized from the above. In this sense, due to the significantly higher concentrations of SO₂ in the smaller amount of flue gases that are produced in combustion conditions in an oxygen-enriched atmosphere, the necessary desulphurization plant will be more compact and more investment justified. The emission of NO_x , in addition to the increase in combustion temperature, also increases with the increase in the proportion of oxygen in the oxidant, which again, in terms of the necessary denitrification of flue gases, is favorable. Additionally, when designing a system for combustion in an oxygen-enriched atmosphere, one should also take into account the possible negative consequences of increased SO_2 and NO_x emissions from the aspect of the harmful effects of these components on the elements in the boiler, as well as possible additional investment and operating costs.

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