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LABORATORY STUDY ON LOW-TEMPERATURE COAL SPONTANEOUS COMBUSTION IN THE AIR OF REDUCED OXYGEN AND LOW METHANE CONCENTRATION

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Original scientific paper

Laboratory tests were conducted to study coal spontaneous combustion at low-temperature (30 ÷ 200 °C) in the air of reduced oxygen (10 ÷ 21 %) and low methane concentrations (0 ÷ 3 %). The production of carbon monoxide in coal spontaneous combustion was analysed. The test results show that both oxygen and methane concentrations affect coal spontaneous combustion. At given oxygen concentration the methane concentration in air had a positive impact on coal spontaneous combustion and the impact increased with methane concentration. This impact was weakened and diminished with the rise of temperature. The initial carbon dioxide temperate at which carbon monoxide started to appear increased with the reduction of oxygen concentration in the air and the increase was somewhat negated by the presence of increased methane concentration in air. However, as the oxygen concentration reduced to a certain value, methane concentration showed little or no influence on coal spontaneous combustion. The low temperature sorption characteristics of oxygen and methane by coal were applied to explain the results. The findings in this study are particularly useful in the prediction and control of coal spontaneous combustion in gassy coal mines.

Keywords: *coal mine; coal spontaneous combustion; low temperature; low methane concentration; oxygen concentration*

Laboratorijska analiza spontanog izgaranja ugljena na niskoj temperaturi u zraku s reduciranim kisikom i niskom koncentracijom metana

Izvorni znanstveni članak

Provedena su laboratorijska ispitivanju u svrhu proučavanja spontanog izgaranja ugljena na niskoj temperaturi (30 ÷ 200 °C) u zraku s reduciranim kisikom (10 ÷ 21 %) i niskim koncentracijama metana (0 ÷ 3 %). Analiziralo se stvaranje ugljičnog monoksida kod spontanog izgaranja ugljena. Rezultati ispitivanja pokazuju da i koncentracije kisika i metana djeluju na spontano izgaranje ugljena. Pri datoj koncentraciji kisika koncentracija metana u zraku je imala pozitivan učinak na spontano izgaranje ugljena, a učinak se povećao s koncentracijom metana. Taj je učinak oslabio i smanjio se porastom temperature. Početna je temperature ugljičnog dioksida, kod koje se ugljični monoksid počeo javljati, porasla smanjenjem koncentracije kisika u zraku, a porast je donekle smanjen povećanjem koncentracije metana u zraku. Međutim, kako se koncentracija kisika smanjivala do određene vrijednosti, koncentracija metana je pokazivala malo ili nikakvog utjecaja na spontano izgaranje ugljena. Uzete su u obzir karakteristike ugljena kod niske temperature sorpcije kisika i metana kako bi se objasnili rezultati. Dobiveni rezultati su od posebne koristi u predviđanju i praćenju spontanog izgaranja ugljena u plinovitim ugljenokopima.

Ključne riječi: *koncentracija kisika; niska koncentracija metana; niska temperatura; spontano izgaranje ugljena; ugljenokop*

1 Introduction

Spontaneous combustion of coal is a serious hazard in coal-related industries such as coal mining, coal transportation and coal storage. If not properly controlled, it can lead to the emission of toxic and explosive gases together with propagation to open fire, and it can become a potential ignition source for an explosion if exposed to a flammable mixture of gas. It is well known that the physical and chemical interaction between coal and molecular oxygen (O_2), or coal oxidation, is the major reason responsible for spontaneous combustion of coal. Coal oxidation is a complicated process involving a number of phenomena such as the release of heat and the emission of gaseous products. If the heat is retained, the coal mass will increase in temperature and the oxidation rate will increase, leading to spontaneous combustion [1÷3].

Research into coal oxidation and its application for the detection and prevention of spontaneous combustion of coal has continued for more than 150 years [4÷9]. One strand of the research work is on the effect of various factors on the oxidation process and characteristics of gaseous products of coal [10÷12]. The gaseous products, in particular carbon monoxide (CO) and ethene (C_2H_4), are important indicators to detect the onset and development stages of spontaneous combustion in coal mines [13÷15]. Coal oxidation and its gaseous products are affected by several factors such as composition and physical properties of coal, temperature, moisture content and partial pressure of O_2 in the gas medium [16÷19].

The atmosphere in underground coal mines often contains methane (CH_4) and has less O_2 concentration than in normal atmospheric air. In-situ measurements indicate that the atmosphere in active goafs can have a wide range of O_2 (0 ÷ 21 %) and CH_4 (0 ÷ 100 % CH_4), and the CH_4 concentration in roadways adjacent to goafs can have up to 3 % [20÷22]. Some field observations suggest that low CH_4 concentration in mine atmosphere tends to shorten the incubation period of spontaneous combustion of coal. For example, the incubation period of spontaneous combustion of the 3_L coal seam in Tengdong coal mine, Shandong, China, was measured to be between 3 to 6 months in normal air condition and field observation indicated that the incubation period was only 29 days in the goaf with 0,5 to 4 % CH_4 concentration.

A number of investigators have observed a dependence of the rate of coal oxidation on oxygen concentration in the gas medium [23], they suggested that the rate of oxygen consumption over a wide range of oxygen concentration can be expressed as a power of the partial pressure of oxygen in the oxidation medium with the exponent varying between zero and unity. However, in these studies no CH_4 was present in the gas medium. In fact, there is little published literature available on how coal oxidation and spontaneous combustion are affected by the combined effect of the reduced partial pressure of

O_2 and the existence of low CH_4 concentration in mine atmosphere.

CO is the most commonly used gas indictor in the prediction of coal spontaneous combustion in coal mines as its appearance and production rate indicate the stages of coal spontaneous combustion [24÷26]. This laboratory study aims to understand the effect of the reduced partial pressure of O_2 (10 to 21 %) and low CH_4 concentration (0 ÷ 3 %) in mine atmosphere on coal spontaneous combustion in the temperature range from 30 to 200 °C.

2 Test method

The test apparatus consists of a gas mixing system, a temperature control system, a gas analysing system, and a coal sample cylinder, as shown in Figure 1. The apparatus is designed to simulate coal spontaneous combustion under various gas conditions and analyse the gaseous products of spontaneous combustion. The gas mixing system can make gases of any mixture of O_2, N_2 and CH_4 and provide steady gas flow to the sample cylinder. The temperature control system can provide controllable temperature environments for coal spontaneous combustion. The gas analysing system is used to analyse the composition of the gaseous products of coal spontaneous combustion.

The gas mixing system consists of an O_2 cylinder, a N_2 cylinder, a CH_4 cylinder, a cylinder for mixed gas, gas pressure regulators, gas pressure stabilisers, gas flow metres, gas flow valves and gas pipelines. A mixed gas of any combination of O_2, N_2 and CH_4 concentrations can be obtained by adjusting flow rates from the cylinders of O_2, N_2 and CH_4. The cylinder for mixed gas is connected to the coal sample container and ready for use.

The temperature control system includes an enclosed tank, a temperature sensor and an electric heating unit. The inner and outer layers of the tank use stainless steel and cold-rolling carbon steel respectively and the gap between the layers is filled with high density aluminium silicate fibre for heat in simulation. A Pt100 temperature sensor is installed to monitor the tank temperature and stainless steel heating pipes are used to control the tank temperature. The heating pipe lines are installed at the bottom and on both sides of the tank. The system is designed to control the tank temperature in the range of 0 and 650 °C and the rate of temperature increase between 1 and 20 °C/min. The gas analysing system uses a coal mine specific gas chromatography GC-4085B. It can detect CO, O_2, N_2, CO_2, CH_4, C_2H_4, C_2H_6, C_2H_2, C_3H_8 and C_4H_{10}. It takes about 4 to 8 min to analyze each sample. The minimum detection limit of CO is 0,5 ppm. A gas drying device is installed on the gas inlet side of the gas chromatography.

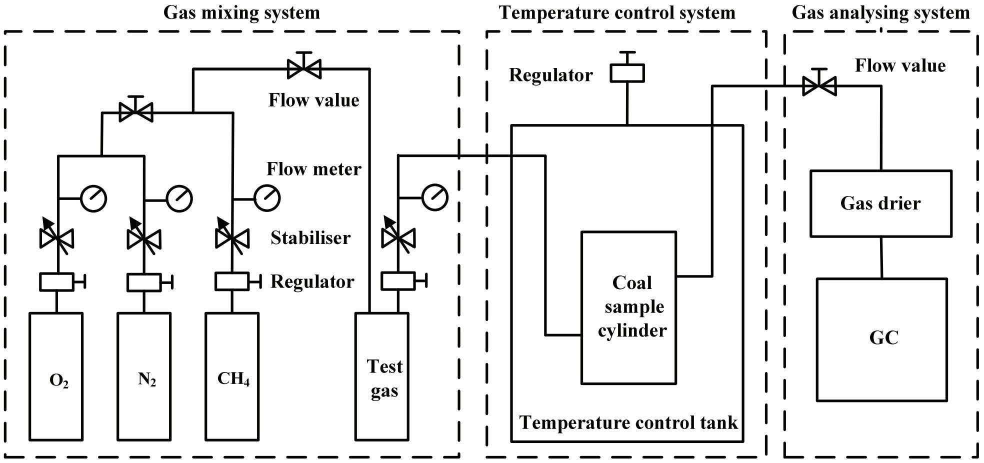


Figure 1 Test apparatus

The coal sample cylinder is shown in Fig. 2. The cylinder is 350 mm high and 100 mm in diameter. The top of the cylinder is sealed with a sealing cap and the bottom of the cylinder is welded and sealed. The gas inlet is on the left bottom of the cylinder and the gas outlet is on the right top of the cylinder. A layer of meshed copper wires is inserted 80 mm above the bottom of the cylinder to ensure even distribution of gas inflow and coal sample is placed on the copper wires. A layer of asbestos is covered on the top of coal sample to prevent the gas outlet from blockage. Three temperature sensors are installed inside the cylinder and they are labelled as 1#, 2# and 3# from the top to bottom of the cylinder. The sensors are located 230 mm, 180 mm and 130 mm respectively above the bottom of the cylinder.

Bituminous coal was sourced from the 3_L coal seam in Tengdong coal mine, Shandong, China. Lumps of coal were taken from a freshly exposed coal face of the mine with a chain saw by removing a layer of coal from the face of 25 cm thick to avoid the possibility of per oxidation. The coal was sealed in plastic cling wrap upon its retrieval from the face and filled with nitrogen. The coal was then transported to a laboratory in Shandong University of Science Technology, crushed and sieved into samples of particle sizes of 0,18 to 0,38 mm. Its proximate analysis results (air dry basis) show that it contains 1,47 % moisture, 7,58 % ash, 53,55 % fixed carbon, 37,4 % volatile matter and 0,45 % total sulphur.

For each test run, the cylinder was loaded with coal sample of 200 g and put in the temperature control tank. The inlet gas of preset compositions was fed into the cylinder at a steady flow rate of 100 ml/min. The start test temperature was set at 30 °C which was in line with the in situ temperature of the 3_L coal seam. The temperature increase rate was controlled at 1 °C/min and the final test temperature was set at 200 °C. The gaseous product at the temperature range of 30 to 200 °C was sampled and analyzed of its CO concentration with the gas chromatography. The tests were conducted for a total of 12 coal samples in the gas of various compositions of O_2, CH_4 and N_2. Tab. 1 lists the compositions of gas in the tests.

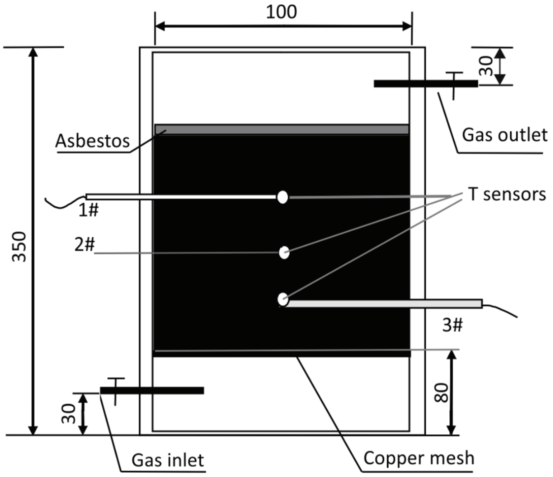


Figure 2 Coal sample cylinder

Table 1 The inlet gas compositions

Number	O_2 %	CH_4 %	N_2 %
1	10	0	90
2	10	0,82	89,18
3	10	1,94	88,06
4	10	3,06	86,94
5	15	0	85
6	15	0,82	84,18
7	15	1,94	83,06
8	15	3,06	81,94
9	21	0	79
10	21	0,82	78,18
11	21	1,94	77,06
12	21	3,06	75,94

Table 2 CO production (ppm) in gases of 10 % O_2 and 0 ÷ 3,06 % CH_4

Temperature ℃	CH_4 0 %	CH_4 0.82 %	CH_4 1.94 %	CH_4 3.06 %
30	0	0	0	0
40	0	0	0	0
50	0	0	2	5
60	9	11	12	14
70	23	23	25	31
80	66	68	59	51
90	76	82	71	62
100	106	116	109	93
110	121	125	114	103
120	165	176	177	132
130	461	452	457	426
140	751	765	745	726
150	1156	1178	1187	1125
160	2012	1990	2063	1941
170	2599	2639	2569	2487
180	3289	3360	3362	3218
190	4116	4140	4095	3983
200	5568	5600	5598	5439

3 Results and discussion

Tab. 2 lists the CO productions of coal spontaneous combustion in gases containing10 % O_2 and 0 ÷ 3 % CH_4 in the temperature range from 30 to 200 °C. For gases containing 15 % and 21 % O_2 the CO productions were listed in Tabs. 3 and 4 respectively.

Table 3 CO production (ppm) in gases of 15 % O_2 and 0 ÷ 3,06 % CH_4

Temperature °C	CH_4 0 %	CH_4 0,82 %	CH_4 1,94 %	CH_4 3,06 %
30	0	0	2	6
35	0	0	5	9
40	0	3	6	11
45	5	3	8	9
50	9	8	11	15
55	8	11	13	18
60	11	13	16	22
65	18	23	22	25
70	26	29	26	32
75	29	32	35	38
80	56	61	58	63
85	59	67	72	78
90	72	79	81	88
95	91	98	89	92
100	106	113	108	127
110	125	131	133	146
120	146	155	159	176
130	456	507	523	546
140	646	638	618	668
150	1978	2566	2437	2618
160	6360	6669	6546	6807
170	9139	8916	8190	8286
180	9060	9066	9280	9065
190	8940	9328	9390	9486
200	10606	10493	10450	10562

Table 4 CO production (ppm) in gases of 21 % O_2 and 0 ÷ 3,06 % CH_4

Temperature °C	CH_4 0 %	CH_4 0,82 %	CH_4 1,94 %	CH_4 3,06 %
30	6	5	8	8
35	8	7	10	10
40	9	10	12	15
45	9	16	15	16
50	14	18	16	18
55	16	18	22	26
60	27	22	26	31
65	28	32	29	38
70	37	41	51	43
75	46	52	64	75
80	73	69	76	83
85	95	86	92	102
90	115	108	112	110
95	128	132	146	152
100	147	156	169	186
110	172	185	198	213
120	213	221	240	268
130	603	663	750	820
140	801	931	859	995
150	2950	3067	3182	3238
160	8978	8878	8650	8853
170	12446	12262	11385	13272
180	15608	15952	15380	15183
190	18152	17870	18328	17954
200	19252	19585	18992	19368

3.1 The Initial CO Temperature

The initial CO temperature is defined as the temperature at which CO starts to appear in coal spontaneous combustion. In this study, the initial CO temperature and production are extracted from Tabs. 2 to 4 and summarized in Tab. 5. It can be seen from Tab. 5 that the initial CO temperature and production varied considerably with O_2 and CH_4 concentrations. In case that no CH_4 was present in the gas medium, the initial CO temperature increased with reduction in O_2 concentration. For example, the initial CO temperature was 30 °C with 21 % O_2 and the temperature increased to 50 °C and 60 °C as O_2 concentration dropped to 15 % and 10 % respectively. In case that low CH_4 concentration was present in the gas medium, the initial CO temperature also increased with reduction in O_2 concentration though the increase was somewhat subdued. For example, with increase in CH_4 concentration from 0 to 3,06 % the initial CO temperature dropped from 60 °C to 50 °C with 10 % O_2 and from 50 °C to 30 °C with 15 % O_2.

The results indicate that the O_2 and CH_4 concentrations affect coal spontaneous combustion. The increase in the initial CO temperature with the reduced O_2 concentration indicates that O_2 decrease in the gas medium has negative impact on coal spontaneous combustion. The subdued increase in the initial CO temperature with the presence of the low CH_4 concentration indicates that CH_4 in the gas medium has a positive impact on coal spontaneous combustion.

Table 5 The initial CO temperature and production

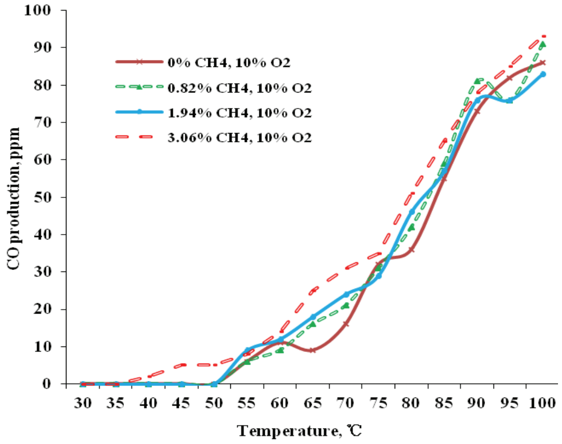
O_2	CH_4 =0		CH_4 = 0,82 %		CH_4 = 1,94 %		CH_4 = 3,06 %	
%	*T* °C	CO ppm	*T* °C	CO ppm	*T* °C	CO ppm	*T* °C	CO ppm
10	60	9	60	11	50	2	50	5
15	50	5	40	3	30	2	30	6
21	30	6	30	5	30	8	30	8

3.2 CO production

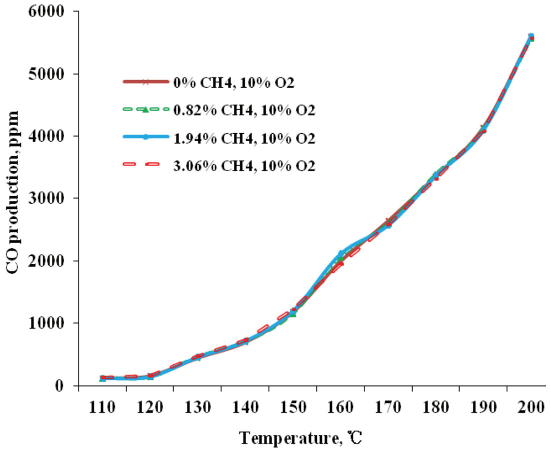
For gases of 10 % O_2 and 0 ÷ 3 % CH_4, the CO production in the temperature range of 30 to 200 °C is shown in Fig. 3. It can be seen from Fig. 3 that at the same temperature the CO production had no obvious and consistent variation with the CH_4 concentration. For example, at 100 °C the CO production was 106, 116, 109 and 93 ppm with the CH_4 concentration of 0 %, 0,82 %, 1,94 % and 3,06 % respectively. This indicates that the presence of low CH_4 concentration in the gas medium of 10 % O_2 has no or little influence on the CO production.

For gases of 15 % O_2 and low CH_4 concentrations, the CO production in the temperature range of 30 to 200 °C is shown in Fig. 4. It can be seen from Fig. 4 that at the same temperature the CO production generally increased with the CH_4 concentration. For example, at 100 °C the CO production was 106, 113, 108 and 127 ppm with the CH_4 concentration of 0 %, 0,82 %, 1,94 % and 3,06 % respectively. This indicates that CH_4 had a positive impact on CO production. However, the positive impact gradually diminished with the rise of temperature. For example, at 200 °C the CH_4 concentration showed no or little influence on the CO production.

For gases of 21 % O_2 and low CH_4 concentrations, the CO production in the temperature range of 30 to 200 °C is shown in Fig. 5. It can be seen from Fig. 5 that at the same temperature the CO production generally increased with the CH_4 concentration. For example at 100 °C the CO production was 147 ppm with the gas of zero CH_4 concentration and the CO production increased to 156, 169 and 189 ppm as the CH_4 concentration rose to 0,82 %, 1,94 % and 3,06 % respectively. This indicates that CH_4 had a positive impact on CO production. However, the positive impacts lowly diminished with the rise of temperature. For example, at 160 °C the CH_4 concentration had no or little influence on the CO production.



(a) The temperature range 30 to 100℃

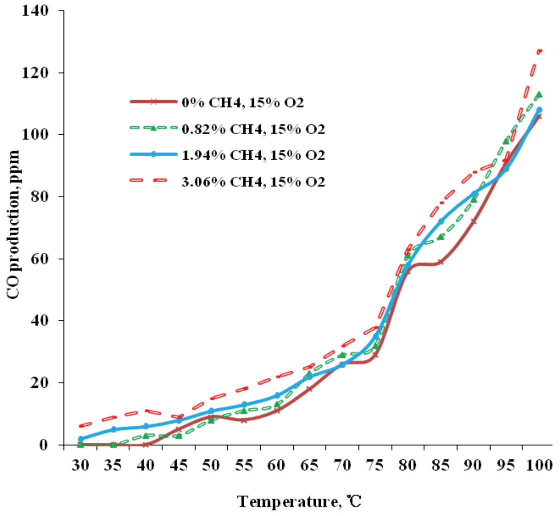


(b) Temperature range of 100 to 200 °C

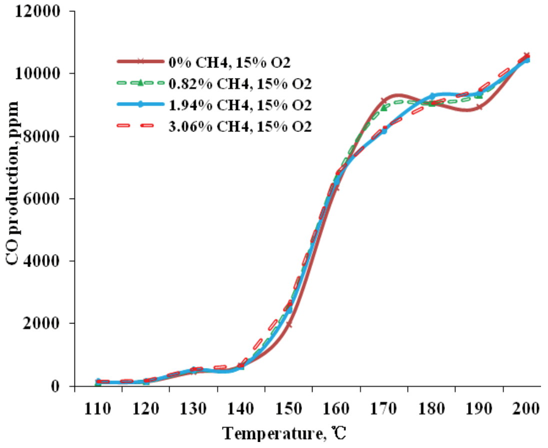
Figure 3 CO production for gases of 10 % O_2 and 0 ÷ 3 % CH_4

A comparison of results in Figs. 3, 4 and 5 shows that at the same temperature and with the low CH_4 concentration the CO production with 21 % O_2 was significantly higher than that with 10 % and 15 % O_2 and this became more obvious with the rise of temperature. For example at 200 °C the CO production with 21 % O_2 was 13000 ÷ 14000 ppm and the CO production dropped to 8000-9000ppm with 10 and 15 % O_2. This indicates that the O_2 concentration in the gas medium had a much more significant effect than the low CH_4 concentration on coal spontaneous combustion.

Low temperature spontaneous combustion of coal is a complicated process and has not yet been fully understood. However it is generally accepted that the gaseous products of coal spontaneous combustion in low temperature such as CO and $CO_2$are formed through the sorption sequence. The sorption sequence is considered to be the three-step processes, i.e. the chemisorption of O_2 on surface of coal pores and formation of unstable carbon-oxygen complexes, decomposition of the unstable solid oxygenated intermediates to gaseous products such as CO and CO_2 and stable solid complexes, and the degradation of the stable solid complexes and generation of new active sites for coal spontaneous combustion. As heat is generated mainly from the chemical adsorption of O_2 by coal and chemical reaction in coal spontaneous combustion, the O_2 concentration in the gas medium is the main reason for the heat accumulation of coal and the subsequent CO production. The reduction in O_2 concentration in the gas medium slows coal spontaneous combustion and CO production.



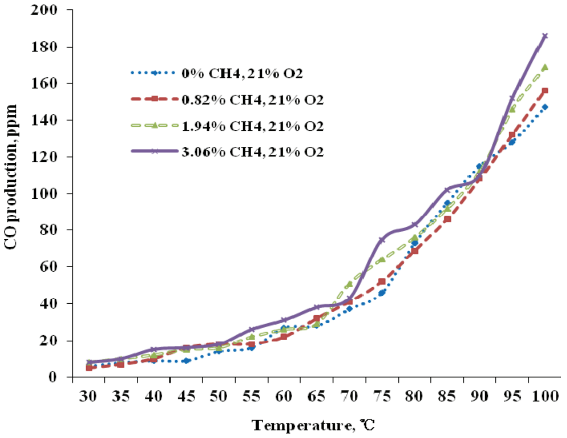
(a) The temperature range 30 to 100 °C



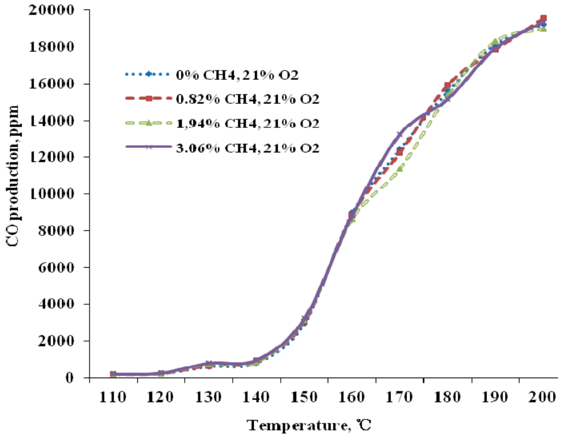
(b) The temperature range 100 to 200 °C

Figure 4 CO production for gases of 15 % O_2 and 0 ÷ 3 %CH_4

In case that low CH_4 concentration is present in coal spontaneous combustion, as coal has higher geophysical adsorption capability of CH_4 than O_2 [27], CH_4 will occupy part of coal surface through physical adsorption and this will reduce the area of coal surface available for O_2 adsorption. The partial occupation of coal surface by CH_4 molecules may hinder the dissipation of heat generated through the chemisorptions and chemical reaction between O_2 and coal, resulting in more heat accumulation on coal surface and accelerate coal spontaneous combustion. With the rise in temperature, more CH_4 desorbs from coal surface and its hindering effect on heat accumulation gradually diminishes and eventually disappears.



(a) The temperature range 30 to 100 °C



(b) The temperature range 100 to 200 °C

Figure 5 CO production for gases of 21 % O_2 and 0 ÷ 3 % CH_4

4 Conclusion

This paper presented the test results of low-temperature spontaneous combustion of coal in the air of 10 ÷ 21 % O_2 and 0 ÷ 3 % CH_4 in an attempt to explain the observed shortened incubation period of coal spontaneous combustion in mine atmosphere. It is concluded that:

(1) Both the initial CO temperature at which CO starts to appear in coal spontaneous combustion and the CO production at the initial CO temperature increase with reduction in O_2 concentration in air. Reducing O_2 concentration in air generates less heat accumulation of coal and slows coal spontaneous combustion.

(2) The presence of low CH_4 concentration in air has a positive impact on coal spontaneous combustion. However, the impact gradually diminishes with the rise in temperature. The partial occupation of coal surface by CH_4 molecules may hinder the dissipation of heat generated through the chemisorptions and chemical reaction between O_2 and coal, resulting in more heat accumulation on coal surface and accelerate coal spontaneous combustion. With the rise in temperature more CH_4 molecules desorb from coal surface and its hindering effect on heat accumulation gradually diminishes and disappears.

(3) O_2 concentration in air has a more profound effect on coal spontaneous combustion than the presence of low CH_4 concentration in air.

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