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Optimization of process parameters in oxygen enriched combustion of biocoal and soma lignite blends by response surface methodology

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ABSTRACT

Co-combustion of coal and biomass in power plants has the potential to reduce emissions compared to burning coal alone. However, the use of biomass with coal in power plants has its own limitations. For this reason, biomass and coal are not often used together in power plants. Torrefaction is a method that can be used to eliminate / reduce all these negative effects. Torrefied biomass (biocoal) prepared under 300 °C and 30 min has similar properties to selected Turkish lignite. Existing power plants will improve CO_2 capture by using oxygen enriched combustion: a promising retrofitting option. In this study Response Surface Methodology (RSM) by using Central Composite Designs (CCD) were performed to obtain the optimal conditions for the oxygen enriched combustion (OEC) of a biocoal / Soma Lignite blends. It was found that the proportion of biocoal in the blend was the most effective parameter for the all responses. Besides, the interactions of the two factors (the oxygen concentration and the proportion of biocoal in the blend) for all responses were successfully described by the Central Composite Design (CCD) model. Also, the process of oxygen enriched combustion optimization results showed that optimum values of oxygen concentration and the proportion of biocoal in the blend to minimize the CO, NOx and the bed temperature values, and to maximize the CO_2 and combustion efficiency values were selected as 22.8 % by vol. and 37.2 % by wt., respectively. On the other hand, CO_2 concentration in the flue gas increased when 50 % biocoal is added to lignite mixture which increases energy efficiency. Since the concentration of CO_2 in the flue gases increased, the CO_2 in the flue gases can be separated and captured by using CCS technology that is considered as the most energy and cost efficient technology.

1. Introduction

Greenhouse emissions caused by fossil fuel combustion is continuously increasing. According to forecasts, global energy demand will increase by one-third from 2015 to 2040 [1]. With increasing incentives, renewables will meet the half of the growth in electricity production until 2040. Today 40 % of electricity generation is met by coal throughout the world. Coal is one of major source of gas emissions with producing almost 50 % of total CO_2 emissions. Energy demand in Turkey will increase at an annual rate of 2.1 % [2]. Turkey is highly dependent on imported energy resources and associated technologies. 73.6 % of energy consumption is met by imports [3]. Coal is the most widespread indigenous energy source of the country. According to recent reports, the total lignite reserve of Turkey is approximately 14.1 billion tons [3]. Technologies that reduce CO_2 emissions must be developed to cope with the dominant role that fossil fuels continue to play in the world.

According to the NASA report [4] in December 2020, the global average concentration of atmospheric CO_2 was about 415 ppm, up roughly 100 ppm (or up 32 %) since measurements began in 1958. Clean coal technologies have become a major research area to reduce CO_2 emissions.

Turkey's total yearlong biomass energy potential is estimated as 32

Abbreviations: AI, artificial intelligence; ANOVA, analysis of variance; BC, biocoal; BIO, red pine biomass; CCD, central composite designs; CE, combustion efficiency; CFB, circulating fluidized bed; CFBC, circulating fluidized bed combustion; df, degrees of freedom; GDF, the general directorate of forestry; HGI, hardgrove grindability index; HHV, higher heating value; IEA, international energy agency; LHV, lower heating value; RSM, response surface methodology; S, soma lignite; Std. Dev, standard deviation; TGA, thermogravimetric analyzer.

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Mtoe and the whole recyclable biomass energy potential is 17 Mtoe [5,6]. Since Turkey has the potential of sustainable forest residues between five and seven million tons per year, it is very important to convert these forest residues into combustible solid fuels. Nevertheless, traditional combustion technologies cannot be used effectively to meet the country's energy needs.

If our fuel has high sulfur properties, low calorie and high ash, then co-combustion in a fluidized bed or pulverized coal combustion may be a good choice to burn this fuel together with biomass with a suitable technology [7,8]. From an environmentally friendly and economic point of view, using an existing facility to co-firing two fuels can be one of the advantages of co-firing. Co-combustion of coal and biomass in power plants has the potential to reduce emissions compared to burning coal alone. Acar and Dincer [9] examined the environmental effects of using fossil fuels and renewable energy sources as fuel for an existing power generation plant and pronounced that coal was the most harmful. Kurkumpas et al. [10] examined the environmental and economic aspects of power plants using lignite and biomass as fuel with a life cycle analysis methodology, emphasizing the significant environmental impacts associated with the use of lignite. Oxy-fuel combustion is a technology that allows coal combustion and CO_2 capture together. Thus, this technology enables coal-based electricity generation to be carried out in a cleaner way by radically reducing CO_2 emissions [11]. In oxy-combustion, coal is burned with an O_2/CO_2 mixture instead of air. For this, the flue gases are returned at certain rates, mixed with O_2 and fed into the combustion chamber. The CO_2 rate in the resulting flue gases can reach 90 % and above. This allows the separation and capture of CO_2 in the flue gases [11]. Generally, Oxy-combustion of coal / biomass is a possible potential to generate a negative CO_2 emission in power plants [12]. Also, if we want to burn biomass with low quality coal a clean technology in terms of emissions, oxy-combustion is a promising method.

We are challenged to use wood biomass in general as a fuel for combustion / co-combustion systems because of its different structure properties and due to transportation difficulties compared to coal. The use of biomass with coal in power plants has its own limitations. For this reason, biomass and coal are not often used together in power plants. Torrefaction is a method that can be used to eliminate / reduce all these negative effects.

Previous studies have widely proposed and reported on the use of biomass blends as feedstock in the oxy-combustion process [6]. However, there are very limited pilot studies on the use of biocoal mixtures as a raw material in oxygen enriched combustion processes [13,14]. Barzegar et al. [15] showed that by burning torrefied pine wood chips with the oxy-fuel combustion method, the lignin decomposition decreased sharply, the activation energy values increased during hemicellulose degradation and remained approximately constant during cellulose degradation. Nudri et al. [16] showed using the RSM methodology to determine the optimal conditions for co-firing of biochar/sub-bituminous coal fuel mixtures that the combustion process has an efficiency of 92.16 % at 774 °C and is optimized with an air flow rate of 28.20 m^3/hr to emit 16.38 % CO_2. None of these studies has performed an oxy-combustion study of coal and torrefied biomass mixtures to further evaluate its properties as a potential solid fuel, and no statistical analysis has been performed to investigate the effects of oxygen dominance and the ratio of biocoal to coal on combustion efficiency and emissions. Optimization techniques are very important in engineering design to proliferate the biocoal in the profitability of thermal processes. Thus, optimization methods have evolved from the simplest mathematical regressions to the most advanced artificial intelligence (AI) methods [17,18].

In this study, the effect of five factors mentioned earlier with two independent variables in oxygen enriched combustion process is considered. Also, the optimal conditions for oxygen enriched combustion of red pine biocoal with Soma lignite fuel blends are determined by using the mathematical and statistical analysis, and the design of experimental technique based on RSM by CCD model. The accuracy and reliability of the results have increased as compared to the previous case and the results have been confirmed with experimental data. Statistical analysis was carried out to evaluate the rule of oxygen concentration and biocoal ratio in the fuel composition on: 1) combustion efficiency, 2) bed temperature, 3) CO_2, CO, NOx emissions. Primary aim of the present study is to propose optimal operational conditions for the co-combustion of biocoal with Soma lignite by producing less emissions. The main difference of this study from other studies in the literature is: oxygen concentration and biocoal ratio in the fuel were statistically evaluated as independent parameters and by examining the effects of these parameters on CO_2 concentration (R1), CO concentration (R2), NOx concentration (R3), combustion efficiency (R4) and bed temperature (R5), it is to reveal which one is more effective.

2. Materials and methods

2.1. Samples and characterization

Biocoal and Soma lignite were used as fuels for this study. The mentioned materials were originally below the particle size of 50 mm. They were divided into pieces of below 2 mm. The samples were not altered in any other way. Torrefied biomass (biocoal) was prepared from pieces of pine wood in the laboratory size of 5 kg per hour of the screw type "Continuous Sunscreen System" which was installed at Ege University. The details of the system are given in Keivani et al. [19]. Fuel analysis were performed according to the ASTM methods. The LECO-AC350 bomb calorimeter was used to measure the higher heating value of both lignite and biocoal according to ISO 1928: 1995 standard test method. In order to determine the approximate analysis of the fuels used in the experiments, the ASTM D 5142-04 standard test was performed using the LECO-TGA 701 Thermogravimetric Analyzer method. In terms of elemental analysis of the samples, the sulfur (S) content was determined according to the standard test methods of ASTM D 4239-05 and the nitrogen (N), hydrogen (H) and carbon (C), content was determined according to ASTM D 5373-02 [8]. The results of analysis are provided in Table 1. Standards used for analysis is given in Keivani et al. [19]. As noted in the following table, the heating value of the biocoal is higher than the Soma lignite.

The silica sand (99.6 % SiO_2) used in the bed in all combustion experiments has a particle density of 2.5 g/cm^3, a mass density of 1.8 g/

Table 1
Analysis of the samples used in the experiments.

	Soma Lignite (S)	Biomass	Biocoal
Proximate analysis			
Moisture (wt. %)	5.14 ±0.1	9.81 ±0.3	0.79 ±0.05
Volatile Matter (wt. %)	32.78 ± 0.3	73.76 ± 0.5	39.22 ± 0.2
Ash (wt. %)	40.49 ± 0.7	0.89 ± 0.04	1.50 ± 0.05
Fixed Carbon (wt. %)	21.6 ± 0.4	15.54 ± 0.8	58.49 ± 0.6
Total	100	100	100
Ultimate analysis			
C (wt. %)	42.40 ± 0.6	44.93 ± 0.4	68.90 ± 0.3
H (wt. %)	1.84 ± 0.01	5.88 ±0.03	5.78 ±0.02
N (wt. %)	0.84 ± 0.02	0.32 ± 0.01	0.63 ± 0.04
S (wt. %)	0.73 ± 0.01	0.03 ± 0.01	0.08 ± 0.03
O (wt. %)	11.51 ± 1.1	47.94 ± 1.3	23.04 ± 1.1
Ash (dry wt. %)	42.68 ± 1.5	0.90 ± 0.02	1.57 ± 0.05
Total (wt. %)	100	100	100
LHV (original kJ/kg)	10,513 ± 1.3	15,950 ± 1.5	22,388 ± 1.2
LHV (dry kJ/kg)	13,464 ± 1.2	18,588 ± 1.1	23,487 ± 1.1
HHV (original kJ/kg)	11,466 ± 1.3	17,506 ± 1.4	23,525 ± 1.6
HHV (dry kJ/kg)	14,108 ± 1.1	19,964 ± 1.2	24,574 ± 1.4
HGI	64 ± 1.5	23 ± 1.3	104 ± 1.2

Abbreviations: (HGI: Hardgrove Grindability Index, LHV*: Lower Heating Value, HHV: Higher Heating Value).

cm^3 and an average particle size of 0.33 mm. In addition, 5 kg of silica sand was used as the inert substrate in each combustion experiment. The average size of sand particles is 344 microns.

The test matrix for the OEC is shown in Table 2.

2.2. Gas analysis

Two gas analyzers were used for the combustion tests. The first is the ABB-AO 2000 online gas analyzer and the second is the GASMET-DX-4000 FTIR flue gas analyzer. ABB measures the amount of CO (0−10,000 ppm, <2%), CO_2 (0−100%, ±1%), NO (0−1000 ppm, <2%), N_2O (0−500 ppm, <2%), SO_2 (0−2000 ppm, <2%), and O_2 (0–25 %, ±1%), in the flue gas on dry basis. This analyzer was used to measure the concentration of oxygen in the enriched air. GASMET-DX-4000 FTIR measures wide range of gas components including hydrocarbons, HCN (0−1000 ppm, <2%), HCl (0−500 ppm, <2%), HF (0−100 ppm, <2%) and H_2O (0–25 % ± 1%). This analyzer was used to measure the concentration of flue gas components.

2.3. Response surface methodology and optimization

The numerous optimization method is based on evaluating the composed effects on the yield by changing several parameters together. By using multiple optimizations, results that are more reliable than single optimization can be obtained and thus, reflection of the real responses of a system can be determined. The statistical experimental design methods are used for the collective effectiveness of the parameters. One of the most obvious and complete set of mathematical and statistical experimental design methods for analyzing the effects of various independent variables is the Response Surface Method [20]. RSM provides a huge advantage by reducing the large number of experiments required to explain the effects and interactions of many parameters, and thus it is widely used by making reasonable experimental design and process optimization [21].

The most commonly used response surface designs in the literature to find optimum values are Central Composite Design (CCD) and Box-Behnken Design (BBD). As discussed by Williges and Simon [22] the most important feature that distinguishes the central composite experimental design from other design methods is the presence of axial points (α). These axial points require two extra levels of testing, low and high, for each test factor. Thus, by the fact that the factors consist of five levels, their quadratic effects are clearly expressed. The alpha term takes different values according to the desired experimental characteristics and the number of factors. In the RSM, CCD is an experimental design useful in response surface methodology to construct a second order (quadratic) model for the response variable without the need to use a full three-surface experimental experiment. The Box-Behnken design is a three-level design that does not contain a full or partial factorial design, unlike the CCD design. All design points are located in the middle of the edges of a cube or hypercube, and all of these points are located on the surface of a sphere. In these designs, effective estimation of the 1 st and 2nd order model coefficients can be achieved. In these designs, effective estimation of the 1 st and 2nd order model coefficients can be achieved. However, for this design to be used, at least three factors must be quantitative. Since there are no axis points in this design, as in the CCD design, the lower and upper limits of the factor levels are not exceeded. The number of factors to consider in this method is 2–6. In this study, the effect of different variables on the response in the study area was investigated, a principal hybrid design- Central Composite Design (CCD) with two variables was performed at three levels. The number of most important factors was set as two (i.e., O_2 concentration (A) and biocoal share in the fuel mixture (B)). These factors are optimized for the maximization of combustion efficiency (R4), and minimization of CO_2 (R1), CO (R2), NOx (R3) and the bed temperature required (R5). Range of factors studied in the CCD is given in Table 3, and they were selected with respect to the process issues and previous studies [23,24].

The total number of experiments for these two variables was 19. Nineteen experiments with five replications in center values were conducted to calculate the net error. The behavior of each response surface (Y) for two independent parameters is explained by the observed quadratic equation Eq. (1).

$$Y = \beta_0 + \sum\nolimits_{i=1}^{2} \beta_i x_i + \sum\nolimits_{i=1}^{2} \beta_{ii} x_i^2 + \sum_i \sum_j \beta_{ij} x_i x_j \tag{1}$$

where, β_0 is the regression coefficients of the independent variables, Y is the predicted response associated with each factor level combination, x_i (i = 1–2) is the levels of the independent variables, β_i is the linear effect, and β_{ii} is the quadratic effect, and β_{ij} (i and j = 1–2) is the second-order interaction coefficients.

The variance analysis (ANOVA) data were computed by Design-Expert 11.0 (trial version) in order to obtain the interaction between the process variables and the responses (i.e.; effects of oxygen concentration (A) and biocoal share in the fuel mixture (B) on concentration of CO, NOx, CO_2, bed temperature and combustion efficiency). The quality of the fit of quadratic model was expressed by the coefficient of determination (R^2), and it's statically significance was checked by the F-test in the same program.

2.4. Experimental system setup and procedure

The design capacity of the experimental system is 30 kWth. Average fuel feeding rate is 7−8 kg/hr. The setup is comprised of a circulating fluidized bed combustor, two fuel feeding hoppers, two cyclones, one return leg, primary and secondary air feeding systems, electrical heaters, ash hoppers, flue gas cooling system, a bag filter and chimney. Fig. 1 displays the schematic setup of the CFBC [7,8]. Combustor is a 4-inch AISI 310S stainless steel having 108 mm inner diameter and 6 m height. Return leg is J valve type and has two air purging points. There are two cyclone separators, induced draft, forced draft and secondary air fans. Start-up is achieved by 15 electrical heaters. 6 of the heaters are on the air supply line to heat air. The rest of the heaters are on the combustor and return leg. Screw Feeders allow fuel feeding at two different points. The feeder motor RPM controls the fuel feeding rate. Primary, secondary air and flue gas flow rate are measured by orifice flow meters. Purge air flow rate is measured by rotameters. O_2 and CO_2 flow rates are measured by mass flow controllers. The oxygen concentration in the inlet stream is analyzed by ABB Magnos 206 paramagnetic online oxygen analyzer and flue gas at the outlet is analyzed by GASMET DX 4000 FTIR. The measured flue gas components are CO_2 (0−100%, ±1%), O_2 (0–25 %, ±1%), H_2O (0–25 % ± 1%), NO_x (0−1000 ppm), N_2O (0−500 ppm), HCN (0−1000 ppm), SO_2 (0−2000 ppm), CO (0−10, 000 ppm). SIEMENS PLC and SCADA systems are controlled and measured, pressures, temperatures, flow velocities. The more detailed explanation of the system and operation procedures can be found in Varol et al. and Atimtay et al. [7,8].

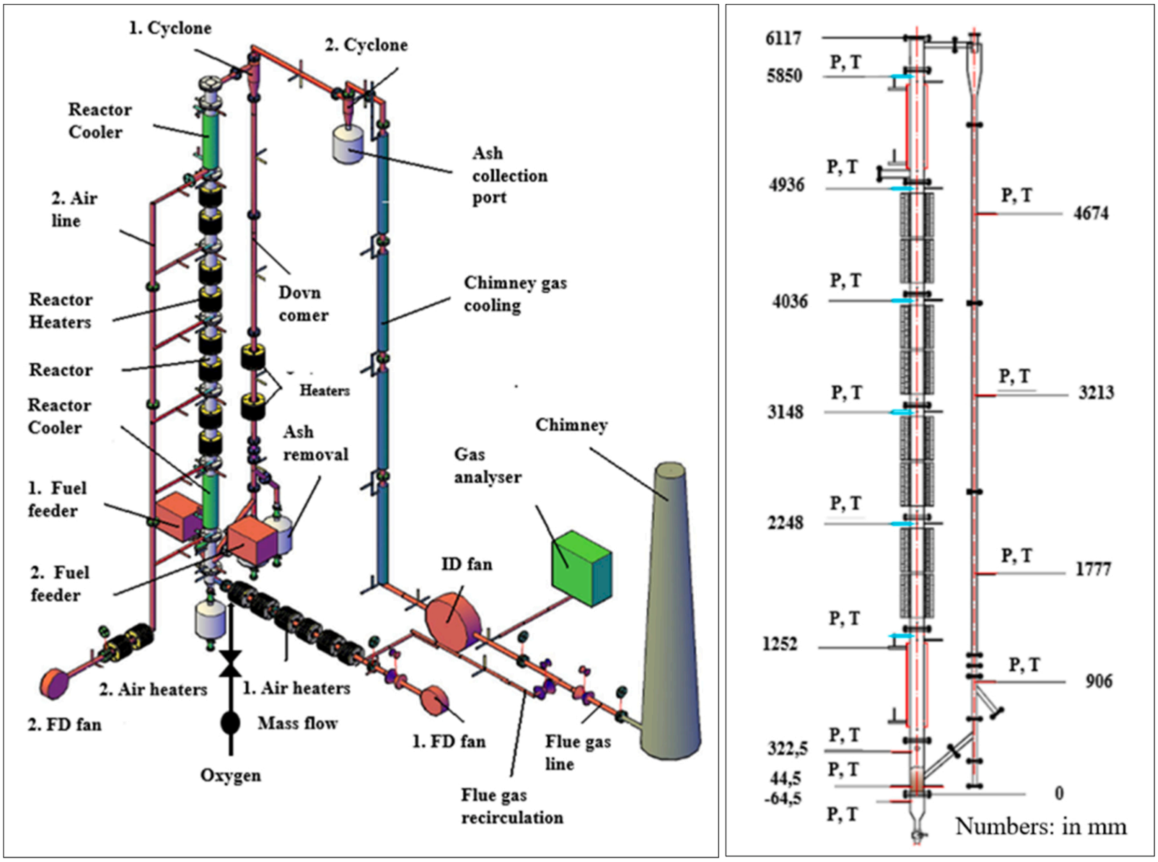
The combustion process was carried out under oxygen-enriched air with oxygen concentrations of 21, 23, 26, and 28 % and the excess air ratio (λ) was 1.4 throughout the experiments. The excess air ratio was adjusted by controlling the amount of feeding fuel. In order to adjust the oxygen concentration at the desired levels, an oxygen tank was connected to the inlet system and the flow was controlled by a mass flow controller and gas analyzer. Each test was started with air combustion

Table 2
Experimental matrix of (OEC) studies.

Fuels (Soma lignite:S; Biocoal: BC), (% by wt.)	O_2 ratio in air (% by vol.)
S	21, 23, 26, 28
90 % S + 10 % BC	21, 23, 26, 28
80 % S + 20 % BC	21, 23, 26, 28
70 % S + 30 % BC	21, 23, 26, 28

Table 3
Range of factors studied in the CCD for Oxygen Enriched Combustion (OEC).

Factor	Name	Units	Low grade	High grade	*α*	Mean	Standard diversion
A	O_2 concentration	% By vol	21	28	1.414	24.58	2.61
B	Biocoal share in the fuel mixture	% By wt.	0	50	1.414	23.68	18.02



(a) (b)

Fig. 1. Experimental system (a: Schematic of the CFBC system; b: temperature (T) and pressure (P) inside the combustor and return leg).

(21 % O_2) and after the system was reached to the steady state and the measurements were recorded, the oxygen was gradually introduced to the system to reach to 23 % oxygen concentration. After the system reached to steady state, the measurements were recorded. This process was repeated to the oxygen concentrations of 26 % and 28 % as well.

Fig. 2 depicts a typical temperature profile during combustion experiments. Each experiment had a duration of approximately 14–18 h. Bed pressure drop control was performed continuously during the test. The temperature was kept constant between 800 and 900 °C during the experiments, which is justified for CFB combustion. During combustion tests, biocoal in the fuel blends was set to 0%, 20 %, 30 % and 50 % by wt. Fig. 2 shows a rapid decrease in temperature due to obstruction or closure of the power supply system.

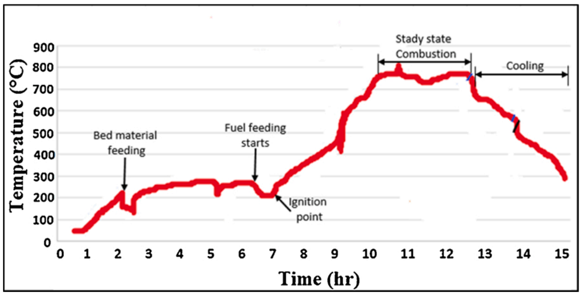


Fig. 2. Temperature profile of an experiment-fuel feeding during fluidized bed combustion system (CFBC).

Liu et al. [25] used the following formula to calculate combustion efficiency:

$$\eta_{CE} = 100 \times \frac{CO_2\% \text{ in flue gas}}{(CO_2 + CO)\% \text{ in flue gas}} \tag{2}$$

3. Results and discussion

3.1. Model analysis using CCD

Simulation runs which used different O_2 concentration (A) and biocoal share in the fuel mixture (B) together with calculated CO_2 (R1), CO (R2), NOx (R3), combustion efficiency (R4) and the bed temperature (R5) responses after simulations are given in Table 4.

Table 4 shows the data of the studied effects of two independent variables, O_2 concentration (A) and biocoal share in the fuel mixture (B) on the five responses CO_2 (R1), CO (R2), NOx (R3), combustion efficiency (R4) and the bed temperature (R5). The data in Table 5 were run through RSM to construct an empirical model for the representation of emissions in terms of O_2 concentration and biocoal share in the fuel

Table 4
Design matrix and results of simulation runs based on the CCD with calculated responses.

Run	Factor 1 A (O_2 concentration) (% by vol.)	Factor 2 B (Biocoal share in the fuel mixture) (% by wt.)	R 1 CO_2 concentration (% by vol.)	R 2 CO concentration (mg/MJ)	R 3 NOx (mg/MJ)	R 4 Combustion Efficiency E(%)	R 5 The bed Temperature T(°C)
1	28	30	19.7	100.4	229.7	98.9	848.4
2	26	20	15.7	167.8	185.6	96.9	862.4
3	28	0	16.2	295	199.8	96.3	873.3
4	26	20	15.7	167.8	185.6	96.2	862.4
5	26	30	19.1	142.2	197.3	98.4	837.3
6	21	0	12.7	520	126.8	93.7	764
7	28	50	20.9	105	243.6	98.4	827.1
8	23	30	15.4	168.1	188.3	97.8	780.5
9	28	20	16.1	152.7	218.4	97.1	871.6
10	21	50	14.9	630.8	168.4	92.9	719.9
11	23	50	19.1	490.9	199.1	94.4	760
12	21	30	12.5	370	150.3	95.9	738.3
13	23	0	14	419.6	156.3	95.6	806.2
14	26	30	19.1	142.2	197.3	98.4	837.3
15	23	0	14	419.6	156.3	95.6	806.2
16	23	20	15	265.3	177.2	96.3	795.4
17	26	0	14.8	418.8	167.6	95.4	863.7
18	26	50	19.5	328.5	217.4	96	805.3
19	21	20	11	330.5	139.3	95.1	749.6

mixture parameters in fluidized bed combustion system.

Table 5
Comparison of the results with some recent oxygen enriched combustion of coal/biomass.

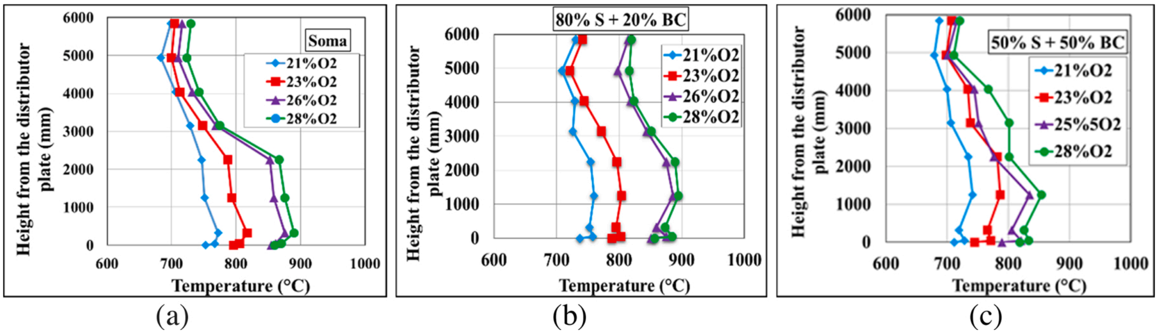
Parameter	Kayahan et al [27]	Engin et al [26]	Liu et al [23]	Varol et al [7]	This Study
Technology	OEC	OEC	OEC	OEC	OEC
Fuel	Lignite/biomass	Lignite	Coal/biomass	Coal/biomass	Lignite/biocoal
Effect of O_2 concentration on bed temperature	Increase	Increase	Increase	Increase	Increase
Effect of O_2 concentration on CO concentration	Decrease	Decrease	Decrease	Decrease	Decrease
Effect of O_2 concentration on CO_2 concentrations	Increase	Increase	Increase	Increase	Increase
Effect of O_2 concentration on NO_X concentrations	Increase	Increase	Increase	Increase	Increase

3.2. *Oxygen enriched combustion (OEC) testing*

The average bed temperature, CO, CO_2, NOx emission and combustion efficiency profiles from co-combustion of Soma lignite/biocoal fuel blends at different O_2 concentrations are presented in Fig. 4.

Fig. 3 shows that as the concentration of oxygen increases along the height of the combustion reactor, the temperature inside the bed also increases for soma lignite alone and for different lignite/biochar mixtures. In addition, a peak temperature was observed in the freeboard, since the volatile content of coal evaporated in the freeboard. The higher the volatile content of the fuel during the combustion process, the higher the temperature in the freeboard will increase accordingly.

Fig. 4 a shows the change of the average bed temperature with O_2 concentration for the co-combustion of Soma lignite and biocoal fuel blends. The average bed temperature increased as the oxygen concentration levels increased in the experiments. As Fig. 4 depicts, the bed temperature decreased slightly for the mixture of Soma lignite mixed with 20 % biocoal at 21 % and 23 % oxygen concentrations. On the other hand, when 50 % biocoal is added to lignite (Fig. 4), comparing the bed temperature at all O_2 concentrations by burning lignite results in decrease in bed temperature. Since the heat capacity of ash is higher than that of gases, and the ash content of soma lignite is higher than that of biocoal, the dense bed temperatures obtained in the combustion of soma lignite increase compared to biocoal. In addition, when high volatile content of biocoal is burned alone, it can have a negative effect on



(a) (b) (c)

Fig. 3. Temperature distribution along the combustor according to the oxygen concentration change with a) Soma lignite alone and mixtures with biocoal (b: 80 % S + 20 % BC; c: 50 % S + %50 BCE).

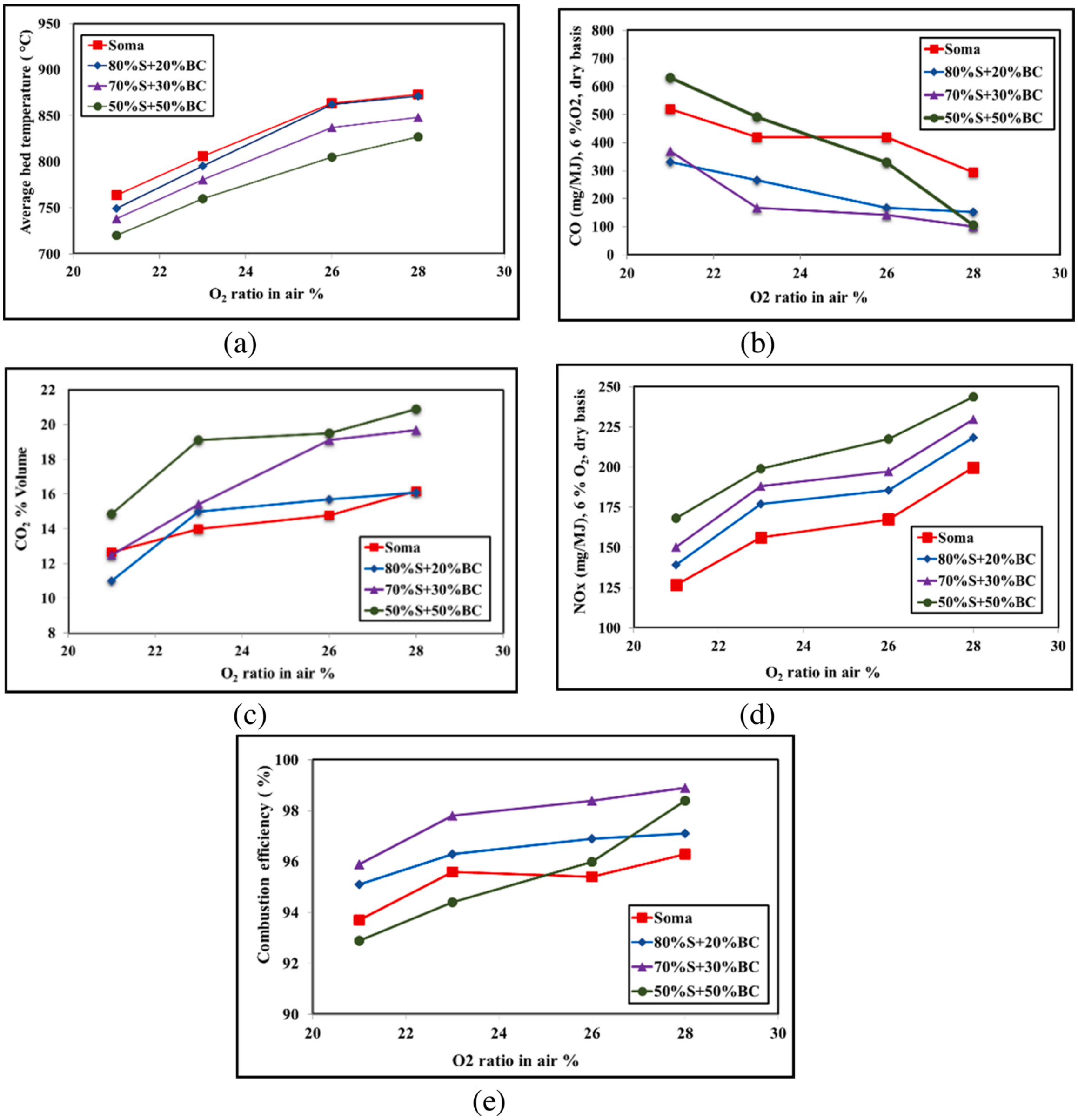


Fig. 4. Effect of O_2 concentration on (a) the average bed temperature, (b) CO conc., (c) CO_2 conc., (d) NOx emission and (e) the combustion efficiency at different O_2 concentrations.

the average bed temperature. In this case, we can improve the temperature by supplying secondary air to eliminate the problem. Fig. 4a shows that when the oxygen concentration increases, the bed temperature decreases in the combustion experiments of soma lignite alone and soma 20, 30, 50 % biocoal mixtures, since biocoal has high volatile content. This indicates that, there is a limit for biocoal contribution for better combustion. On the contrary, Liu et al. [12] showed that the addition of biomass improved the overall temperature and uniformity of its distribution in the oxy-fuel PFB because the structure of biomass is different from biochar. Better combustion in oxygen enriched air is indicated by the decreased in CO emission data. Fig. 4b shows CO concentrations with respect to O_2 ratio in air for soma lignite alone and soma 20, 30, 50 % biocoal mixtures. For the combustion of Soma lignite alone in normal air (21 % by vol. O_2), CO emission values showed a decrease from about 520 mg/MJ to 295 mg/MJ when the combustion air oxygen concentrations was increased from 21 % to 27 % by vol. For the combustion of Soma and biocoal mixture, the highest concentrations of CO was found in the combustion of 50 % by wt. biocoal and coal mixture with air. The findings were validated against experimental results in Engin et al. and Kayahan et al. [26,27].

As shown in Fig. 4c, the CO_2 concentration increases with increase in O_2 concentration for all combustion tests. An increase in the CO_2 concentrations was noted as the share of biocoal in the fuel blends increased, which indicates a better combustion. As the oxygen enrichment and biocoal fraction of the mixture increase, the addition of biochar to the soma lignites increases the synergistic effect of the combustion process. The synergistic effect accounts for the high alkaline and the earth alkaline metal oxide content of the biomass. Kayahan et al. [27] revealed that Oxygen enrichment increased CO_2 concentration in all cases and as biomass share increased in feed blends CO_2 concentrations increased as well.

As the oxygen concentration increased in the oxygen-rich environment, NOx emissions from the combustion of soma lignite alone increased from 127 mg/MJ to 200 mg/MJ (Fig. 4d). NOx emissions from combustion are higher for all blends of biocoal than for soma lignite alone. The results show it is the increase in the proportion of biocoal in the fuel composition that results in higher NOx emissions, although the nitrogen content of biochar (0.63 % by weight) is lower than the amount of nitrogen present in lignite (0.84 % by weight). Biocoal does however have a higher H/N ratio (9.17) than Soma lignite (2.19). Fuels with higher H / N ratios produce more NH_3 and increase NO formation [27]. This is a possible cause for the higher NOx emission for the blends. Previous work by other authors [26,27] should be considered. These previous studies reported that increasing the excess of oxygen supplied to the burner led to an enhancement of NO production from the fuel-N, which seems to be the situation for this test.

Fig. 4e shows the combustion efficiency variation with O_2 concentration for Soma lignite and biocoal mixtures. If balanced on a carbon system, it indicates the conversion of all carbon in the fuel to CO or CO_2.

The combustion efficiency for Soma lignite alone varies between 94 % and 96 % with oxygen concentration in air. By adding biocoal to the soma lignite, we see combustion by adding 20 and 30 % biocoal to the fuel mixture to 99.5 % and increasing it. Due to this, for 50 % additions in addition to biocoal, we see a decrease in combustion efficiency to lower levels in Soma lignite, especially at lower O_2 concentrations. This can be explained by the fact that as the temperature increases, dissociation tends to happen readily rendering the complete combustion become easier [16]. Soma has higher ignition temperature thus requires higher operating temperature (up to 900 °C) for better combustion performance. Blending 50 % biocoal into the coal decrease the average bed temperature of combustion since biocoal and its derivative compounds have higher volatile matter content than Soma, they have a significantly lower ignition temperature. The synergistic effects can lower the ignition temperature and therefore decrease the efficiency of combustion [28]. Table 5 shows the results with some recent oxygen enriched combustion of coal/biomass found in literature. The results of this study are in good agreement with that of others reported in the literature.

3.3. *Validation of model equations and statistical analysis for optimization study*

According to the regression analysis at 95 % confidence interval, the lack of fit on the value and values of p (<0.0001) of) parameter estimation was significant [22]. This indicates that a model fits better with data than linear models. Quadratic models were used to match the data observed by the least squares analysis and the following experimental models were obtained for CO_2, CO, NOx emissions, combustion efficiency and the bed temperature as given in Eqs. 3–7, respectively. The limits for the O_2 concentration and the biocoal share in the fuel blend is 28 % by volume and 50 % by wt respectively. These equations were constructed by considering co-combustion of Soma lignite / biocoal fuel blends at different O_2 concentration [23,24,30].

$$CO_2 = 16.37 + 2.57A + 2.28B + 0.6538AB - 1.36A^2 + 0.9551B^2 \tag{3}$$

$$CO = 196.83 - 138.31A - 13.70B - 078.85AB + 15.65A^2 + 193.39B^2 \tag{4}$$

$$NOx = 184.27 + 33.60A + 21.83B + 1.64AB - 0.6278A^2 + 1.72B^2 \tag{5}$$

$$E = 97.33 + 0.1335A + 3.72B + 0.8027AB - 0.3780A^2 - 1.76B^2 \tag{6}$$

$$T = 823.59 + 58.24A - 25.22B - 2.65AB - 19.33A^2 - 10.08B^2 \tag{7}$$

Where A is the O_2 concentration, B is the biocoal share in the fuel blend. The interaction between O_2 concentration and biocoal share in the fuel mixture is very significant. It has an important effect on the biocoal share in the fuel mixture and emissions in terms of O_2 concentration in co-combustion processes. Therefore, in this study a method has been developed to find the optimum point between these two parameters in co-combustion process. Given this assurance of the statistical significance of the quadratic model used to explain the experimental data at the 95 % confidence level, the model has been tested using the results of analysis of variance (ANOVA). The ANOVA results of the quadratic model for parameters of co-combustion of Soma lignite / biocoal fuel blends at different O_2 concentration were given in the Table 4.11 and Table 4.12 for R1, R2, R3, R4 and R5 responses, respectively. Statistical testing was carried out using F value and p- value for ANOVA, on the basis of the simulation values. F value must remain high, in order to achieve a replicable and confidential regression model. A low p value is expected if the importance of the developed regression is very high [29–31].

Table 6 demonstrates that regression was statistically significant at an F-value of 123.66 for CO_2 concentration; 4.16×10^5 for CO concentration; 16285.1 for NOx concentration; 47.75 for combustion efficiency and 41394.5 for bed temperature values with a very low probability values (p model 0.0001 for all responses) on the co-combustion of Soma lignite / biocoal fuel blends at different O_2 concentrations. It is known that these obtained values are higher than the minimum F value required to provide 95 % confidence level.

Very low p values with high F values, also with high R^2 values in each model, indicate that the installed models are significant and can explain the response of a good relationship between independent factors [17,29]. Given this, in the quadratic model that describes our process, sufficient accuracy 15.84 for CO_2, 20.15 for CO, 22.44 for NOx, 28.16 for combustion efficiency and 41.11 for the bed temperature values, indicate a satisfactory signal for the process. Regression was statistically significant when we realized the importance of second-order statistics (p-model 0.0001 for all responses). These results show that the response equation was suitable for the CCD experiments.

The statistical results of R1, R2, R3, R4 and R5 responses on the co-combustion of Soma lignite / biocoal fuel blends at different O_2 concentration were given in the Table 7. The accuracy of the model was checked with a coefficient of determination R2. According to the ANOVA results, the model explains the high values of 88.93 % for CO_2, 93.23 % for CO, 94.19 % for NOx, 98.15 % for combustion efficiency and 98.39 % for the bed temperature. An acceptable agreement of these results with the adjusted determination coefficient is deemed necessary.

In this study, the adjusted R^2 values of 84.67 % for CO_2, 90.62 % for CO, 91.95 % for NOx, 96.31 % for the combustion efficiency and 97.77 % for the bed temperature values were found. R2 values advocate an excessive correlation between observed values and predicted values. This indicates that the regression model provides a plausible explanation of the relationship between the three independent variables and the two responses [30,32].

3.4. *Parametric interactions of the factors on each response*

Fig. 5 describes the response surface profiles for the calculated R1, R2, R3, R4 and R5 values of co-combustion of Soma lignite / biocoal fuel blends at different O_2 concentrations and biocoal shares, respectively. There are significant and moderate interactions among the variables considered for the R1, R2, R3, R4 and R5 values due to the curved nature of 3D surfaces. Fig. 4a indicates the mutual interaction of R1 with A, B; R2 with A, B; R3 with A, B; R4 with A, B and R5 with A, B, respectively. The plots for the interactions between A and B are given in Fig. 5. As Fig. 5 demonstrates, increasing two independent variables above and below the center points increases the CO_2 (R1), CO (R2) and NOx (R3) values. In contrast to other responses, biocoal contribution (A) up to about 40 % and as the oxygen concentration (B) increases, the combustion efficiency (R4) values always increase, but after the 40 % biocoal contribution, the combustion efficiency (R4) values decrease (Fig. 4). Also, Fig. 5 shows that increasing oxygen concentration (B) above and below the center points increases T (R5), but on the contrary as the biocoal share (B) increases, T (R5) values decrease. Other studies in the literature reported a similar O_2 concentration and biocoal share effect [23,24,33].

3.5. *Determination of optimum parameters of the co-combustion of Soma lignite / biocoal fuel blends*

The optimum magnitudes of the most significant parameters to

Table 6
ANOVA results of independent variables for each equation term using Response Surface Quadratic Model.

	Sum of Squares	df	Mean Square	F-value	p-value	
CO_2						
Model	123.66	5	24.73	20.88	< 0.0001	significant
A-O_2 concentration	65.26	1	65.26	55.10	< 0.0001	
B-Biocoal share in the fuel mixture	47.69	1	47.69	40.26	< 0.0001	
AB	2.17	1	2.17	1.83	0.1988	
A^2	5.70	1	5.70	4.81	0.0471	
B^2	3.91	1	3.91	3.30	0.0923	
Lack of Fit	15.40	10	1.54			
CO						
Model	4.161E+05	5	83226.45	35.79	< 0.0001	significant
A	1.894E+05	1	1.894E+05	81.45	< 0.0001	
B	1727.90	1	1727.90	0.7431	0.4043	
AB	31587.31	1	31587.31	13.59	0.0027	
A^2	749.64	1	749.64	0.3224	0.5798	
B^2	1.603E+05	1	1.603E+05	68.96	< 0.0001	
Lack of Fit	30226.96	10	3022.70			
NOx						
Model	16285.15	5	3257.03	42.15	< 0.0001	significant
A	11174.62	1	11174.62	144.61	< 0.0001	
B	4388.27	1	4388.27	56.79	< 0.0001	
AB	13.72	1	13.72	0.1775	0.6804	
A^2	1.21	1	1.21	0.0156	0.9025	
B^2	12.66	1	12.66	0.1638	0.6923	
Lack of Fit	1004.60	10	100.46			
Efficiency						
Model	47.75	9	5.31	53.16	< 0.0001	significant
A	0.0198	1	0.0198	0.1979	0.6669	
B	4.81	1	4.81	48.15	< 0.0001	
AB	3.26	1	3.26	32.63	0.0003	
A^2	0.4319	1	0.4319	4.33	0.0672	
B^2	12.85	1	12.85	128.78	< 0.0001	
A^2B	0.4100	1	0.4100	4.11	0.0733	
AB^2	1.12	1	1.12	11.24	0.0085	
A^3	1.32	1	1.32	13.18	0.0055	
B^3	5.50	1	5.50	55.10	< 0.0001	
Lack of Fit	0.6533	6	0.1089	1.33	0.4391	not significant
Temperature						
Model	41394.46	5	8278.89	158.65	< 0.0001	significant
A	33574.00	1	33574.00	643.38	< 0.0001	
B	5857.63	1	5857.63	112.25	< 0.0001	
AB	35.78	1	35.78	0.6857	0.4226	
A^2	1143.00	1	1143.00	21.90	0.0004	
B^2	435.36	1	435.36	8.34	0.0127	
Lack of Fit	678.39	10	67.84			

Table 7
Statistical values for each fitted model of each response using response Surface Quadratic Model.

Responses	Std. Dev	Mean	C.V. %	R^2	Adjusted R^2	Predicted R^2	Adeq Precision
CO_2 (R1)	1.09	16.07	6.77	0.8893	0.8467	0.7590	15.8376
CO (R2)	8.22	296.59	16.26	0.9323	0.9062	0.7683	20.1478
NOx (R3)	8.79	184.44	4.77	0.9419	0.9195	0.8891	22.4395
E (R4)	0.3159	96.28	0.3281	0.9815	0.9631	0.8391	28.1640
T (R5)	7.22	810.99	0.8907	0.9839	0.9777	0.9620	41.1164

minimize the CO, NOx and the bed temperature values, and to maximize of CO_2 and combustion efficiency values for co-combustion of Soma lignite / biocoal fuel blend parameters [23]. Model equations were used for optimizations. Two independent variables were adjusted based on the data of five responses requested (CO_2, CO, NOx emissions, temperature and combustion efficiency). If the combustion efficiency is high, the co-combustion of Soma lignite / biocoal fuel blend process is more economical. The reduction in the CO and NOx reduces the emissions values of co-combustion process. Also, the increase in the CO_2 values increase the CO_2 capture efficiency and reduces capital and operational costs of co-combustion process [23,24]. Since the concentration of CO_2 in the flue gases increased, the CO_2 in the flue gases can be separated and captured by using CCS technology that is considered as the most energy and cost efficient technology. The optimization criteria specified in the Materials and Methods section were entered into the Design Expert 11 trial program. Optimization solutions proposed by the program were obtained. Optimization procedure carried out with desirability function. The utility performance technique is one of the most widely used engineering applications for parameter optimization [17]. According to this function, the O_2 concentration and biocoal share in the fuel mixture values of every determined response is transformed to a dimensionless desirability value (d). The function has a value between 0 and 1, the increase in the desirability is directly related to the value of d so that the corresponding response increases with it. In this study, for co-combustion of Soma lignite / biocoal fuel blend in oxygen-enriched atmosphere, optimum values of O_2 concentration (A) and biocoal share in the fuel mixture (B) to minimize CO, NOx and the bed temperature values and to maximize CO_2 and combustion efficiency values

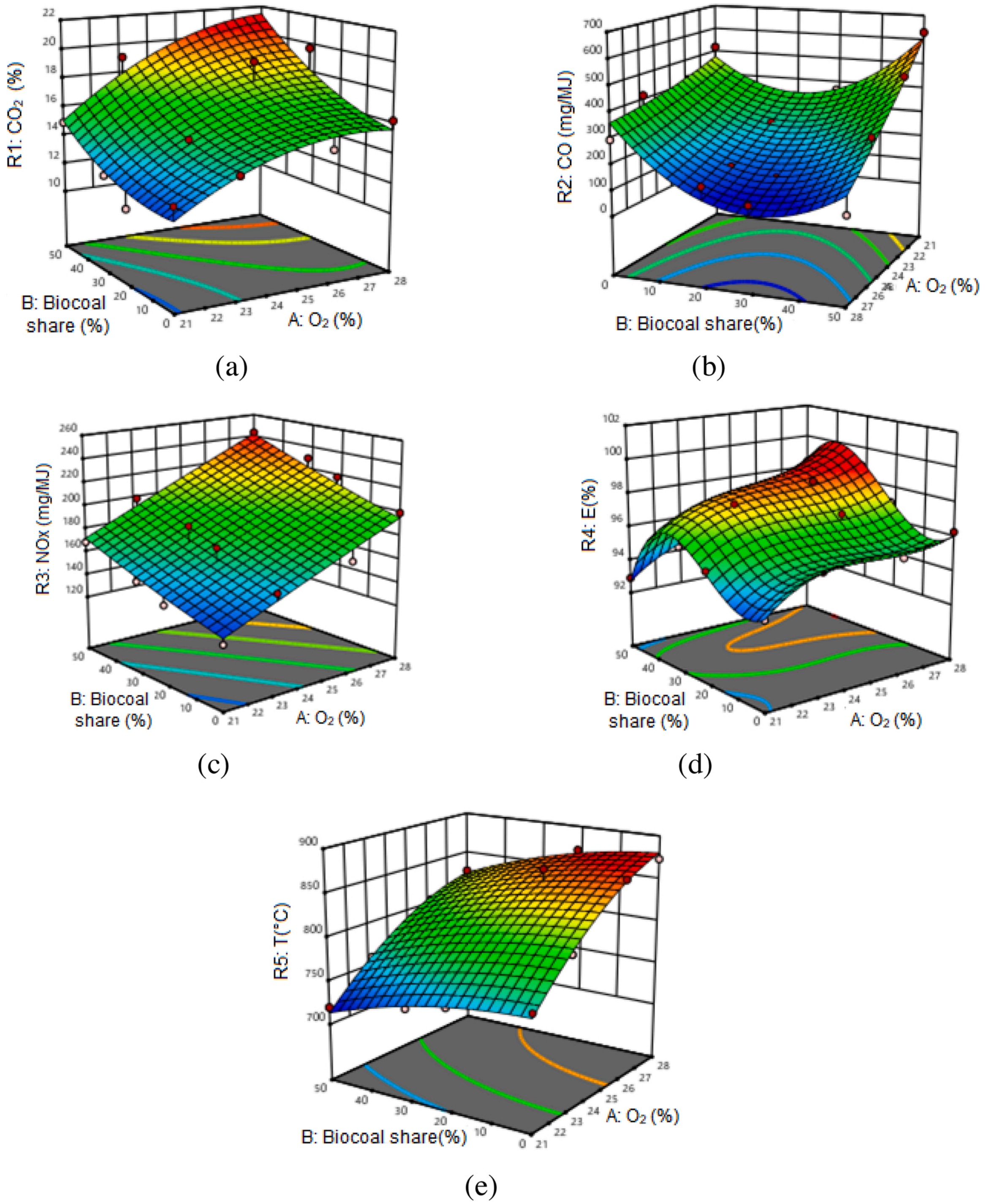


Fig. 5. 3D response surface plots showing simultaneous effect of O_2 concentration and the average bed temperature on (a) CO_2 emission, (b) CO emission, (c) NOx emission and (d) combustion efficiency, (e) Temperature.

were calculated to be 22.8 % and 37.2 % by vol., respectively, with desirability of 0.75 for biocoal share in the fuel mixture and 0.73 for O_2 concentration. The optimum values of 5 responses (CO_2, CO, NOx emissions, the bed temperature and combustion efficiency) were obtained as 16.09 % by vol., 321.41 mg/MJ, 179.39 mg/MJ, 778.48 °C and 97.81 %, respectively (Fig. 5).

Fig. 6 demonstrates the desirability of each dependent and independent parameter for the co-combustion of Soma lignite / biocoal fuel blend process. The first 2 bars show the independent parameters, i.e. O_2 concentration and biocoal share in the fuel mixture. The other bars show the dependent parameters, like CO_2, CO, NOx emissions, the bed temperature and the combustion efficiency. They are the responses obtained. The most effective parameter on co-combustion of Soma lignite / biocoal fuel blend is biocoal share in the fuel mixture. As shown in Fig. 6, desirability for responses can be arranged in decreasing order as E (0.82) > T (0.62) > CO (0.58) > NOx (0.55) > CO_2 (0.52).

4. Conclusions

The optimum values of oxygen concentration and the proportion of biocoal in the blend for co-combustion of red pine biocoal with Soma lignite fuel blends was determined using the mathematical and

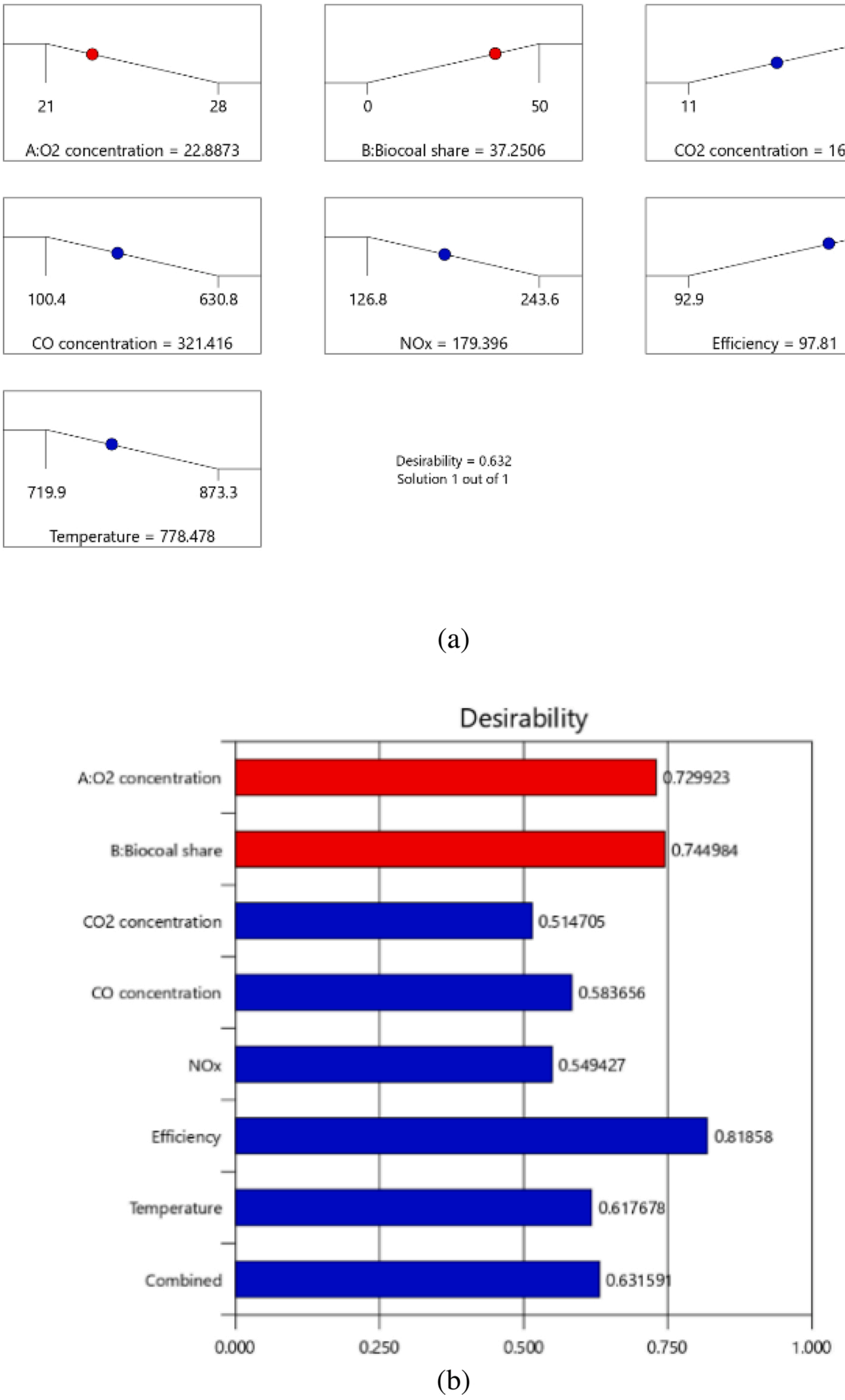


Fig. 6. Desirability bars for (a) optimum process condition with the highest desirability at optimum point, (b) optimization of the 5 selected goals.

statistical analysis and design of experimental technique based on Response Surface Methodology (RSM) by Central Composite Design (CCD) model. O_2 concentration and biocoal share in the fuel mixture were independent parameters, and CO_2, CO, NOx emissions, the bed temperature and the combustion efficiency were chosen as dependent variables (responses). ANOVA study statistically showed that models based on CO_2, CO, NOx emissions, the bed temperature and the combustion efficiency as a function of two parameters (oxygen concentration and the proportion of biocoal in the blend) were very significant, and the experimental results showed their importance. Differently from other studies, the results in this study underline the dominance of oxygen concentration and the proportion of biocoal in the blend for the whole responses obtained. By applying the RSM, it was found that the proportion of biocoal in the fuel blend was the most effective parameter among the five responses. Besides, the interactions of the two factors for all responses were successfully described by central composite design (CCD) model. The optimum values of oxygen concentration and the proportion of biocoal in the blend to minimize the CO, NOx and the bed temperature values, and to maximize the CO_2 and combustion efficiency values were selected as 22.8 % by vol. and 37.2 % by wt., respectively. On the other hand, CO_2 concentration in the flue gas increased when 50 % biocoal is added to lignite mixture which increases energy efficiency. Since the concentration of CO_2 in the flue gases increased, the CO_2 in the flue gases can be separated and captured by using CCS technology that is considered as the most energy and cost efficient technology. Also, there are possible energy savings because the amount of energy flowing out with the exit air decreased with increasing oxygen concentration due to the amount of nitrogen flowing out decreased [34]. As the oxygen concentration in the combustion air increased, more energy is available from the combustion thus less fuel is required.

Authorship statement

All persons who meet authorship criteria are listed as authors, and all authors certify that they have participated sufficiently in the work to take public responsibility for the content, including participation in the concept, ideas, literature review, design, analysis, writing, drafting or revision of the manuscript and take intellectual responsibility for its content. Furthermore, each author certifies that this material or similar material has not been and will not be submitted to or published in any other publication before its appearance in the *journal of CO2 utilization*.

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Data availability

The authors are unable or have chosen not to specify which data has been used.

Declaration of Competing Interest

The authors report no declarations of interest.

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