Thermogravimetric Analysis of the Effects of Oxygen Concentration and Coal Granularity on Coal Spontaneous Combustion Characteristics

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***Abstract* — In order to study the effects of oxygen concentration and coal granularity on coal spontaneous combustion characteristics, the thermogravimetric analysis (TG analysis) for this field is made and the analysis of the TG curves show that: both coal powder granularity and oxygen concentration have major effects on the spontaneous combustion characteristics of coal; as coal powder granularity reduces, TG curve moves forward, the reaction interval of combustion is shortened, ignition occurs beforehand, the boundaries of the TG curves of coal powder spontaneous combustion are more obvious, the maximum spontaneous combustion rate occurs beforehand, the ignition temperature drops, the activation energy of the low temperature section gets smaller, and the extent to which it reduced is related to the type and the surface structure of coal; the higher the oxygen concentration, the more complete the reaction of coal and the greater the reaction rate; the increase in oxygen concentration shortens the total time of reaction.**

Keywords - oxygen concentration, coal granularity, TG analysis, coal spontaneous combustion characteristics

I INTRODUCTION

Coal spontaneous combustion is a complex physico-chemical process and its spontaneous combustion characteristics are subject to many factors. The research and analysis on the affecting factors are of great signification to the mechanism of coal spontaneous combustion(1-3). Characterized by powerful quantization, fewer requirements on sample numbers, high accuracy and good repeatability, it allows accurate measurement of the changes in quality and the change rate of substance(4-6). The author made a TG analysis experimental study of different granularity and oxygen concentration for the coalsamples from Huating Coal Mine. The effects of granularity and oxygen concentration on coal spontaneous combustion characteristics were obtained from an analysis of the TG curves of the combustion process of coal samples. This provides a theoretical basis for the further study on the natural characteristics of coal.

II. EXPERIMENTALCONDITION

A TG209 TG analyzer provided by Netzsch of Germany was used for the TG analysis of the samples taken from Huating Coal Mining, and the detailed experimental program see Table 1 and 2.

TABLE I.EXPERIMENTAL SCHEME OF TG ANALYSIS OF DIFFERENT COAL GRANULARITY（OXYGEN CONCENTRATION 21%）

NO。	Granularity（mm）	Coal quality (mg)	Initial temperature (℃)	Heating rate (℃.min^{-1})	Test environment oxygen concentration
1	0.45~0.90	27.559			
2	0.3~0.450	25.800	32	4	21%
3	0.20~0.30	29.086			
4	0.125~0.2	28.515			

TABLE II.EXPERIMENTAL SCHEME OF TG ANALYSIS OF DIFFERENT OXYGEN CONCENTRATION (GRANULARITY 0.200~0.300MM)

NO。	oxygen concentration（%）	Coal quality (mg)	Initial temperature (℃)	Heating rate (℃.min^{-1})	Test environment(ml/min) N_2	O_2
1	21	29.086			40	11．0
2	18	31.200			40	9．0
3	15	31.200			40	7．5
4	13	31.452	32	4	40	6．0
5	11	30.750			40	5．0
6	9	30.380			40	4．0
7	7	30.518			40	2．0

III. Experimental Results and Analysis

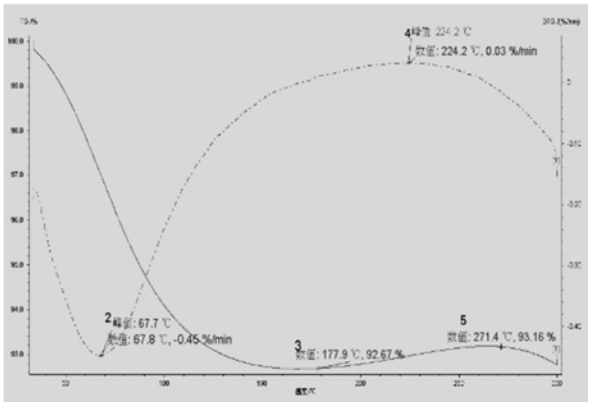
The TG curve of coal showed that, at the initial stage, coal sample compound was mainly physical adsorption, and chemical adsorption and chemical reactions were at a slow rate. The amount of physical adsorption by coal samples was greater than the desorption amount, causing the initial weight gain of coal samples; the chemical adsorption of coal is a reversible exothermic process at a greater rate and presents easy attainment of equilibrium. At that time, dry air will take water and methane, carbon monoxide and vapor that are obtained in some coal samples, causing weight loss after physical adsorption equilibrium. Subsequently, coal samples were heated at a certain rate of temperature rise, and the internal energy of the heated molecules increased, physical adsorption decreased while the chemical adsorption increased and was accompanied by chemical reactions; oxygen consumption rate increased, so did oxygen absorption rate.

When the temperature rose to a critical temperature, the rate of physical and chemical adsorption of coal samples reached the maximum, indicating the decomposition or depolymerization of alkyl side chains, oxygen-containing functional groups, bridges and other small molecules around the condensed aromatic systems of coal sample molecular structures and the release in the form of small molecule volatile, which caused a weight loss. The further increase in temperature accelerated the cracking of the small molecules in the coal molecular structures and thereby the active structure growth and enhanced its adsorption amount; eventually a dynamic equilibrium was attained when the desorption amount and the amount produced in chemical reactions were equal to the amount of adsorption oxygen. After a stable period, increasing active groups were exposed, seeing an increasing oxygen adsorption amount and accelerating the cracking of small molecules and initiating the fractures of big molecular structures. Due to a large amount of oxygen consumption in chemical reactions at the previous stage, the available oxygen and gases for reaction was depleted, and massive available pores in coal surface might absorb a large amount of oxygen, displaying a slowing weight loss rate and an increasing mass ratio of coal samples, until the coal started spontaneous combustion and generated vast amounts of heat, alkanes, alkenes, CO and CO_2 when the coal samples lost weight again(7-9).

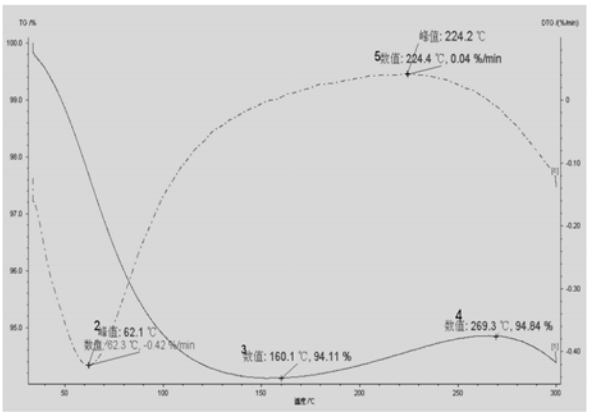
With the continuous coal spontaneous combustion, there was a point of maximum weight loss rate. This shows that violent chemical reactions occurred in coal molecules, combustion rate significantly increased and massive gases were produced, leading to a significant weight loss in coal samples. As manifested in TG curves, the weight loss curves leveled off and the combustion temperature of coal was reached, signaling the end of the ignition process of coal samples' spontaneous combustion.

A.Effects of Coal Granularity on Pyrolysis Products

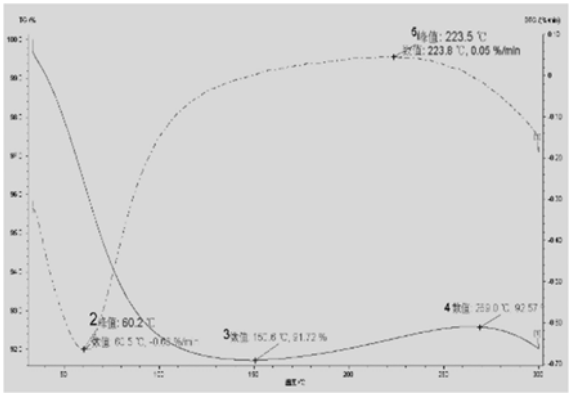
Coal powder fineness is an indicator having considerable effects on spontaneous combustion characteristics. When powdered coal is of small granularity, pyrolysis products are independent of the granularity; the coal powder of large granularity is different from finelypowdered coal in spontaneous combustion characteristics, and the surface can provideconditions for secondary reactions. The pyrolysis products near coal grain center want escape, and cracking, condensation or polymerization of them and thereby carbon deposits may occur during migration. The larger the granularity, the greater the deposits and the fewer the obtained pyrolysis products will be. Moreover, coal specific surface area increases and thus the activation energy decreases as coalpowdergranularity decreases, and coal powderis more spontaneously flammable.



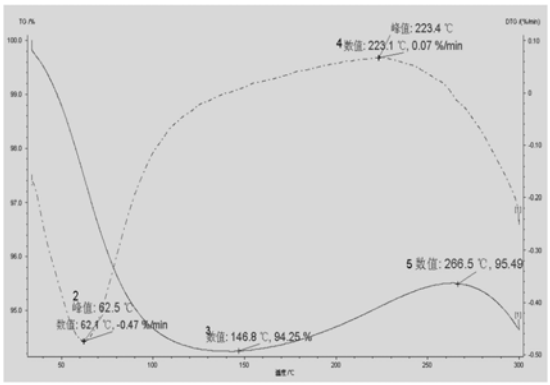
Figure（a） TGA curve chart（granularity 0.45~0.90mm）



Figure（b） TGA curve chart（granularity 0.30~0.45mm）



Figure（c） TGA curve chart（granularity 0.20~0.30mm）



Figure（d） TGA curve chart（granularity 0.125~0.20mm）

Figure 1 TGA curve chart of different coal granularity

TABLE III.TG ANALYSIS OF DIFFERENT COAL GRANULARITY（OXYGEN CONCENTRATION 21%）

NO.		TGA			DTG		
		①Maximum point of weight ratio high adsorption T	③Inflection point of weight ration	⑤Minimum point of weight ratio active T		②Maximum point of weight loss critical T	④Minimum point of weight loss speedup T.
1	T℃ Weight ratio%		177.9 92.67	271.4 93.16	T℃ Weight loss rate	67.8 0.45	224.2 0.03
2	T℃ Weight ratio%		160.1 94.11	269.3 94.84	T℃ Weight	62.3 0.42	224.4 0.04
3	T℃ Weight ratio%		150.6 91.72	269.0 92.57	T℃ Weight	60.5 0.66	228.5 0.05
4	T℃ Weight ratio%		146.8 94.25	266.5 95.49	T℃ Weight	62.1 0.47	223.1 0.07

Note： DTG curve weight loss rate，unit：%/min

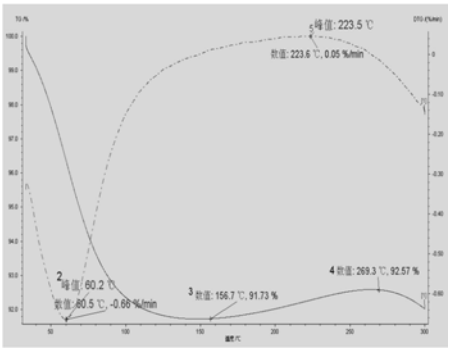
Fig. 1 and Table 3 show that weight losses were different in thermal decomposition of the coal samples of different granularity. The smaller the coal powder granularity, the more obvious the boundaries of the TG curves of coal powder spontaneous combustion and the more prematurely the maximum spontaneous combustion rate appeared, leading to lower ignition temperature and beforehand ignition. Table 3.6 shows that, at the critical temperature of the coal samples that are 0.2~0.3mm in granularity, the point of maximum weight loss rate of 0.66%/min appeared the earliest at 60.5℃. At the inflection temperature (cracking temperature), the mass ratio was the minimal (92.67%); the smaller the coal powder granularity, the less the activation energy of the high and low temperature section of coal powder spontaneous combustion, and the extent to which it reduced is related to the type and the surface structure of coal; coal powder granularity has significant effects on its spontaneous combustion characteristics, and its physical structure and spontaneous combustion characteristics got improved as coal powder granularity decreased; the weight loss in thermal decomposition decreased as coal sample granularity increased. This indicates that, as coal powder granularity increases, reaction specific surface area decreases and thermal decomposition reactions are not complete; as coal granularity decreases, the total time of spontaneous combustion of coal samples is shortened; as coal sample granularity increases, coal specific surface area increases and the combustion conditions get more favorable and more surface activity structures are exposed to air, and these situation increasethe corresponding oxygen consumption rate and coal combustion rate, and the combustion promotes the decrease in coal granularity, further increases the combustion rate and shortens the reaction time.

B. Effects of Different Oxygen Concentration on Spontaneous Combustion Characteristics

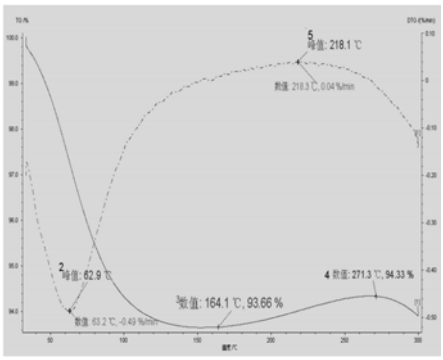
Coal-oxygen compound is a combination of solids and single gases, and its reaction rate is proportional to the concentration of reactive gases, and basically presents a linear relationship.

Fig. 2 shows the TG curves of the samples of the

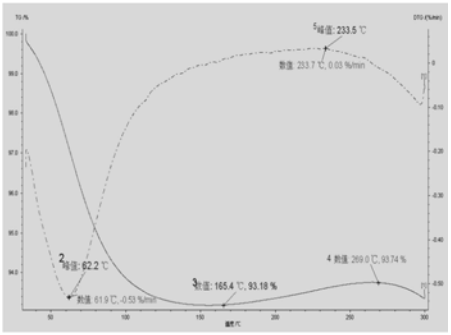
same granularity from Huating Coal Mine at different oxygen concentration.



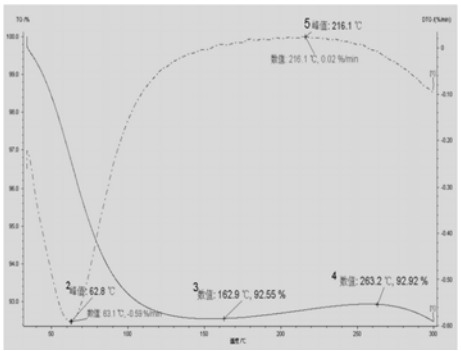
Figure（e） TGAcurve chart（oxygen concentration 21%）



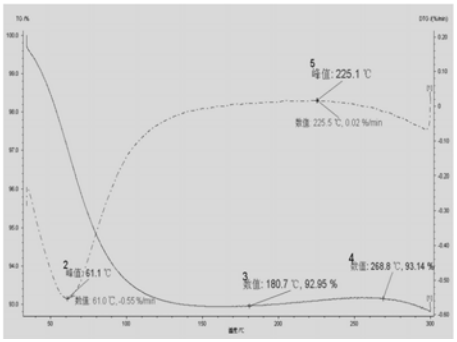
Figure（f） TGAcurve chart（oxygen concentration18%）



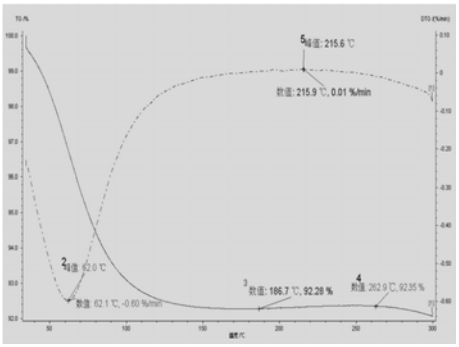
Figure（g） TGAcurve chart（oxygen concentration 15%）



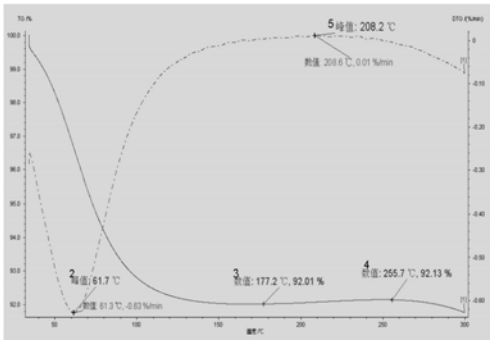
Figure（h） TGAcurve chart（oxygen concentration13%）



Figure（i） TGAcurve chart（oxygen concentration 11%）



Figure（j）TGAcurve chart（oxygen concentration 9%）



Figure（k） TGAcurve chart（oxygen concentration7%）

Figure 2 TGA curve chart of different granularity

TABLEⅣ.TG analysis of different oxygen concentration（granularity 0.125~0.200mm）

NO.		TGA			DTG		
		①Maximum point of weight ratio high adsorption T	③Inflection point of weight ration	⑤Minimum point of weight ratio active T		②Maximum point of weight loss critical T	④Minimum point of weight loss speedup T.
1 21%	T℃ Weight ratio%		156.7 91.73	269.3 92.57	T℃ Weight loss rate	60.5 0.66	223.6 0.05
3 15%	T℃ Weight ratio%		165.4 93.18	269.0 93.74	T℃ Weight loss rate	61.9 0.53	233.7 0.03
4 13%	T℃ Weight ratio%		162.9 92.55	263.2 92.92	T℃ Weight loss rate	63.1 0.59	216.1 0.02
5 11%	T℃ Weight ratio%		180.7 92.95	268.8 93.14	T℃ Weight loss rate	61.0 0.55	225.5 0.02
6 9%	T℃ Weight ratio%		186.7 92.28	262.9 92.35	T℃ Weight loss rate	62.1 0.6	215.9 0.01
7 7%	T℃ Weight ratio%		177.2 92.01	255.7 92.13	T℃ Weight loss rate	61.3 0.63	208.6 0.01

Note： DTG curve weight loss rate，unit：%/min

Fig. 2 and Table 4 show that there were perceptible differences between the TG curves at different oxygen concentration. As oxygen concentration increased, TG curves moved forward and got close to the low temperature section, and the maximum weight loss rate of coal samples reduced; with the oxygen concentration of 21% and at the critical temperature of coal samples, the point of maximum weight loss rate of 0.66%/min appeared the earliest; at the inflection temperature (cracking temperature), the minimum mass ratio (maximum weight loss) of 92.67% appeared the earliest at 157.7 ℃. This indicates that the higher the oxygen concentration, the more complete the coal reaction and the greater the reaction rate; as oxygen concentration increases, the ignition time of coal spontaneous combustion decreases.

IV. CONCLUSIONS

(1) Both coal powder granularity and oxygen concentration have major effects on the spontaneous combustion characteristics of coal samples;

(2) As coal granularity decreases, TG curve moves forward, the reaction interval of combustion is shortened, ignition occurs beforehand, the boundaries of TG curves of coal powder spontaneous combustion are more obvious, the maximum spontaneous combustion rate occurs beforehand, ignition temperature drops, the activation energy of the low temperature section gets smaller, and the extent to which it reduces is related to the type and the surface structure of coal;

(3) The spontaneous combustion characteristics of coals vary obviously under the action of different oxygen concentrations. With the oxygen concentration of 21% and at the critical temperature of coal samples, the point of maximum weight loss rate of 0.66%/min appears the earliest; at the inflection temperature (cracking temperature), the minimum mass ratio (maximum weight loss) of 92.67% appeared the earliest; the higher the oxygen concentration, the more complete the coal reaction and the greater the reaction rate; the increase in oxygen concentration leads to the shortened total reaction time.

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