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Combustion behaviour of Turkish lignite in O_2/N_2 and O_2/CO_2 mixtures by using TGA–FTIR

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ABSTRACT

The pyrolysis and combustion behaviour of a low calorific value Turkish lignite with high sulphur and ash content in air and oxy-fuel conditions were investigated by using non-isothermal thermo-gravimetric method (TGA) coupled with Fourier-transform infrared (FTIR) spectrometer. Pyrolysis tests were carried out in nitrogen and carbon dioxide environments which are the main diluting gases of air and oxy-fuel environment, respectively. Pyrolysis results show that weight loss profiles are almost the same up to a temperature of 720 °C in these two environments, indicating that CO_2 behaves as an inert gas in this temperature range. However, further weight loss takes place in CO_2 atmosphere at higher temperatures due to CO_2–char gasification reaction. Combustion experiments were carried out in four different atmospheres: air, oxygen-enriched air environment (30% O_2–70% N_2), oxy-fuel environment (21% O_2–79% CO_2) and oxygen-enriched oxy-fuel environment (30% O_2–70% CO_2). Combustion experiments reveal that replacing nitrogen in the gas mixture by the same concentration of CO_2 does not affect the combustion process significantly but only leads to slight delay in combustion. Overall comparison of derivative thermogravimetry (DTG) profiles shows that oxygen content in the combustion environment is the most effective parameter irrespective of the diluting gas. As O_2 concentration increases profiles shift through lower temperature zone, peak and burnout temperatures decrease, weight loss rate increases and complete combustion is achieved at lower temperatures and shorter times. During pyrolysis and combustion tests gaseous products CO_2, CO, H_2O, CH_4, SO_2 and COS in flue gas were identified and analyzed by using FTIR. Results indicate that higher CO and COS formation takes place during pyrolysis due to gasification reaction. Gaseous species evolution trends in combustion tests are found to be almost identical in oxygen enriched conditions independent of the diluting gas.

1. Introduction

Today, demand for electric power continues to increase due to population growth, technological and economical development. With 826 billion tones of proved coal reserves, coal combustion has an important role in energy production worldwide [1]. Growing concern about greenhouse gas emissions and their potential impact on climate change necessitate investigation of alternative technologies for reduction of CO_2 emissions from coal fired power plants. Conventional technologies for removing CO_2 from the stack gas in the existing coal fired power plants are expensive since CO_2 is diluted (typically about 14% by volume on a dry basis) [2]. The cost of gas separation can be reduced by increasing the concentration of CO_2 in the flue gas. Oxy-fuel combustion technology is suggested as one of the new promising technologies for capturing CO_2 from power plants. This technology is based on burning coal in a mixture of oxygen and recycled flue gas (RFG) leading to CO_2 concentrations greater than 95% in the exhaust gas. Recycled flue gas is used to control flame temperature and supply the volume of missing N_2. Oxy-fuel combustion technology for coal-fired power generation has been briefly described and reviewed in detail recently [3–5]. Oxy-fuel combustion is found to differ from air combustion in combustion characteristics such as burning stability, char burnout, gas temperature profiles and heat transfer due to differences in gas properties between CO_2 and N_2, the main diluting gases in oxy-fuel and air, respectively. Previous studies on oxy-fuel combustion mainly revealed that similar temperature profiles with air case are achieved at higher oxygen concentrations, around 30% in the combustion environment, as the higher heat capacity of CO_2 causes delay in combustion process [2–7].

Non-isothermal thermo-gravimetric analysis (TGA) technique is a rapid, inexpensive and simple method that has been widely used in studying the pyrolysis and combustion behaviour of coal and evaluating the relative burning properties of coal samples.

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There exists a considerable number of studies carried out for the investigation of combustion behaviour high rank coals in oxy-fuel environment by TGA [8–13]. However, an investigation of oxy-fuel combustion characteristics of indigenous lignites by TGA–FTIR technique is not available to date. Therefore, pyrolysis and combustion behaviours of an indigenous lignite with low calorific value, high ash and sulphur contents are studied in air and oxy-fuel conditions by using a TGA–FTIR combined system.

2. Experimental

2.1. Sample

A typical low quality indigenous lignite from Çan town of Çanakkale province in Turkey was used in pyrolysis and combustion tests. Proximate analysis of the Çan lignite was performed by using LECO TGA-701. Ultimate analysis was carried out with LECO CHNS-932. Calorific values of the fuels are measured by using AC-500 bomb calorimeter. Analyses were performed according to ASTM standards. The Jeol JSM-6400 scanning electron microscope (SEM) configured with a Noran energy dispersive spectrometer (EDS) was utilised for determination of ash composition. Proximate, ultimate and ash analyses of Çan lignite together with its calorific value are briefly summarized in Table 1. As can be seen from the table, Çan lignite can be characterized by its low calorific value, high ash content (~25%) and high total sulphur content (~4%).

2.2. Experimental setup and method

In the present work TGA/DTG were used to determine pyrolysis and combustion characteristics of lignite samples. TGA system was coupled with FTIR spectrometer for determination of evolved gases during pyrolysis and combustion experiments. Fig. 1 shows a schematic diagram of the experimental setup consisting of Perkin Elmer Pyris STA 6000 thermo-gravimetric analyzer, Spectrum 1 FTIR spectrometer and a mass flow controller (MFC) for each gaseous species. TGA and FTIR were connected by a heated line with a temperature of 270 °C in order to prevent the condensation of gases. FTIR spectra were collected with 4 cm^{-1} resolution, in the range of 4000–700 cm^{-1} IR absorption band.

About 12 mg of coal sample with particle size less then 100 μm was held initially at room temperature for 1 min and then heated with a heating rate of 40 °C/min from room temperature up to 950 °C during each experiment. In pyrolysis tests, samples were held at 950 °C for an additional 60 min. The required combustion environments were formed by mixing two gases in the desired ratio by using two different mass flow controllers in order to regulate the flow rates of the gases. The total gas flow was set to 70 ml/min for pyrolysis and 45 ml/min for combustion experiments.

Table 1
Proximate, ultimate and ash analysis of Çan lignite.

Ultimate analysis (as received, % by wt.)		Proximate analysis (as received, % by wt.)	
C	37.31	Moisture	16.35
H	3.3	Ash	28.78
O	10.02	Volatile matter	29.79
N	0.91	Fixed carbon	25.08
$S_{Combustible}$	3.33		
Ash	28.78	LHV (MJ/kg)	9.89
Total moisture	16.35		
S_{Total}	3.49		

Ash analysis (% by wt.)

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	SO_3	TiO_2
43.13	18.2	15.78	7.63	0.48	2.0	0.63	11.08	1.07

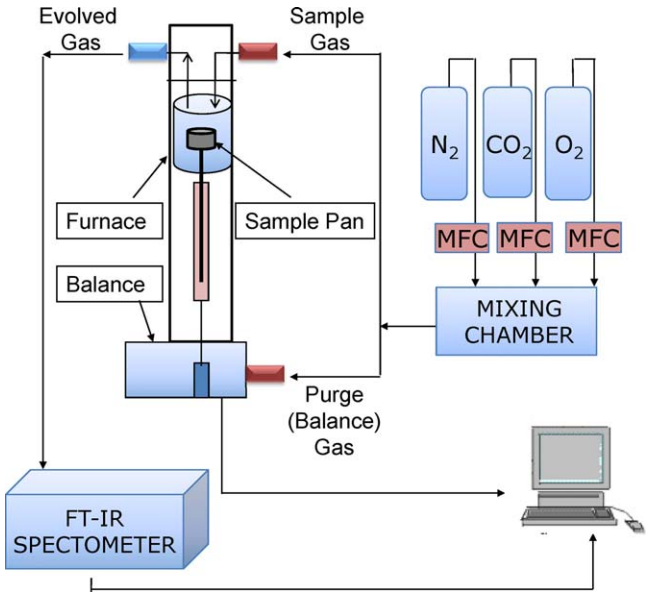

Fig. 1. Schematic diagram of the experimental setup.

Pyrolysis tests were carried out under nitrogen and carbon dioxide atmospheres which are the diluting gases of air and oxy-fuel environments, respectively. Four combustion tests were performed in air environment to investigate the effect of combustion environment on burning process of lignite sample. The base case was considered as lignite combustion in air environment. In oxygen-enriched air case the sample is burned in 30% O_2–70% N_2 atmosphere. In oxy-fuel combustion tests, the volume of N_2 used in the base case was replaced with an equal volume of CO_2. In the last case, combustion of lignite sample was investigated in oxygen-enriched oxy-fuel environment, that is, in 30% O_2–70% CO_2 atmosphere.

TGA and DTG profiles obtained during pyrolysis and combustion experiments were used to determine some characteristic parameters such as initial decomposition temperature (T_{in}), peak temperature (T_{max}), ignition temperature (T_{ig}) and burnout temperature (T_b). T_{in} represents the initiation of weight loss and is defined as the temperature at which the rate of weight loss reaches 1%/min after initial moisture loss peak in DTG profile [22]. T_{max} is the point at which maximum reaction rate occurs. Different from initial decomposition temperature, ignition temperature T_{ig} is defined as the temperature at which coal starts burning. It is taken as the temperature at which the weight loss curves in the oxidation and pyrolysis experiments diverge [20]. The last characteristic temperature considered is burnout temperature which represents the temperature where sample oxidation is completed. It is taken as the point immediately before reaction ceases when the rate of weight loss is 1%/min [21].

A linear relation between spectral absorbance at a given wavenumber and concentration of gaseous components is given by Beer's Law. In this study, the points of absorbance at a certain wavenumber are plotted against temperature in order to obtain a formation profile for each evolved gas observed in the spectra during experiments. The IR wavenumbers of CO_2, CO, H_2O, CH_4, SO_2 and COS are 2360, 2112, 1540, 3016, 1340 and 2042 cm^{-1}, respectively. Formation profiles of NO_x related species such as NO and NO_2 are not reported due to overlap of their absorption bands with the characteristic absorption bands of water in the range of 3900–3500 and 1900–1350 cm^{-1}.