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INVESTIGATION OF THE INFLUENCE OF PRESSURE ON THE COMBUSTION OF ALPAGUT LIGNITE

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This research presents the combustion behavior of lignite under different reaction pressures. Lignite from Alpagut, Çorum of Turkey was combusted in its run off mine (ROM) condition under three different pressure levels of 172, 345, 517 kPa (25, 50, 75 psi). Experiments were done in a fully controlled temperature regime in an isolated combustion tube that operated in coordination with a continuous gas analyzer. Combustion behavior of lignite under different pressures was characterized by effluent gas analysis method. The changes in the amounts of consumed oxygen, evolved carbon oxides as well as variations in the temperature were assessed. The combustion efficiency and effectiveness of lignite was evaluated in terms of thermal features, from the viewpoint of reaction kinetics and by the computation of instantaneous fuel consumption at critical points. It was seen that combustion of lignite tended to turn from a steady profile to a considerably rapid one with increase in pressure, proving to be highly sensitive to the applied pressure level. Also, different levels of pressure resulted in distinctive combustion behavior not only from the view of thermal characteristics, but also in terms of reaction kinetics.

Keywords: *combustion, lignite, pressure, reaction kinetics, thermal features*

Introduction

In spite of the increasing trend for seeking alternative energy sources, fossil fuels remain to be the major source of energy. Coal is the most abundant fossil fuel, comprising approximately three quarters of the total recoverable fossil fuels in the world with an even distribution compared to other sources [1]. Coal is expected to play an increasingly important role in the 21st century in meeting the energy requirements [2, 3]. Despite the abundance and availability of the coal reserves it is a conflict that coal ranks as the second most commonly used energy source [2]. The most important reason underlying this fact is the environmental concerns. Air-pollutants and after-use particulate and washery tailings disposal are serious problems related to coal utilization and strict environmental regulations restrains use of coal and prospective projects [4, 5]. Development of environment friendly and cost effective combustion technologies is one of the most important issues for the utilization of coal. Efforts have focused on clean coal technologies during the last three decades. Among a number of alternative combustion facilities introduced, several were practically employed. Fluidized bed combustor (FBC), pressurized fluidized bed combustor (PFBC), integrated gasification combined cycle (IGCC), pulverized coal injection (PCI) and a variety of fixed bed reactors are the important ones characterized by their efficiencies and minimized environmental impacts [3, 4]. These new technologies, however, require many parameters be optimized and closely controlled during operation. Among these parameters, pressure probably accounts as the most important one. It was shown by many scientists that changes in pressure would lead to extensive deviations and/or modifications in the combustion profile [3, 6–12]. Sun and Zhang examined the ignition of coal particles at elevated pressure levels using a pressurized thermogravimetric analyzer [7]. Ignition temperature of coal particles was reported to decrease with an increase in pressure, claiming that higher pressure levels contributed to the initiation of oxidation. Wang *et al.* developed a rapid approach to examine the liability of coals to oxidation [8]. He monitored the low-temperature oxidation of coal particles under extended pressure and found that increasing the pressure had a dramatic effect on the effectiveness and the intensity of coal combustion. Liakos *et al.* studied the effect of pressure on the combustion of pulverized coal through a two-dimensional, two-phase combustion model [9]. It was reported that pressure had a significant effect on the reaction kinetics and the reaction mechanism. All these studies in literature show that pressure is of profound importance for coal combustion as it

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directly dictates the nature of reaction and the efficiency of operation, particularly in the currently used sophisticated combustion technologies.

The use of thermal analysis for solid fuels and particularly for coal is very reliable for investigating combustion behavior with regard to a number of variables. Two of the very recent studies utilizing thermal analysis for the characterization of coal combustion were carried out by Kizgut *et al.* and Elbeyli and Pişkin [13, 14]. Kizgut *et al.* used TG/DTG to investigate the thermal characteristics of a chemical-treated ultra-clean Turkish coal [13]. Elbeyli and Pişkin combined TG/DTG/DSC analysis and reaction kinetics concept to determine the combustion and pyrolysis behavior of cleaned Tuncbilek lignite [14]. The use of oxyreactive thermal analysis (OTA) has been studied by Cebulak *et al.* [20]. Cebulak *et al.* concluded that OTA is a new and a useful tool to study the relations between the physicochemical properties and thermal reactivity of carbon materials like anthracite, diamond and graphites. The effect of heating rate on different bituminous coal as well as electrographite and sodium silicate samples were studied by Bigda and Mienowksi through thermogravimetry and kinetic analysis [21]. All these studies and a vast number of other researches that were not cited here imply that thermal analysis is one of the essential ways of identifying reaction behavior and interpreting thermal decomposition characteristics of all fossil fuels and other materials, offering a great flexibility and a broad perspective of research, as has been mentioned by Stenseng *et al.* [22].

This research investigates the effect of air pressure on the combustion behavior of a Turkish lignite by evaluating the combustion profiles at three different pressure levels. The aim was to assess the variations in the efficiency, the intensity and the effectiveness of lignite combustion as the pressure was changed. The combustion behavior is expressed from the view of thermal features, reaction kinetics as well as amount of consumed fuel at certain instants of on-going reaction.

Experimental

Material and methods

Lignite sample

In this research, Alpagut lignite from Çorum-Turkey was used for the combustion experiments. Alpagut lignite is of moderate quality and has the general characteristics of most Turkish lignites. The ultimate and proximate analyses of Alpagut lignite are given in Table 1.

Table 1 Proximate and ultimate analysis of the Alpagut lignite sample (as received)

Proximate analysis		Ultimate analysis	
Moisture/%	22.01	C/%	42.92
Ash/%	15.60	H/%	2.94
Volatile matter/%	28.99	S/%	1.17
Fixed carbon/%	33.40	N/%	1.66
Cal. value/ kcal kg^{-1}	4300		

Combustion experiments and thermal characteristics

Combustion experiments with Alpagut lignite were conducted in a closed tube reactor that enables close control of the operational parameters. The reaction assembly used simulates the actual-scale with comparatively lower sample loading and proved to produce reliable results in several previous studies about the thermal decomposition of fossil fuels [13–21]. The combustion tube consists of a reactor, auxiliary heater parts, a temperature commander and a continuous gas analyzer (Fig. 1). The gas analyzer simultaneously processes the effluent gas quantitatively as the reaction continues. The lignite sample is put inside the reactor and ignited through heating by electrical coils around. For the combustion tests, the initial temperature was set at 20 and increased to 100°C at a heating rate of 5°C min^{-1} under nitrogen flow. The reactor was retained at this temperature for 30 min to achieve thermal equilibrium. Then, the heating rate was reduced to 2°C min^{-1} and purge air injection replaced nitrogen at 1.5 L min^{-1} rate. Combustion tests were performed under constant pressures of 172, 345 and 517 kPa (25, 50 and 75 psi).

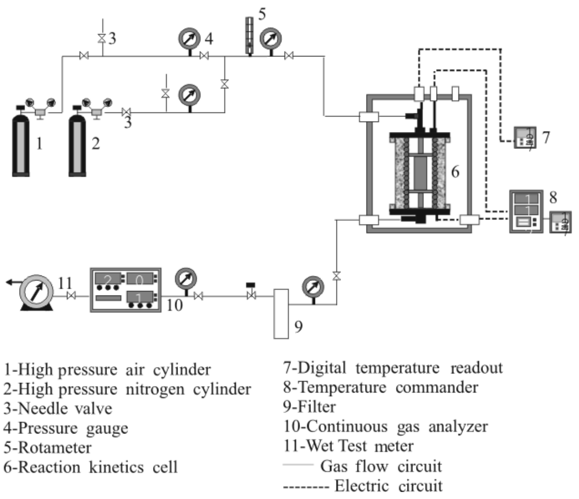

Fig. 1 Schematic diagram of the reaction kinetics system

Once the reaction is started the changes in the temperature of the reaction core, oxygen consumption and the composition of the effluent gas are monitored and recorded. O_2 peak temperatures and times that mark the maximum reaction level, the burnout temperatures and times and the residue amounts at the end of combustion are the most indicative thermal features. In this context, these parameters were assessed via effluent gas analysis method to determine the effectiveness of combustion as a function of the combustion pressure.

Reaction kinetics

To determine the reaction kinetics, a model that incorporates the principles of Arrhenius kinetics and effluent gas analysis data was used. The model was first developed by Weijdema for the interpretation of oxidation kinetics of in-situ combustion oil-recovery process [22]. The model relied on the measurement of rate of oxygen consumption as a function of time. The model was then improved by Fassihi *et al.* and made more concise and flexible [23]. The method proved to be a reliable tool for the computation of reaction kinetics with a variety of fossil fuels, such as oil, oil shales, tar sands, etc. [14, 19, 25]. According to the model,

$$\text{Rate of } O_2 \text{ consumption} = q\Delta O_2/(AL) \tag{1}$$

where q=constant gas flowrate (moles per unit time), ΔO_2=change in the concentration of O_2 concentration in the exit gas, A=cross sectional area of the sample pack, L=length of the sample pack.

Equation 1 is commonly expressed as:

$$q\Delta O_2/(AL) = A_r PO_2^m C_f^n \exp(-E/RT) \tag{2}$$

where A_r=Arrhenius constant, PO_2=partial pressure of oxygen, Pa; m=reaction order with respect to partial pressure of oxygen; C_f=Fuel concentration, s^{-1}; n=reaction order with respect to fuel concentration; E=activation energy, kJ mol^{-1}; R=universal gas constant, J mol K^{-1}; T=absolute temperature, K.

The rate of O_2 consumption may also be expressed in terms of the rate of decrease of fuel amount, which provides;

$$q\Delta O_2/(AL) = -\alpha dC_f/dt \tag{3}$$

In Eq. (3), α is the proportionality factor and is equal to the amount of oxygen that reacts with 1 g of fuel. It is possible to determine O_2 consumption at any instant by combining Eqs (2) and (3);

$$q\Delta O_2/(AL) = A_r P_{O_2}^m C_f^n \exp(-E/RT) = -\alpha dC_f/dt \tag{4}$$

Integration of Eq. (4) between $t=t_1$ and $t=\infty$ yields;

$$\alpha C_f(t) = \int_{t_1}^{\infty} [q\Delta O_2/(AL)] dt \tag{5}$$

where C_f=0 at t=∞. Again from Eq. (4)

$$C_f^n(t) = (q\Delta O_2/(AL)]1/A_r P_{O_2}^m C_f^n \exp(-E/RT) \tag{6}$$

and if Eq. (6) is substituted in Eq. (5);

$$\Delta O_2 / \left(\int_{t1}^{\infty} O_2 dt \right)^n = \beta \exp(-E/RT) \tag{7}$$

where

$$\beta = (q/AL)^{n-1} A_r P_{O_2}^m / \alpha^n \tag{8}$$

The temperature should be increased at a constant rate and the relative reaction rate of the combustion is computed via the changes in the amount of oxygen consumed. Oxygen consumption at different temperatures is obtained by graphical integration of the values on the left hand side of Eq. (7), i.e. by graphical integration of the $\Delta O_2=f(t)$ curve. Next, the logarithm of these values *vs.* $1/T$ is plotted to obtain '$-E/2.303R$' as the slope and $\log\beta$ as the intercept of the curve [22]. A computer program [18] is used to compute the relative reaction rate, processing the changes in reaction temperature, O_2 consumption and the quantities of the effluent gases. A plot of the relative reaction rates *vs.* inverse temperatures gives a straight line with its slope equal to the activation energy, E_A, of the reaction. The log–log plot of the true intercept of the straight line *vs.* O_2 partial pressure provides the Arrhenius constant, A_r, as the intercept of the curve [26].

Instantaneous fuel consumption

It is possible to characterize the liability of lignite to combustion at a given condition by the identification of thermal characteristics and by an assessment of the reaction kinetics and activation energy. However, both approaches take the reaction entirely and can not reflect the characteristic of the on-going reaction at a certain instant. As lignite combustion is a highly heterogeneous reaction in its nature, it would be necessary to identify the intensity of combustion at a desired instant to make comparisons among the distinctive phases driven by different mechanisms. In order to determine and compare the extent of combustion at critic instants and at various phases, a new approach has been introduced using the relative reaction rate concept. Relative reaction rate equation expresses the oxidation rate of any fuel as follows [22–29]:

$$R_c = A_r \exp\left(-\frac{E}{RT}\right) P_{O_2}^m C_f^n \tag{9}$$

where R_c=relative reaction rate; A_r=Arrhenius constant; E=activation energy, kJ mol^{-1}, R=universal gas constant; T=temperature, K; P_{O_2}=partial pressure of oxygen, Pa; C_f=instantaneous fuel consumption, g.

Using the instantaneous fuel consumption term (C_f), direct computation of the amount of combustible matter oxidized at any instant is possible once the activation energy and Arrhenius constants are provided.

Results and discussion

The profiles for the carbon oxides emissions and O_2 consumption *vs.* time and temperature at different pressures are given in Figs 2–4. Combustion of lignite showed a single peak of maximum O_2 consumption as well as CO_2 and CO evolutions, but occurring at different time and temperatures with respect to pressure level (Figs 2–4). This peak shows the maximum point of reaction. Combustion of lignite tends to accelerate until this peak, as revealed by the increases in O_2 consumption and carbon oxides evolution. Beyond this peak, combustion tends to decelerate on account of the depletion of the combustible matter.

O_2 consumption increased with increase in pressure from 172 to 517 kPa, as seen in Table 2 and Fig. 5. Accordingly, it may be suggested that increasing the pressure led to increases in the extent of combustion, which was probably due to enhanced diffusion of reactant gas through the pores involved by the combustible mass. The amount of combustible matter affected by O_2 became greater, as O_2 flow through the inner and deep sections was enforced by raises in the pressure. It should also be noted that with increase in pressure O_2 distribution throughout the entire lignite mass became more homogenous and a higher level of O_2 saturation could be achieved. Parallel to the increase in the effectiveness of combustion, evolution of CO and CO_2 also increased as the products of combustion (Figs 2–4). Higher O_2 consumption at higher pressures was in agreement with previous studies that reported an increase in the extent of combustion of hydrocarbon species with rises in pressure [8, 19, 20, 29].

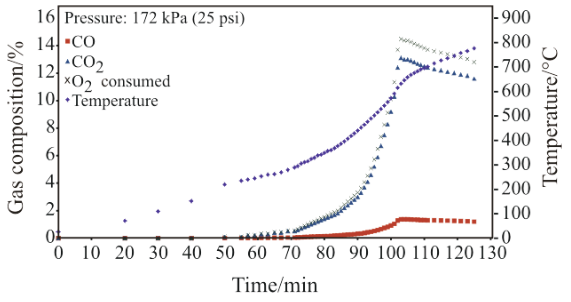

Fig. 2 Gas compositions and temperature *vs.* time of combustion at 172 kPa (25 psi)

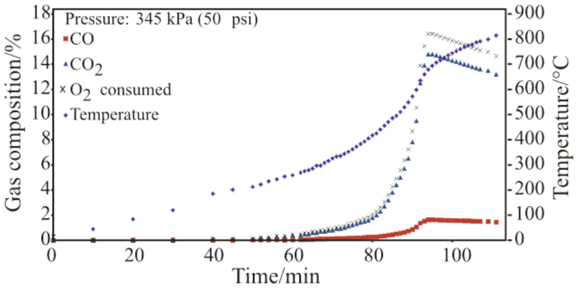

Fig. 3 Gas compositions and temperature *vs.* time of combustion at 345 kPa (50 psi)

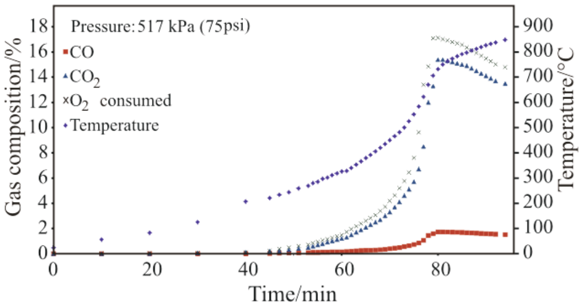

Fig. 4 Gas compositions and temperature *vs.* time of combustion at 517 kPa (75 psi)

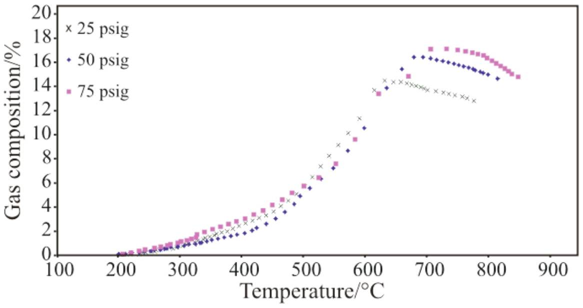

Fig. 5 O_2 consumption curves and peak temperatures at 172, 345 and 517 kPa

With increases in pressure, higher O_2 peak temperatures were also observed (Table 2, Fig. 5). It is well known that the heat release during the oxidation of a fuel is directly proportional to the extent of O_2 consumed [30]. Accordingly, the increase in the peak temperatures is attributed to the improved O_2-combustible matter contact at higher pressures that led to an increase in the amount of combustible matter oxidized and heat released per unit time. Similar findings were reported for the pressure-related combustion of oil shales [20, 29]. Moore also reported O_2 peak temperatures to increase due to accelerated O_2 diffusion with rises in pressure [31]. In addition, earlier peak temperatures were observed as the pressure was increased to 345 and 517 kPa (50 and 75 psi) which reflected an increase in the relative rate of reaction due to increased effectiveness of O_2 diffusion. The amount of residue at the

Table 2 O_2 peak temperatures and times, O_2 consumption and peak values according to different pressure levels

Pressure level/		O_2 peak temp./°C	Peak time/min	O_2 consumption/%	Residue/%
kPa	psi				
172	25	632	103	14.48	25.21
345	50	679	94	16.43	22.07
517	75	732	80	17.13	19.62

end of combustion decreased as the pressure was increased and the lowest residue was achieved at 517 kPa (Table 2). This observation confirms that the combustible matter that remained unreacted at 172 kPa, was oxidized as a result of improved O_2 diffusion at higher pressures and a more complete combustion could be achieved.

From the view of reaction kinetics it was seen that increases in combustion pressure resulted in decreases in the activation energies (Table 3). This suggests that the reactivity of lignite is improved as the pressure was increased and the liability of lignite particles to combustion increased. The improvement in the reactivity was governed by two main factors [19, 32]. A more homogenous distribution and a more effective in-situ flow of O_2 leading to a more uniform combustion regime; and the rapid replacement of the consumed O_2 with fresh O_2, being strongly enforced at higher pressures.

To evaluate the role of pressure on combustion of Alpagut lignite with respect to the instantaneous fuel consumption approach, first, the certain instants that would be indicative have been sorted. When the combustion profiles at 172, 345 and 517 kPa were examined, it was seen that the differences due to pressure became apparent after the 50th min of reaction (Figs 2–4) and this made 50th min as the first critical instant. In order to evaluate the combustion in a further phase of reaction, 65th min of reaction was marked. In addition, the amount of fuel consumption at the O_2 peak temperature would be meaningful as the combustion reaches its highest level at this point. The earliest peak was recorded at 517 kPa at the 80th min of reaction. This instant was also encountered to enable a qualitative comparison of the effectiveness of combustion. The instantaneous fuel consumption values for the above mentioned instants and the corresponding temperatures are given in Table 3. It is seen that the fuel consumption increased at the 50th, 65th and 80th min as the pressure was increased. The temperature levels achieved at these instants also increased, confirming the increased amount of fuel oxidation and heat release with rise in pressure. The highest peak temperature at the earliest peak time at 517 kPa should also be noted as it points out the contributing effect of pressure on the extent of combustion. This means that the pressure level influences not only the effectiveness of the reaction, but the relative rate of the reaction as well, as revealed by the thermal indicators, activation energies and the amounts of fuel consumed instantaneously.

Table 3 Activation energy values and instantaneous fuel consumption values computed at critical instants according to different pressure levels

Pressure/		Time/min	Temp./°C	Ins. fuel cons./ $g\cdot10^{-4}$	Act. energy/ kJ mol^{-1}
kPa	psi				
172	25	50	220	0.206	46.10
		65	262	0.83	
		80	349	3	
		103 (peak)	632	38.37	
345	50	50	212	0.25	43.52
		65	286	1.44	
		80	418	4.53	
		94 (peak)	679	48.05	
517	75	50	239	0.95	38.79
		65	327	3.4	
		80 (peak)	732	57.75	

Conclusions

In view of the findings achieved in this study, the following conclusions were derived.

- The extent of combustion increased with the increases in the pressure level. This was due to more adequate O_2 diffusion, which led to a more effective interaction among the reactant gas and the combustible matter.
- Increasing the pressure resulted in a more complete combustion and higher combustion efficiency, as indicated by the higher temperature levels achieved and lesser residue left. This was also expressed by the increases in the quantities of fuel burned at the specific instants as the pressure was increased.
- Increasing the pressure favored the reaction kinetics and increased the reactivity of lignite, significantly improving the liability of the combustible matter to ignition. This was governed by the abundance of O_2 even at the most immersed sections as a result of high pressure.
- Combustion behavior of lignite was highly pressure-sensitive, turning from a steady reaction to a more rapid and a vigorous one as the pressure was increased.
- It may be misleading to suggest pressure increases to be in favor of combustion or low pressures as a foe. However, it was clearly understood that pressure is a significant operational parameter which provides a great flexibility between a steady going combustion and a rapidly flowing one. Thus, it can be adjusted in accordance with the object of utilization or the design of the combustor facility, enabling direct control on the reaction behavior of lignite.

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