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Spontaneous combustion in six types of coal by using the simultaneous thermal analysis-Fourier transform infrared spectroscopy technique

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Abstract Using simultaneous thermal analysis-Fourier transform infrared spectroscopy, we analyzed the oxidation and exothermic behaviors of six types of coal based on various factors, such as characteristic temperature, heat release, and gas release, to establish a foundation for prevention and control of spontaneous combustion in six types of coal in China. According to the experimental results, a decrease in the metamorphic grade of coal causes an increase in the amount of volatile matter, the heat release rate, and the total heat released. The apparent exothermic onset temperature and initial temperature for the release of H_2O, CO_2, CO, and CH_4 during the nonisothermal oxidation process of coal took place earlier, indicating that the oxidation reaction occurred more easily in lower-grade coal, increasing the hazards of spontaneous combustion. Moreover, when decomposing, coal releases large amounts of CH_4, which may cause gas explosions in coal mines. Therefore, technology facilitating the detection of CH_4 and prevention of explosions should be developed for use in the coal industry.

Keywords Apparent exothermic onset temperature · Characteristic temperature · Nonisothermal oxidation process · Oxidation and exothermic behaviors · Simultaneous thermal analysis-Fourier transform infrared spectroscopy technique

List of symbols

A_{ad}	Ash content (%)
FC_{ad}	Fixed carbon content (%)
M_{ad}	Moisture content (%)
T_1	Characteristic temperature of maximum evaporation of water and desorption of gases (°C)
T_2	Characteristic temperature of maximum oxidization mass gain (°C)
T_3	Characteristic temperature of ignition point (°C)
T_4	Characteristic temperature of maximum mass loss rate (°C)
T_5	Characteristic temperature of terminal residual (°C)
T_{D1}	Temperature at the maximum endothermic reaction with evaporation of water and desorption of gases (°C)
T_{D2}	Exothermic onset temperature of oxidation reaction (°C)
T_{D3}	Temperature at the maximum heat release rate (°C)
T_{D4}	Temperature at the end of the combustion reaction (°C)
V_{ad}	Volatile contents (%)

Introduction

In China, spontaneous combustion of coal has caused numerous accidents, resulting in economic loss and human casualties. On June 23, 2014, the spontaneous combustion of coal triggered a gas explosion in a mining area that killed 22 people in Chongqing, China [1]. In the same year (November 26, 2014), 24 people perished in Liaoning

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Province [2]. Undoubtedly, feasible control of spontaneous combustion of coal is still a crucial problem that has yet to be solved [3–8].

Spontaneous combustion of coal is a self-accelerating, nonlinear, and physical and chemical reaction process. In general, coal–oxygen compound theory has been applied broadly and developed completely for studying the coal spontaneous combustion [3]. The theory holds that when coal comes into contact with oxygen, it can generate an exothermic reaction because of physical adsorption, causing the coal's surface temperature to increase. During this process, certain functional groups that are easily oxidized on the coal's surface may undergo an oxidation reaction and release heat, resulting in the accumulation of thermal energy. If ventilation in the coal mine (i.e., heat removal) is less than the heat generated, the temperature rise may reach the ignition point of coal [9–13]. A spontaneous combustion of coal reaction may occur, resulting in fire, explosion, or the release of toxic gases [14–18].

In this study, six types of coal with different metamorphic grades, anthracite, lean coal, coking coal, gas coal, nonstick coal, and lignite, were tested to determine the characteristics of oxidation reaction, mass loss, and heat release, as well as which functional groups of gases were released by simultaneous thermal analysis-Fourier transform infrared spectroscopy (STA-FTIR). Therefore, the characteristics of spontaneous combustion of coal could be effectively identified and used for developing preventative technology, to help mitigate the risk of disaster and unpredictable environmental pollution [19, 20].

Experimental

Samples

The six types of coal used and listed from high to low metamorphic grades are anthracite (Yah-Ma-Zhuang, Henan, China), lean coal (Sang-Shu-Ping, Shaanxi, China), coking coal (Shi-Hao-Cun, Henan, China), gas coal (Nantun, Shandong, China), nonstick coal (Shi-Cao-Cun, Ningxia, China), and lignite (Jungar Banner, Inner Mongolia). All samples were pretreated. First, the oxidation layer on the coal surface was peeled to retain the central part. Second, the retained coal was broken under N_2 to prevent oxidation. Finally, the broken coal was collected and dried at low temperature to yield the experimental sample. Each type of coal underwent industrial and element analysis prior to the experiments, and the results are presented in Table 1. According to the industrial analysis results, an increase in the metamorphic grade corresponded with a decrease in the fixed carbon content (FC_{ad}) but an increase in the volatile contents (V_{ad}). Coking coal had the largest ash content (A_{ad}), and the largest moisture content (M_{ad}) appeared in lignite. From the element analysis results, a decrease in the metamorphic grade corresponded with lower carbon content and higher hydrogen and oxygen content. These results correspond to the basic properties of coal with differing metamorphic grades.

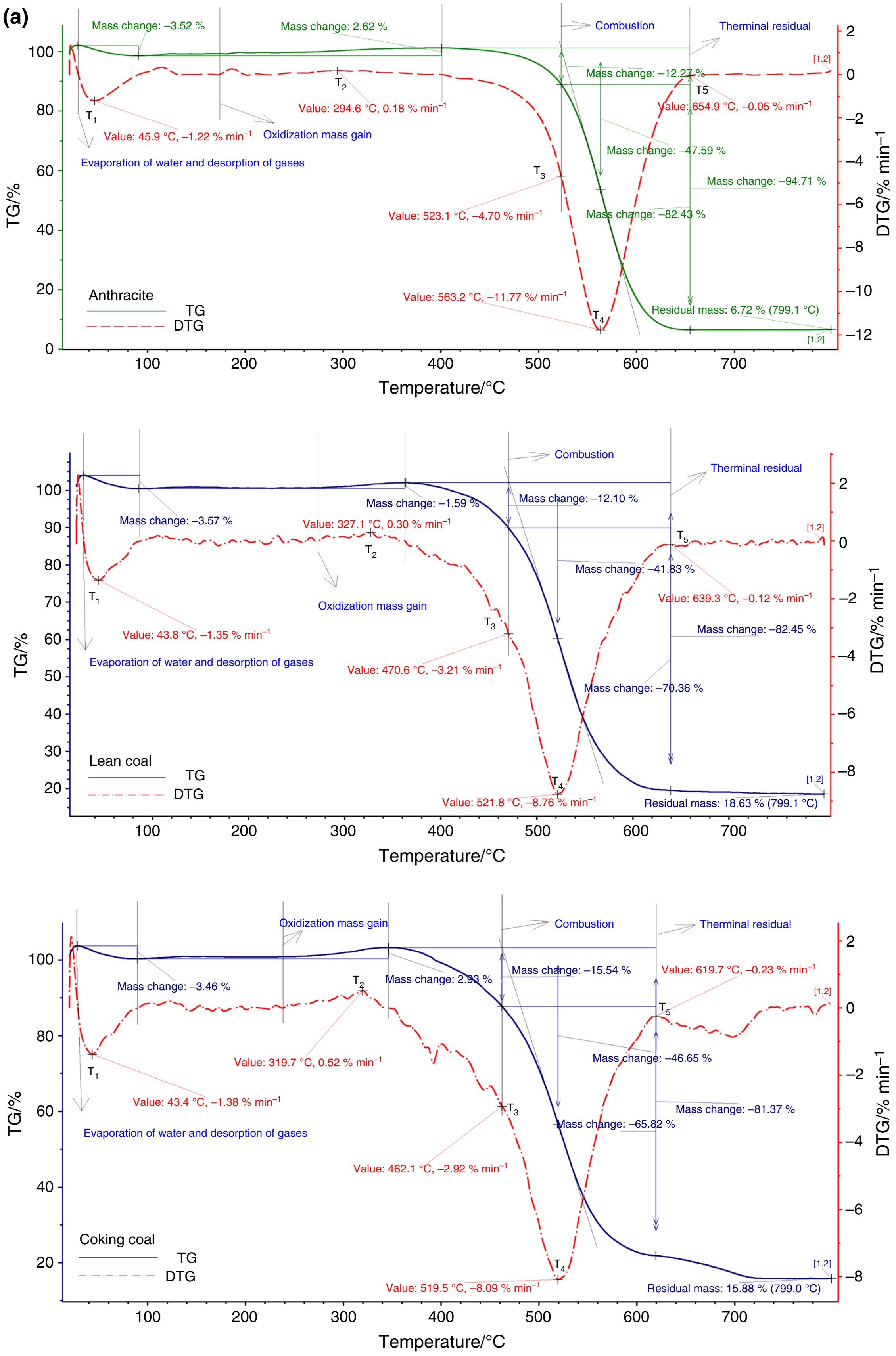
Fig. 1 Oxidation characteristic analysis for six types of coal by TG ▶ tests **a** (1/2), **b** (2/2)

Simultaneous thermal analysis-Fourier transform infrared spectroscopy

The characteristic temperature of different oxidation stages for six types of coal was investigated by using STA-449-F3 (NETZSCH, Bavaria, Germany), which consists of thermogravimetry (TG) and differential scanning calorimetry (DSC) techniques. Furthermore, the change in the functional groups of released gases with increasing temperature was observed through VERTEX70v FTIR (Bruker Corporation, MA, USA). The related experimental data, including the mass loss, mass loss rate, exothermic onset temperature of oxidation, and heat of oxidation reaction, as well as which functional groups of gases were released

Table 1 Results of industrial and element analysis for six types of coal

Samples	Industrial analysis/mass%					Element analysis/mass%			
	M_{ad}	A_{ad}	V_{ad}	FC_{ad}	C	H	O	N	S
Anthracite	3.10	7.27	5.54	84.11	90.04	3.61	2.79	0.38	0.19
Lean coal	0.61	10.78	13.55	75.06	84.53	3.73	4.27	0.14	3.37
Coking coal	0.52	22.28	28.34	63.29	78.23	3.85	6.42	0.14	0.63
Gas coal	3.45	8.83	32.86	55.31	74.42	5.05	7.31	0.44	0.29
Nonstick coal	10.82	6.23	33.01	49.94	67.37	4.66	11.56	0.24	0.43
Lignite	11.09	18.95	33.25	36.71	59.09	6.10	17.02	0.35	0.59


(a)
Mass change: –3.52 %
Mass change: 2.62 %
Combustion
Thermal residual
Mass change: –12.27 %
Value: 294.6 °C, 0.18 % min–1
Value: 654.9 °C, –0.05 % min–1
Value: 45.9 °C, –1.22 % min–1
Oxidization mass gain
Evaporation of water and desorption of gases
Mass change: –47.59 %
Mass change: –94.71 %
Value: 523.1 °C, –4.70 % min–1
Mass change: –82.43 %
Value: 563.2 °C, –11.77 %/ min–1
Residual mass: 6.72 % (799.1 °C)
Anthracite
TG
DTG
T1
T2
T3
T4
T5
[1.2]
TG/%
DTG/% min–1
Temperature/°C
Mass change: –1.59 %
Mass change: –12.10 %
Mass change: –3.57 %
Value: 327.1 °C, 0.30 % min–1
Mass change: –41.83 %
Value: 639.3 °C, –0.12 % min–1
Value: 43.8 °C, –1.35 % min–1
Mass change: –82.45 %
Value: 470.6 °C, –3.21 % min–1
Mass change: –70.36 %
Lean coal
Value: 521.8 °C, –8.76 % min–1
Residual mass: 18.63 % (799.1 °C)
Mass change: –15.54 %
Value: 619.7 °C, –0.23 % min–1
Mass change: –3.46 %
Mass change: 2.93 %
Value: 319.7 °C, 0.52 % min–1
Mass change: –46.65 %
Value: 43.4 °C, –1.38 % min–1
Mass change: –81.37 %
Mass change: –65.82 %
Value: 462.1 °C, –2.92 % min–1
Coking coal
Value: 519.5 °C, –8.09 % min–1
Residual mass: 15.88 % (799.0 °C)

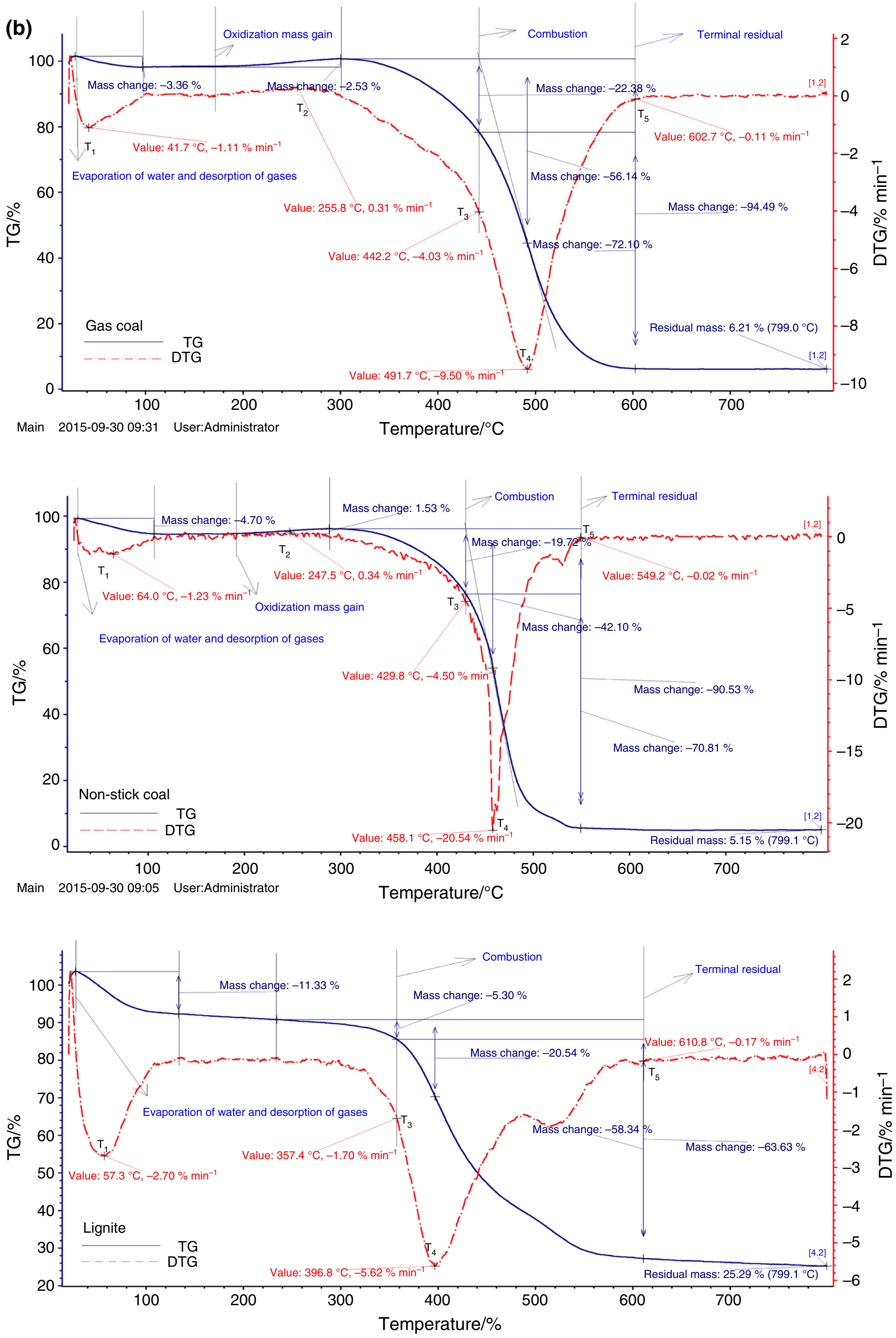


Fig. 1 continued

Table 2 Characteristic temperature of six types of coal by TG tests

Sample	Anthracite	Lean coal	Coking coal	Gas coal	Nonstick coal	Lignite
T_1/°C	45.904	43.775	44.415	41.743	64.032	57.319
Mass$_1$/%	98.642	98.647	98.647	98.924	97.308	95.248
DTG$_1$/% min^{-1}	−1.217	−1.352	−1.511	−1.108	−1.226	−2.699
T_2/°C	400.996	362.761	334.865	300.218	288.302	234.219
Mass$_2$/%	99.118	98.097	101.055	99.175	96.802	87.661
DTG$_2$/% min^{-1}	0.000	0.000	0.001	0.000	0.000	−0.087
T_3/°C	523.088	470.615	446.115	442.194	429.762	357.419
Mass$_3$/%	87.108	86.426	87.165	77.144	76.968	82.544
DTG$_3$/% min^{-1}	−4.694	−3.208	−3.113	−4.026	−4.500	−1.696
T_4/°C	563.163	521.752	519.480	491.711	458.066	396.819
Mass$_4$/%	52.576	57.925	56.632	43.900	54.483	67.834
DTG$_4$/% min^{-1}	−11.771	−8.756	−8.092	−9.504	−20.538	−5.619
T_5/°C	654.891	639.291	617.265	602.725	549.189	610.819
Mass$_5$/ %	6.400	18.831	22.650	6.154	5.667	26.262
DTG$_5$/% min^{-1}	−0.049	−0.119	−0.101	−0.106	−0.024	−0.175

with heating, could be started, measured, and recorded simultaneously by using STA-FTIR. The size of the sample and reference crucible used in STA was 10.0 mL. The temperature ranges of TG, FTIR, and DSC were set from 20.0 to 800.0 °C with a heating rate of 10.0 °C min^{-1}. Based on the structure of coal, the scanning range of infrared spectroscopy was from 4000 to 650 cm^{-1}, and the resolution was 4 cm^{-1}. The scanning times for each test were 32.0. The experiment was conducted in a 1:4 of oxygen/nitrogen environment with a flow rate of 50.0 mL min^{-1}.

Element analysis and industrial analysis

Element analysis of samples was investigated by using elemental analyzer EA 2400 II (PerkinElmer, USA). The mass of the sample was 50.0 mg. The furnace temperature was from 100.0 to 1100.0 °C. The related experimental data included C, H, O, N, and S contents.

Industrial analysis of samples was by industrial analyzer WBGF-6000 (Wanbo Instrument Co., Ltd., PR China). The mass of the sample was 800.0 mg. Oxygen pressure and nitrogen pressure were set at 0.2 MPa. The temperature was set from 20.0 to 800.0 °C. The experimental data included moisture content (M_{ad}), ash content (A_{ad}), volatile contents (V_{ad}), and fixed carbon content (FC_{ad}).

Results and discussion

TG results

TG was used to observe the mass loss of samples to determine properties of composition, thermal stability, and oxidation or degradation behaviors. Figure 1 shows the TG and differential thermogravimetry (DTG) results for six types of coal, and the characteristic temperature of oxidation reaction for coal could be classified by five features: characteristic temperature of maximum evaporation of water and desorption of gases, characteristic temperature of maximum oxidization mass gain, characteristic temperature of ignition point, characteristic temperature of maximum mass loss rate, and characteristic temperature of terminal residual, identified as T_1 to T_5, respectively. According to the results, the T_2 values of six types of coal—from high to low metamorphic grades—were 401.0, 363.0, 335.0, 300.0, 288.0, and 234.0 °C, indicating a declining trend. Because the metamorphic grade was lower, there was more volatile content in the coal, causing the oxidation rate to increase, so that the T_2 took place earlier. For the same reasons, T_3–T_5 for six types of coal also showed a similar trend. The T_3 values were 523.0, 470.0, 446.0, 442.0, 429.0, and 357.0 °C after the metamorphic grade was decreased. The T_4 values were 563.0,

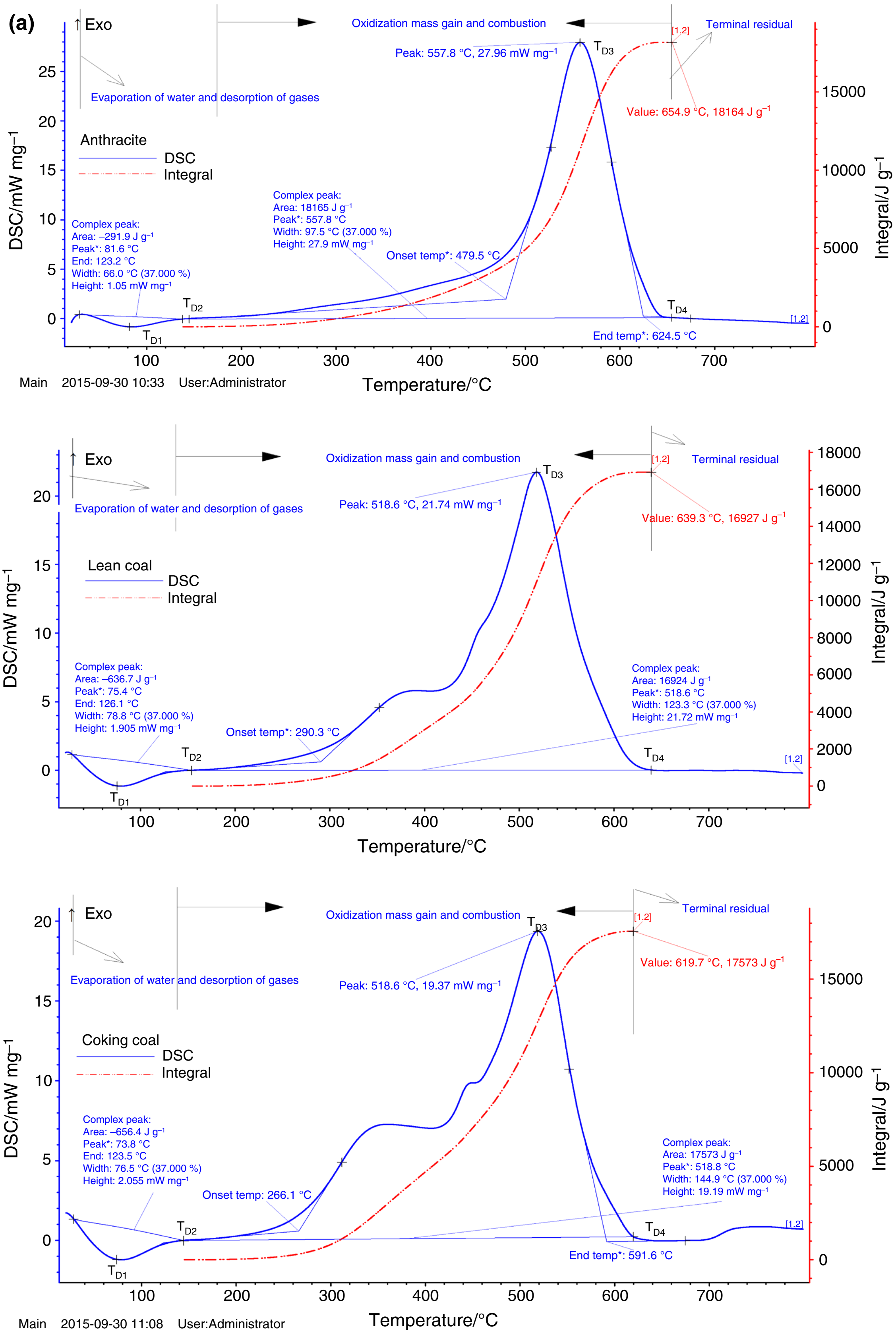


Fig. 2 Oxidation characteristic analysis for six types of coal by DSC tests **a** (1/2), **b** (2/2)

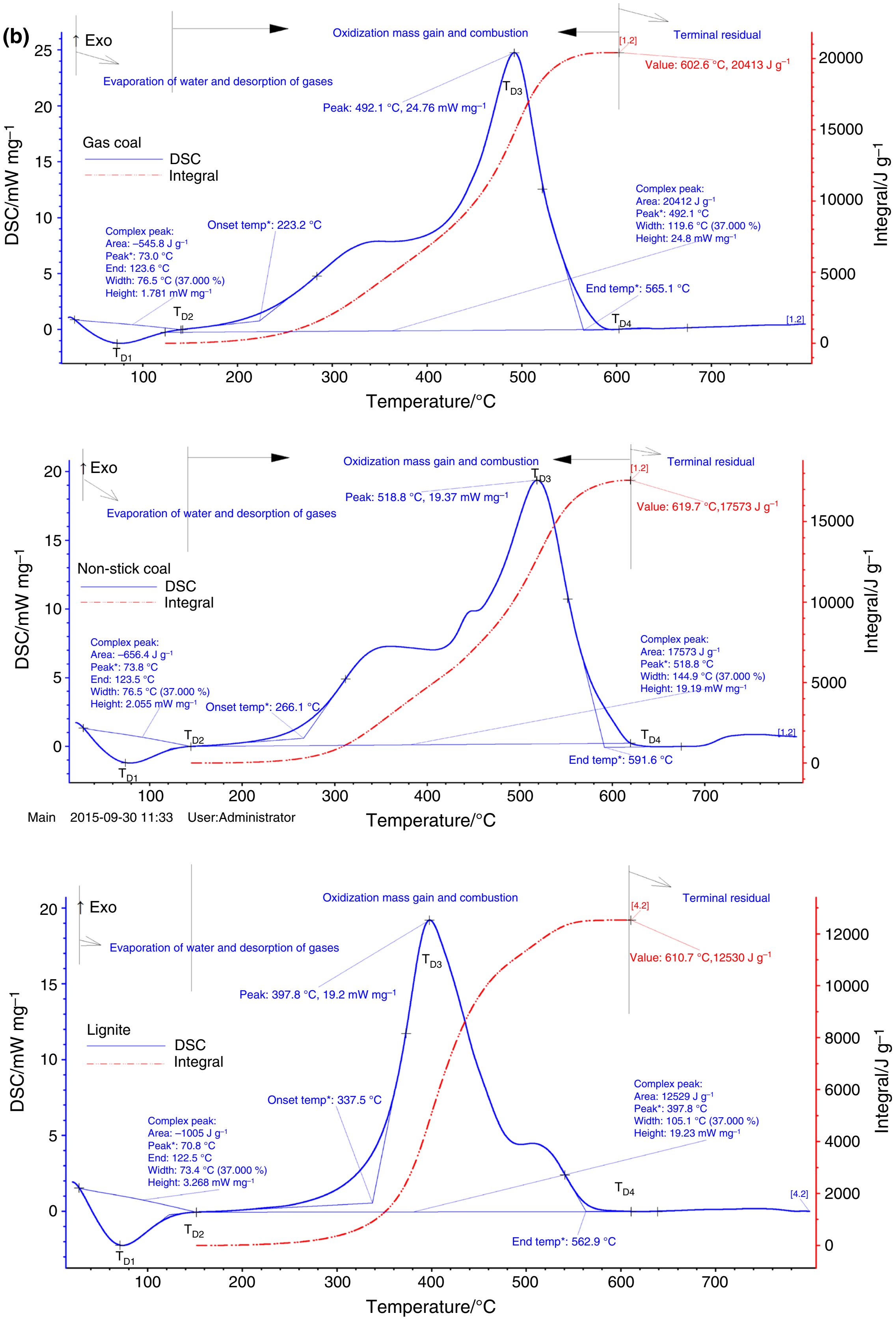


Fig. 2 continued

Table 3 Characteristic temperature of six types of coal by DSC tests

Sample	Anthracite	Lean coal	Coking coal	Gas coal	Nonstick coal	Lignite
T_{D1}/°C	86.14	75.39	73.85	72.95	73.00	70.80
T_{D2}/°C	123.20	126.11	123.52	123.60	118.81	122.53
T_{D3}/°C	557.80	521.75	519.48	491.71	458.07	396.82
T_{D4}/°C	654.89	639.29	619.66	602.73	549.19	610.83
Maximum heat releasing/mW mg^{-1}	27.96	21.74	19.37	24.76	52.18	19.2
Total heat of reaction/J g^{-1}	18,165	16,924	17,573	20,412	20,251	12,529

522.0, 519.0, 492.0, 458.0, and 397.0 °C, and the T_5 values were 655.0, 639.0, 617.0, 603.0, 549.0, and 611.0 °C after the metamorphic grade was decreased. Therefore, the metamorphic grade of coal is a crucial factor in determining spontaneous combustion. During exploitation, storage, or disposal, coal of a lower metamorphic grade should be cautiously handled and subject to prevention and control measures, to mitigate the risk of the spontaneous combustion of coal. However, in the current study, nonstick coal had a maximum DTG of -20.54 % min^{-1}, and lignite had a minimum DTG of -5.62 % min^{-1}. Therefore, when coal reached the combustion stage, the degree of violence of the combustion reaction for coal bores no relationship with the metamorphic grade, but was correlated with other factors, including composition of rock of coal, molecular structure, minerals content and distribution, and ash content. Various characteristic temperatures tested by applying TG for six types of coal are displayed in Table 2.

DSC results

Figure 2 shows the DSC experimental results for six types of coal. There were two thermal stages from the DSC curves. The first stage was an endothermic reaction caused by the evaporation of water and desorption of gases, which could correspond to the T_1 of the TG results. The second stage was an exothermic reaction corresponding to the TG results from oxidation mass gain to combustion (from T_2 to T_4). All DSC curves were defined according to four characteristic temperatures. T_{D1} was the temperature of maximum endothermic reaction with evaporation of water and desorption of gases, T_{D2} was the exothermic onset temperature of oxidation reaction, T_{D3} was the maximum heat release rate, and T_{D4} was the end of the combustion reaction. We found that T_{D1} occurred earlier with a lower metamorphic grade of coal; thus, the evaporation of water and the desorption of gases depended on the metamorphic grade of the coal. At the second stage, there was more than one exothermic peak during the heat release process for all coals, except for anthracite, which had only a single exothermic peak. This is because anthracite has a higher metamorphic grade, meaning that it has pure coal composition and a simple molecular structure. Moreover, gas coal showed the largest total heat of exothermic reaction, which is inconsistent with its metamorphic grade. This phenomenon could indicate that when the oxidation reaction of coal occurred, the intensity of the reaction depended on the physical or chemical properties of coal itself. In addition, because the total heat of exothermic reaction can be used to evaluate the severity of spontaneous combustion of coal, the heat generated by gas coal during the oxidation process must be immediately removed, leading to excessive heat accumulation in the coal seam. Related characteristic temperatures obtained from the DSC test are listed in Table 3.

FTIR results

Because of the inherent complexity of coal, FTIR is utilized to analyze the time, characteristics, and concentration of gases released during the oxidation process. Figure 3 presents a 3D diagram (temperature–wave number–absorbance) of FTIR compared with the TG thermal curve for six types of coal. We observed that the temperature of gases released reasonably matched the characteristic temperature of the TG test, and the temperature of maximum gases released was also close to the maximum mass loss

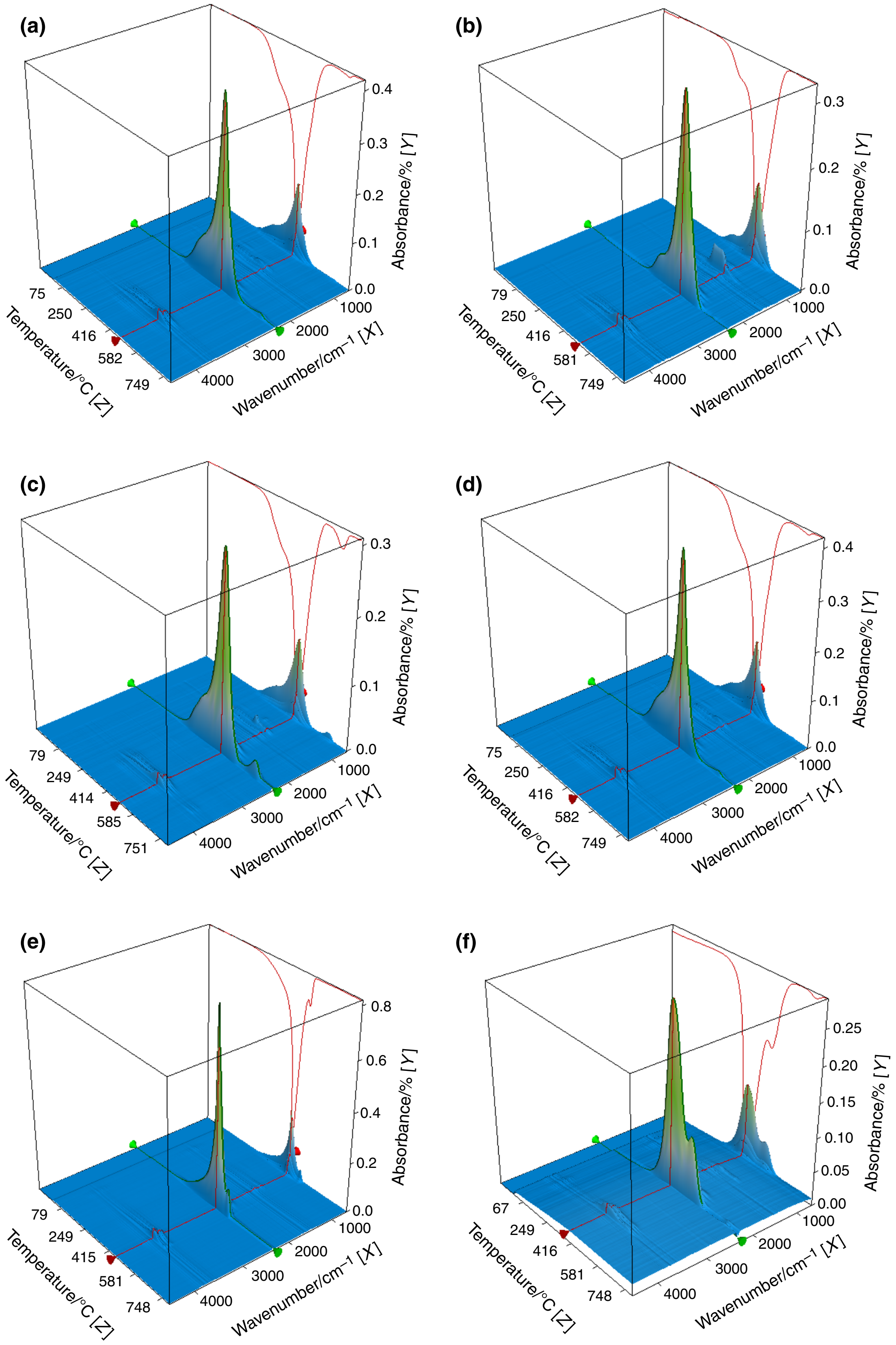


Fig. 3 3D results for gases releasing of **a** anthracite, **b** lean coal, **c** coking coal, **d** gas coal, **e** nonstick coal, and **f** lignite by using FTIR tests

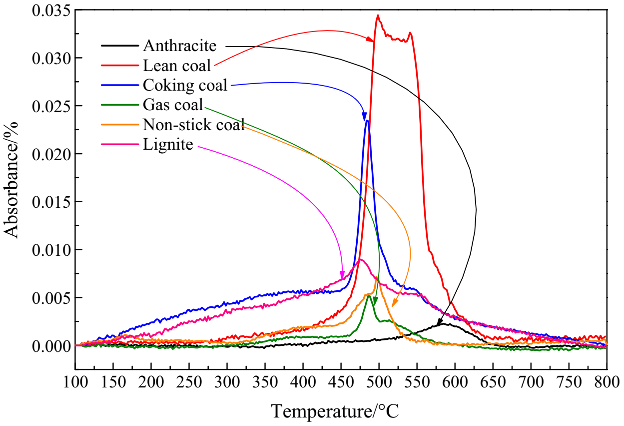


Fig. 4 Absorbance versus temperature diagram for CH_4 releasing of six types of coal

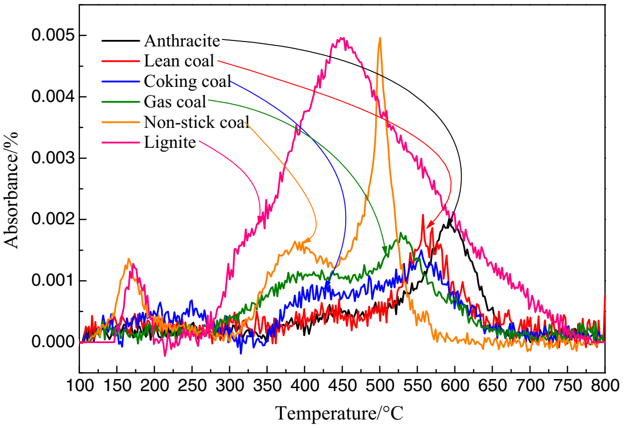


Fig. 5 Absorbance versus temperature diagram for H_2O releasing of six types of coal

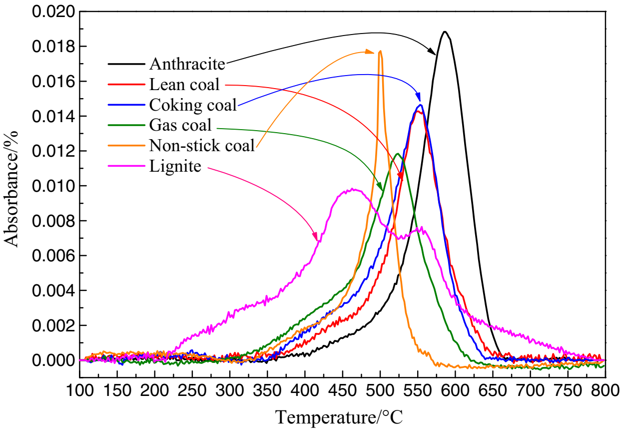


Fig. 6 Absorbance versus temperature diagram for CO releasing of six types of coal

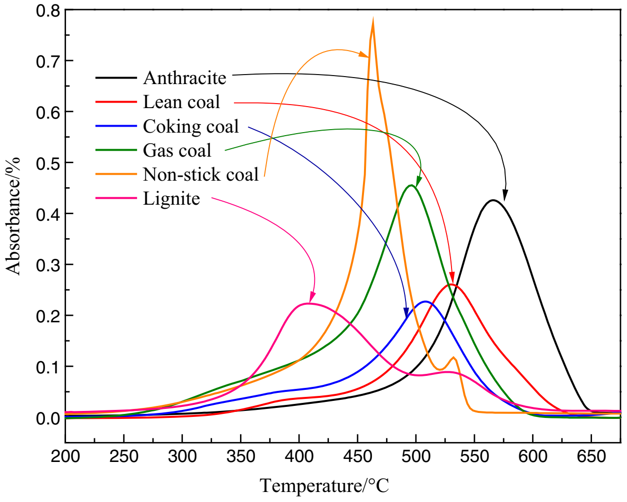


Fig. 7 Absorbance versus temperature diagram for CO_2 releasing of six types of coal

temperature. Therefore, the composition analysis for these released gases could be applied for further analysis of the oxidation process and evaluation of spontaneous combustion of coal. Figures 4, 5, 6 and 7 show the absorbance versus temperature for four index gases: CH_4, H_2O, CO, and CO_2, respectively. According to the literature, the wave numbers of 2650–2200 cm^{-1} and 850–400 cm^{-1} that are simultaneously present indicate CO_2 stretching; the wave numbers of 3100–2800 cm^{-1} combined with 1400–1100 cm^{-1} indicate CH_4 stretching; the wave numbers of 3700–3625 cm^{-1} indicate –OH stretching under dissociated conditions; the wave numbers of 1650–1350 cm^{-1} indicate –OH stretching of an in-of-plane ring bend, which could confirm the presence of H_2O; and the wave numbers of 2200–1900 cm^{-1} indicate CO stretching [21, 22]. H_2O stretching appeared in nonstick coal and lignite at ca. 170.0 °C because they contained more free and bound water than other types of coal. Moreover, the initial temperature of H_2O, CO, or CO_2 released took place earlier with lower metamorphic grades of coal. These results demonstrate the combustion reaction of higher metamorphic grades coal, which are difficult to process and are the same as the TG results. Another cause of spontaneous combustion of coal is CH_4, because high CH_4 and oxygen concentrations could cause a gas explosion. CH_4 is produced because of the oxidation of coal and CH_4 adsorption by coal in nature. According to the FTIR results, CH_4 was released from 450.0 to 600.0 °C, and lean coal released the most CH_4 during the combustion process. Anthracite released the least. Therefore, reinforced control technology for CH_4 release should be developed for various types of coal, mitigating the risk of a gas explosion. The characteristic parameters of the release of CO, H_2O, and CO_2 coal are presented in Table 4.

Table 4 FTIR analysis for gases releasing of six types of coal

Sample	Characteristic peak	Onset temperature/°C	Peak temperature/°C	Final temperature/°C	Height of peak/%
Anthracite	1344 cm^{-1} CH_4	348.865	584.973	700.918	0.25 E−2
Lean coal		239.243	498.541	679.838	3.40 E−2
Coking coal		441.622	483.784	582.865	1.84 E−2
Gas coal		428.973	489.000	660.865	0.56 E−2
Nonstick coal		291.946	496.432	572.324	0.70 E−2
Lignite		277.189	475.351	620.811	0.54 E−2
Anthracite	1604 cm^{-1} H_2O	348.865	593.405	700.918	1.97E−3
Lean coal		294.054	559.676	679.838	2.08E−3
Coking coal		280.973	555.459	694.595	1.50E−3
Gas coal		230.811	530.162	713.568	1.79E−3
Nonstick coal		114.865	502.757	574.432	4.96E−3
Lignite		146.487	452.162	766.270	4.96E−3
Anthracite	2115 cm^{-1} CO	363.622	589.189	669.297	1.88E−2
Lean coal		363.622	559.676	679.838	1.43E−2
Coking coal		353.081	555.459	641.892	1.47E−2
Gas coal		283.514	523.838	648.216	1.22E−2
Nonstick coal		332.000	500.649	572.324	1.80E−2
Lignite		216.054	464.811	778.919	0.98E−2
Anthracite	2356 cm^{-1} CO_2	337.994	565.289	648.267	0.207
Lean coal		325.834	529.726	622.773	0.125
Coking coal		311.612	508.389	607.964	0.106
Gas coal		252.341	496.535	598.482	0.222
Nonstick coal		247.082	463.343	553.435	0.368
Lignite		223.036	408.815	626.931	0.104

Conclusions

Our study confirmed the behaviors of spontaneous combustion of coal for six different metamorphic grades of coal, and various characteristic temperatures and index gases for coal spontaneous combustion were also determined by using the STA-FTIR technique. The main findings of this study are as follows:

1. From the industrial and element analysis results, six types of coal were defined completely. In addition, according to the TG results, coal with a lower metamorphic grade can readily trigger coal spontaneous combustion. However, when a combustion reaction occurs, the vigor of spontaneous combustion of coal has no relationship with the metamorphic grade, but is rather related to inherent physical and chemical properties of coal itself.
2. When the metamorphic grade of coal is lower, the temperature of maximum endothermic reaction with evaporation of water and desorption of gases also happens earlier. Gas coal has the maximum heat of combustion reaction when oxidation reaction occurs; therefore, heat removal in a gas coal mine should be strictly governed to mitigate the risk of spontaneous combustion of coal.
3. With a decrease in the metamorphic grade of coal, the initial temperature of H_2O, CO, or CO_2 release takes place earlier, indicating that the combustion reaction proceeds easily. Moreover, when decomposing, coal can release great amounts of CH_4, which may cause a gas explosion in a coal mine. Therefore, CH_4 detection technology should be implemented for coal use, storage, mining, transportation, and disposal.

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