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**STUDY ON
SUSCEPTIBILITY TO SPONTANEOUS COMBUSTION OF
ANTHRACITE IN VIETNAMESE COAL MINES**

Le Trung Tuyen

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Abstract

Regardless of the common sense that anthracite hardly starts spontaneous combustion because of few radicals which absorb oxygen, a dozen and half spontaneous combustions at several anthracite coal mines in northern coal fields in Vietnam have been reported since May 2004. Those spontaneous combustion incidents force the relevant coal mines stagnation of coal production, abandon of some parts of coal resources, long term firefighting activities, etc.

Site data such as gas analysis of the gas samples derived from inside the sealing showed different features from the cases of spontaneous combustion at bituminous or sub-bituminous coal mines. In addition it was found that the proposed methods for estimating the susceptibility to spontaneous combustion of bituminous or sub-bituminous coal cannot be applied to anthracite.

From such facts, the author and his group had started research works at the coal mine sites as well as laboratories to find out the mechanism of spontaneous combustion of anthracite coal, a suitable method for evaluating the susceptibility to spontaneous combustion of anthracite coal, key factors which give positive impacts on starting spontaneous combustion of anthracite coal and other characteristics of spontaneous combustion of anthracite coal.

Through both site study and laboratory study, the author could get some new findings relevant to the study purposes shown above as well as the author had found some more fields that need further researches to solve the problems of spontaneous combustion of anthracite coal.

This paper describes the research results on spontaneous combustion of anthracite coal and based on those results, proposes a method to evaluate the susceptibility of anthracite coal to spontaneous combustion and index gasses to detect spontaneous combustion at anthracite coal mines. In addition, unsolved matters that might relate with spontaneous combustion of anthracite coal are also reported as future research subjects.

要 約

無煙炭は、反応遊離基がほとんど無いため、ほとんど自然発火をすることはない、という従来の一般的な考えにもかかわらず、ベトナム北部の炭田の幾つかの無煙炭炭鉱では 2004 年 5 月以降、20 回近くの自然発火が報告されている。それらの自然発火の発生は、関係炭鉱に、生産の停滞、一部資源の放棄、また、長期にわたる消火活動などを強いる結果となっている。

自然発火の発生した炭鉱の密閉内から採取したガスの分析結果など現場のデータは、瀝青炭や亜瀝青炭の炭鉱の事例とは異なった様相を呈している。加えて、従来提案されている石炭の自然発火性向を評価する手法は、無煙炭には適用できないことが判明した。

以上の事実から、筆者とその研究グループは、無煙炭の自然発火について、自然発火の発生機構、無煙炭の自然発火性向の妥当な評価手法、無煙炭の自然発火発生に寄与する主要因の解明、その他、無煙炭自然発火の諸特性などについて現場研究と共に実験室における研究を実施した。

現場と実験室における研究の双方から無煙炭の自然発火に関する研究目的に関連し、新しい知見を得ることが出来たと共に、今後さらに研究を深めていく必要のある分野も明らかになってきた。

本論文は、無煙炭の自然発火に関する上記の研究結果について報告する、またそれらの結果に基づき、無煙炭の自然発火性向評価手法と無煙炭の自然発火検知の指標ガスについて提案を行う。加えて、将来の研究課題として明らかになった未解決の問題についても紹介する。

Acknowledgements

From 2004, I started my research on coal spontaneous combustion issue since we have been facing with the coal spontaneous combustions at some underground coal mines in Vietnam. Eighteen (18) time of coal spontaneous combustions have occurred at underground coal mines in Vietnam, up to date. It has been hard work to account for the problems not only for me but also for other colleagues who involved into coal spontaneous combustion in Vietnam because of our limited experiences in this field. However, our experiences as well as our knowledge have been improved to solve that problem with the help from our colleagues in the coal mine field from foreign countries. When I started to conduct my research on this field, I have received tremendous help and encourages from our side (Vinacomin, IMSAT- Institute of Mining Science and Technology) as well as my colleagues from Japan and Poland. I am so happy to have a chance to become a part of this job to take in part to solve coal spontaneous combustion issue.

I am especially grateful to my former supervisor Prof. Dr. Toyoharu NAWA who gave me a chance to comeback for study and get Ph.D. in Hokkaido University where I had got Master degree.

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Thank you.

Introduction

Until the first coal spontaneous combustion recognized at a Vietnamese coal mine in May 2004, the issue of coal spontaneous combustion has never been considered hazardous for underground coal mines in Vietnam. Therefore, occurrences of coal spontaneous combustions at underground coal mines had been considered as rather fires from other external initial causes such as electric equipment or belt conveyer.

Since May 2004, eighteen (18) spontaneous combustions have occurred up to date at Hong Thai Coal Mine, No. 91 Enterprise Coal Mine, Khanh Hoa Coal Mine, Mao Khe Coal Mine and Ha Lam Coal Mine all of which produce anthracite coal. To account for the spontaneous combustion, such coal mines have been forced to conduct long term firefighting measures, to abandon some coal resources around the area of spontaneous combustion, to change the mining plan and to cope with other problems such as safety measures against gas explosion, toxic gasses from the combustion. All those countermeasures have reduced the productivities of the relevant coal mines remarkably.

[Purposes of the Study]

To account for such spontaneous combustion, the author and his group had started research works at the coal mine sites as well as at laboratories having the main purposes shown below:

- ✓ To find out the mechanism of spontaneous combustion of anthracite coal,
- ✓ To find out a suitable method for evaluating the susceptibility of anthracite coal to spontaneous combustion,
- ✓ To find out key factors which give positive impacts on starting spontaneous combustion of anthracite coal, and,
- ✓ To find out other characteristics of spontaneous combustion of anthracite coal such as incubation period to spontaneous combustion.

The author and his group have been conducting various site studies such as site survey and gas analysis of gas samples from sealing, and laboratory studies on gas generation from heated coal samples, self-oxidation tests of coal samples, porosity measurements of coal sample and oxygen absorption of coal samples at ambient temperature. Conventional methods for evaluating the susceptibility to spontaneous combustion of coal sample have been adopted to anthracite coal samples on trial if those are applicable to anthracite coal or not.

This paper describes those study results in the following chapters:

In Chapter 1, “General Overview of Coal Industry of Vietnam” is introduced. Main topics of the brief descriptions are “Coal fields in Vietnam”, “Geological background of relevant coal fields”, “Variety of coal winning methods”, “Coal developing policy of VINACOMIN*¹”, “Coal production of Vietnam”, “Coal developing plan” and “Safety record of Vietnamese coal mines”.

In Chapter 2, “Spontaneous Combustions at Anthracite Coal Mines in Vietnam” are introduced. History of 18 time spontaneous combustions at Hong Thai Coal Mine (5 times), No. 91 Enterprise Coal Mine (4 times), Khanh Hoa Coal Mine (6 times), Mao Khe Coal Mine (1 time) and Ha Lam Coal Mine (2 times) are briefly introduced. Gas analysis results of some coal samples derived from inside the sealing walls of spontaneous combustion areas of those coal mine, it had been shown that except for methane gas (CH_4), no any higher rank hydrocarbon gasses such as C_2H_6 , C_2H_4 , C_3H_8 , had not been observed and higher concentration of hydrogen gas (maximum: $\text{H}_2 = 4.32\%$) had been observed..

In Chapter 3, “Gas Generation from Heated Coal” is discussed to compare with those site data shown in Chapter 1, heating test of anthracite coal samples has been conducted at the laboratory by supplying steady air flow through coal samples. The results showed consistency between site data and laboratory data. Higher rank hydrocarbon gasses had been observed in case of heating test of Vietnamese sub-bituminous coal.

In Chapter 4, “Conventional Methods for Evaluating the Susceptibility of Coal to Spontaneous Combustion” are discussed. By using Russian method, Japanese method and Polish method several anthracite coal samples has been examined to check if those methods are applicable or not to anthracite coal. Because those methods have been developed by using mainly bituminous or sub-bituminous, all those conventional methods showed “much less susceptibility” (Russian method), “hard to start spontaneous heating” (Japanese method) and “medium, low or too low” (Polish method) for all the subjected anthracite coal samples. Namely, it was shown that those three conventional methods cannot be applied to anthracite coal directly.

In Chapter 5, “Susceptibility of Anthracite Coal to Spontaneous Combustion” is discussed by using the data of an adiabatic oxidation tester (SHIMAZU SIT-2) to which anthracite coal samples have been subjected. When test results on self-oxidation rate of anthracite coal sample have been compared with the results of bituminous or sub-bituminous coal samples, it had been shown that self-oxidation rate of anthracite coal samples from coal mines which have spontaneous combustion showed much higher values at lower temperature range less than $150\text{ }^\circ\text{C}$

*¹ Vietnam National Coal-Mineral Industries Holding Corporation Ltd.

and much lower values at higher temperature range greater than 150 °C than those of bituminous or sub-bituminous.

In Chapter 6, “Other Effects on Susceptibility of Anthracite Coal to Spontaneous Combustion” are discussed in relation with the porosity of coal sample, the original moisture of coal sample and easiness to start self-oxidation coal samples, and the oxygen absorption capacities of anthracite coal samples, and others. Among those factors, it was found that porosity and original moisture has significant effect on the proneness to start oxidation of anthracite coal.

Effects of coal sampling method and sample storage period on the reliable laboratory data to decide the susceptibility of coal sample to spontaneous combustion are also discussed.

In Chapter 7, “Conclusion and Recommendation” are described based on the purposes of the study as well as the unsolved problems which need future study are also described.

In Annex 1, “Past researches on the mechanism of coal spontaneous combustion” including spontaneous combustion of other coals such as bituminous or sub-bituminous are introduced and discussed.

In Annex 2, “Site situations of spontaneous combustions of anthracite coal in Vietnam” are briefly introduced from both mining conditions and geological conditions.

Chapter 1: General Overview of Coal Industry in Vietnam

1.1 Introduction of Vietnamese coal industry

Vietnamese coal exploitation history ascends back more than 180 years to the French's colony time. Until the independence, Vietnamese coal industry had been small scale in both technical and coal production aspects. Coal had been produced in coal mines which were owned by local enterprises and private coal sectors. After the Innovation and Development (Doi Moi) Policy, Vietnamese coal industry has made rapid changes in its organization as well as the scale of coal production. The coal production and business in Vietnam had been shared by local enterprises and not unified until the establishment of VINACOMIN. In 1994, Vietnam National Coal-Mineral Industries Holding Corporation Ltd. (VINACOMIN) has been established based on the decision of the government¹⁻¹⁾. Based on the government policy, VINACOMIN has been one of the largest economic group companies in Vietnam which plays an important role in the government's strategies to ensure efficient and effective exploitation of mineral resources. VINACOMIN also plays important role in engineering security for the government in economic development. According to the government decision¹⁻²⁾, VINACOMIN has been fulfilling the responsibilities to ensure the national energy security. Following the decision, VINACOMIN has to cover the functions as follows:

- ✓ To enhance geological exploration for new coal fields including the Red River Delta Coal Field;
- ✓ To construct new coal mines, especially underground coal mines and to increase coal production at existing coal mines;
- ✓ To enhance mechanization in underground coal mines;
- ✓ To plan and to construct infrastructures in mining area such as coal washing plants, transportation system, etc.;
- ✓ To conduct feasibility study on the development of Red River Delta Coal Basin including underground coal gasification;
- ✓ To construct a terminal for coal import in the Southern part of Vietnam;
- ✓ To co-invest foreign coal mines so as to ensure the import of coal resource;
- ✓ To develop human resources for mining activities.

VINACOMIN is the dominant coal producer in Vietnam which is managing over 95% of total coal handling, supplying anthracite and lignite for power generation industry from both domestic and foreign coals. Beside the VINACOMIN, the Thai Nguyen Iron and Steel Corporation (TISCO) is a subsidiary of the Vietnam Steel Corporation, which organizes Phan Me underground coal mine in Thai Nguyen province that produces sub-bituminous coal.

1.2 Coal resources in Vietnam¹⁻³⁾

In Vietnam, wide range various coal distributes with geological ages from Palaeozoic Era to Cenozoic Era as shown in Table 1-1 and Figure1-1. Coal types are anthracite, semi-anthracite, bituminous and lignite; however, the major coal fields as useful resources were found in Quang Ninh Coal Field (Triassic Period in Mesozoic Era) of North-East of Vietnam, which covers 72% of geological reserves of anthracite coal.

Belt like shape Quang Ninh Coal Field distributes from west at Ha Tuyen Province (now divided in to Ha Giang Province and Tuyen Quang Province) through Thai Nguyen Province and Hon Gai to east at Cam Pha with total length around 300 km. This coal belt is divided into two groups of the southern main belt that passes through Uong Bi and Hon Gai to Cam Pha and shorter northern belt at Bao Dai-Yen Tu Graben existing in the mountainous area with the maximum height of 1,068 m. On the other hand, the main belt of Hon Gai Graben is located at the hilly area with the height 200 or 400 m, which are divided by rivers that run north to south. (See Figure 1-1).

Table 1-1: Coal distribution in Vietnam and Geological Era

Geological Time		Coal	Area
Palaeozoic era	Middle to Late Devonian	Anthracite	Binh Tri Province
	First to Middle Carboniferous	Low rank Anthracite	La Khe Suite
	Late Permian	Anthracite	Thanh Hoa Province,
Mesozoic era	Middle Triassic	Low rank coal	Thanh Hoa Province
	Late Triassic	High rank Anthracite	Quang Ninh Province
	Jurassic	Low rank coal	Phu Quoc Island
Cenozoic era	Neogene	Bituminous	Lower Red River Area
	Quaternary	Low rank coal	All over Vietnam

Coal bearing formations develop on the base rock of limestone or chart of Palaeozoic era. Coal bearing formations show differences in sedimentation due to blocking movement of base rocks and in addition, due to blocking movement after sedimentation of coal bearing formations, which show differences in their erosions. Then after due to impingement movement between the South-China Plate and Indo-China Plate, folds from east to west or from north-east to south-west and accompanied various faults were created.

Coal bearing formations in Quang Ninh Province belong to the later part of Triassic Period in Mesozoic era and has seam thickness from 1,000 to 4,500 m in which 60 coal seams are

included at the maximum. The thickness of the coal seams varies from 0.1 to 92.2 m widely and total amount of all the coal seam varies from 5 to 217 m.

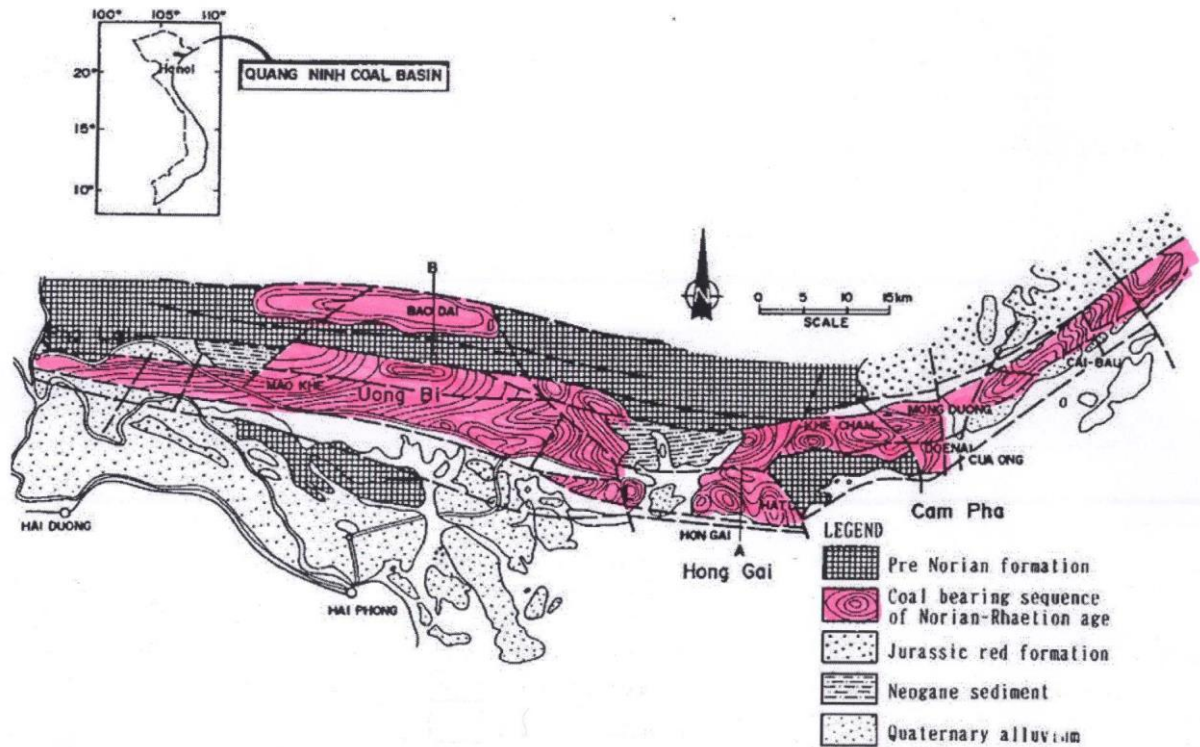


Figure 1-1: Quang Ninh Coal Field¹⁻³⁾

As shown in Figure 1-1, Quang Ninh Coal Graben is divided by several big faults that run from west to east into two coal bearing formations of the Hon Gai Graben and the Bao Dai Graben which are separated by a Palaeozoic formation upheaval belt. The Hon Gai Graben has the size of 150 km in length and 10 km in width, which is divided into Cai Bau Area, Cam Pha Area, Hon Gai Area and Uong Bi-Mao Khe Area by several big faults and the Bao Dai Graben has the size of 30 km in length and 4 or 5.5 km in width which has overall Bao Dai-Yen Tu fold anticline. In both the Hon Gai Graben and the Bao Dai Graben, most of anthracite resources are included and most of anthracite coal in Vietnam is produced from those two grabens.

The structure of coal bearing formations change depending on the coal field area. Coal seams in Uong Bi-Mao Khe area have the dip inclination of 25° or 45° toward north with the strike from west to east. At the central part of the Hon Gai Graben, the strike of the fold turns to north-south which is roughly perpendicular to the belt expansion of the Hon Gai Graben. At the Cam Pha area, folds become more complex due to small scale block movements. Inclination of coal seams is 45° or 65°.

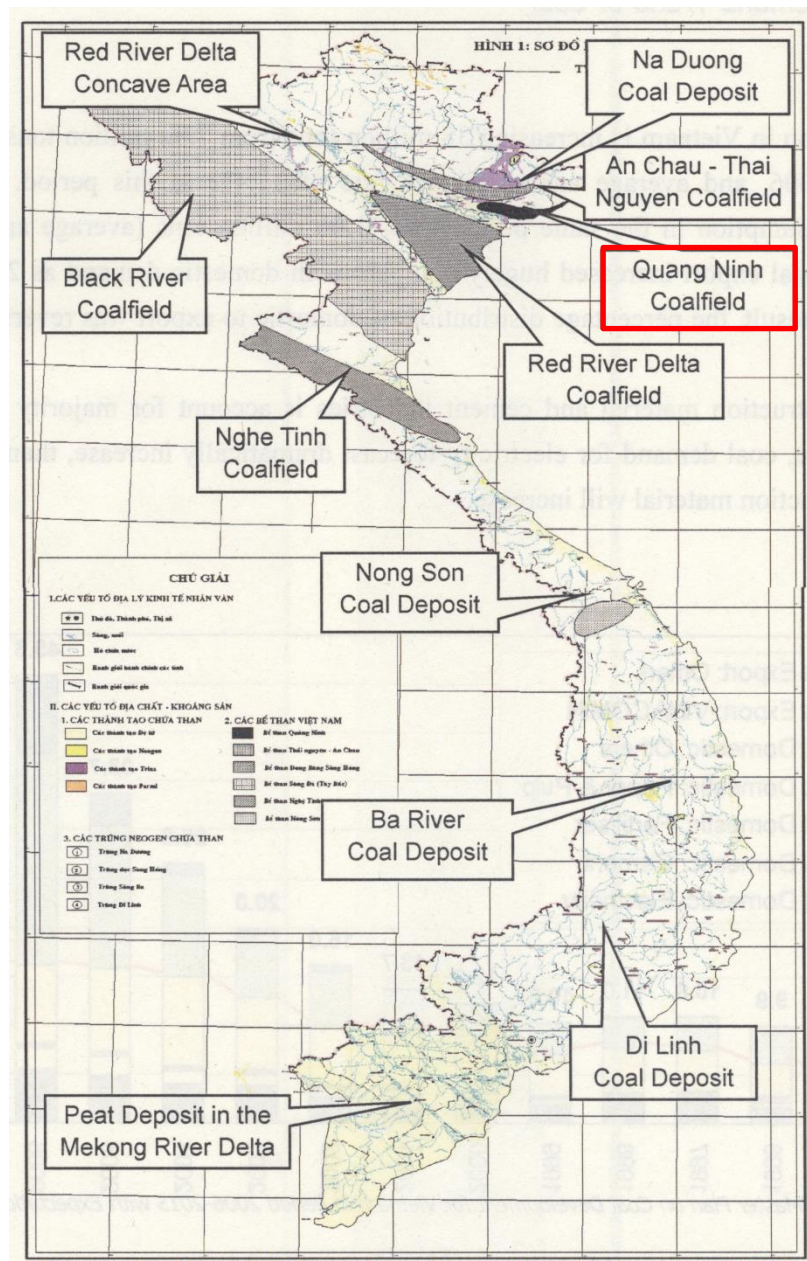


Figure 1-2: Coal fields in Vietnam

Table 1-2: Coal parameters of Quang Ninh Coal Field

Parameters	Unit	Min.	Max.		Average
Humidity	%	0.87	2.18		1 to 1.15
Ash-dry basis	%	9.73	23.64		16
Volatile matter	%	7.62	9.78		8.52
Calorific value	kcal/kg	8,120	8,685		8,300
Specific gravity	g/cm ³	1.45	1.50		1.48
Sulfur	%	0.29	0.36		0.32

1.3 Major coal winning methods in Vietnam¹⁻⁴⁾

1.3.1 Preparing schema and opening coal seam

As mentioned before, most coal resources in Quang Ninh area are located at natural hilly terrains which are convenient for preparing of coal winning by cross-cut roadways from the mountainside or hillside surface at an initial phase. Lately, when the underground coal mine goes deeper area, coal resources are developed by vertical shafts, incline shafts and cross-cut roadways.

The level layout is the most important, which has been applied at Mao Khe, Thong Nhat and Khe Cham coal mines that are featured with great rise areas and divided into separated blocks. Thus, the layout consist of two directions: the level for major independence transportation level and level for dividing into sub levels. The coal developing concept at Mao Khe Coal Mine or Hong Thai Coal Mine is shown in Figure 1-3.

The block (sub-panel) development method for a steeper coal seam is also applied at some coal mines, in which exploiting areas are developed by sub-level galleries as if multiple coal seams. The coal seam in the block is developed separately by rise galleries dug from the transport roadway up to ventilation roadway. These blocks are divided into sublevel by roadway in seam drifts toward two side of the rise. The exploiting areas are also prepared according to the schema of sub level coal faces by the in-seam roadway drift systems above. The series of coal seams or levels in the seam are separately developed.

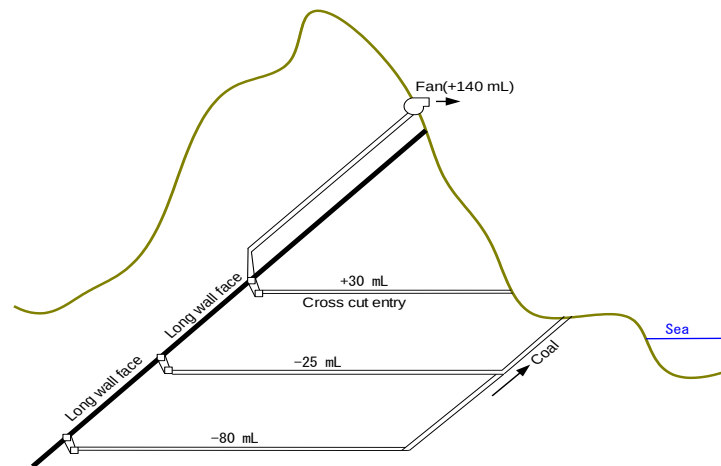


Figure 1-3: Coal developing concept at Mao Khe Coal Mine or Hong Thai Coal Mine

1.3.2 Method of coal winning

The geological conditions of coal seam in Quang Ninh Coal Field vary with their dip angle, coal seam thickness and coal quality itself. Therefore, choice of a coal wining method is not fully satisfied with all requirements for efficient development as well as for safety as well. From the viewpoints of coal spontaneous combustion itself, the selection of the coal wining method must satisfy the requirements for preventing coal spontaneous combustion. Recently, Vietnamese coal

mine adopted several main coal winning methods. Taking account for the coal seam conditions, coal winning schemas have mainly been used at Quang Ninh Coal Field as shown below.

(1) Long wall panel that exploits full coal seam thickness by drilling and blasting method

In this method, coal is extracted by drilling and blasting, long wall is supported by hydraulic single props or sliding beams. The upper rock layers are fully caved. This method is applied to coal seams that have a thickness around 3 m or less with a gentle slope. Blasting bore holes are arranged at the floor and roof of the coal seam. The residual coal ratio inside the goaf area is higher because the coal at floor part is not fully extracted (Figure 1-4).

(2) Long wall panel that exploits the coal by a coal cutting machine

The coal winning schema of this method is basically same as the “Long wall panel which exploits full coal seam thickness by drilling and blasting method (Figure 1-4)” shown above except for that coal is cut by a coal cutting machine (Figure 1-5).

(3) Long wall panel which slices the bottom part of the coal seam by drilling and blasting method with caving upper part of the coal seam

This is a method applied for a thicker coal seam that cannot be covered by the height of available support systems. The bottom part of coal seams is exploited by a long wall coal face by using a coal cutting machine same as (2), but the upper part coal seam is recovered from the backward support system from the caved parts (Figure 1-6).

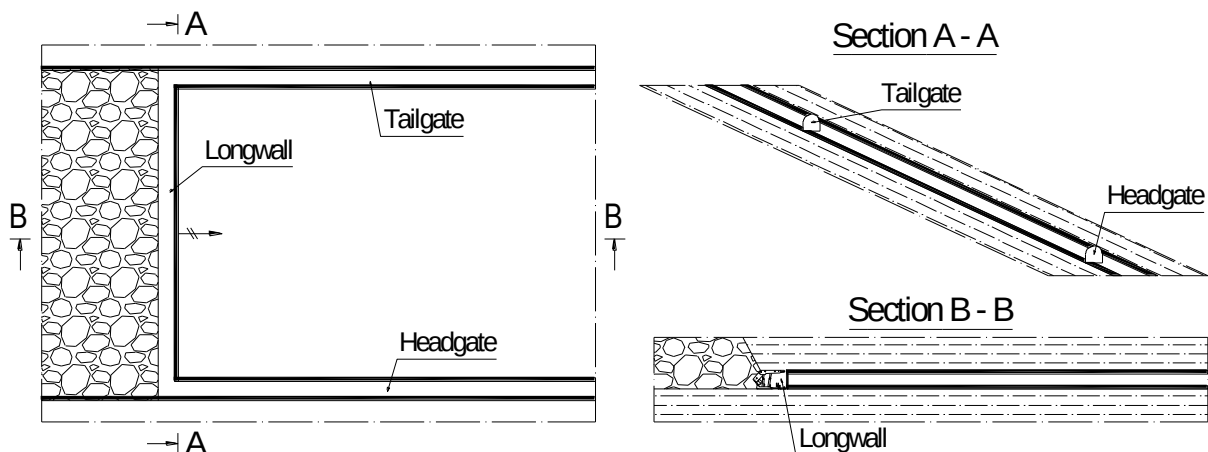


Figure 1-4: Coal winning schema of a long wall face with drilling and blasting

(4) Long wall panel that exploits coal by a coal cutting machine with supports of self-advancing with upper part coal caving

This is a quite new mining method that is being applied at 4r biggest underground coal mines in Quang Ninh Coal Basin.

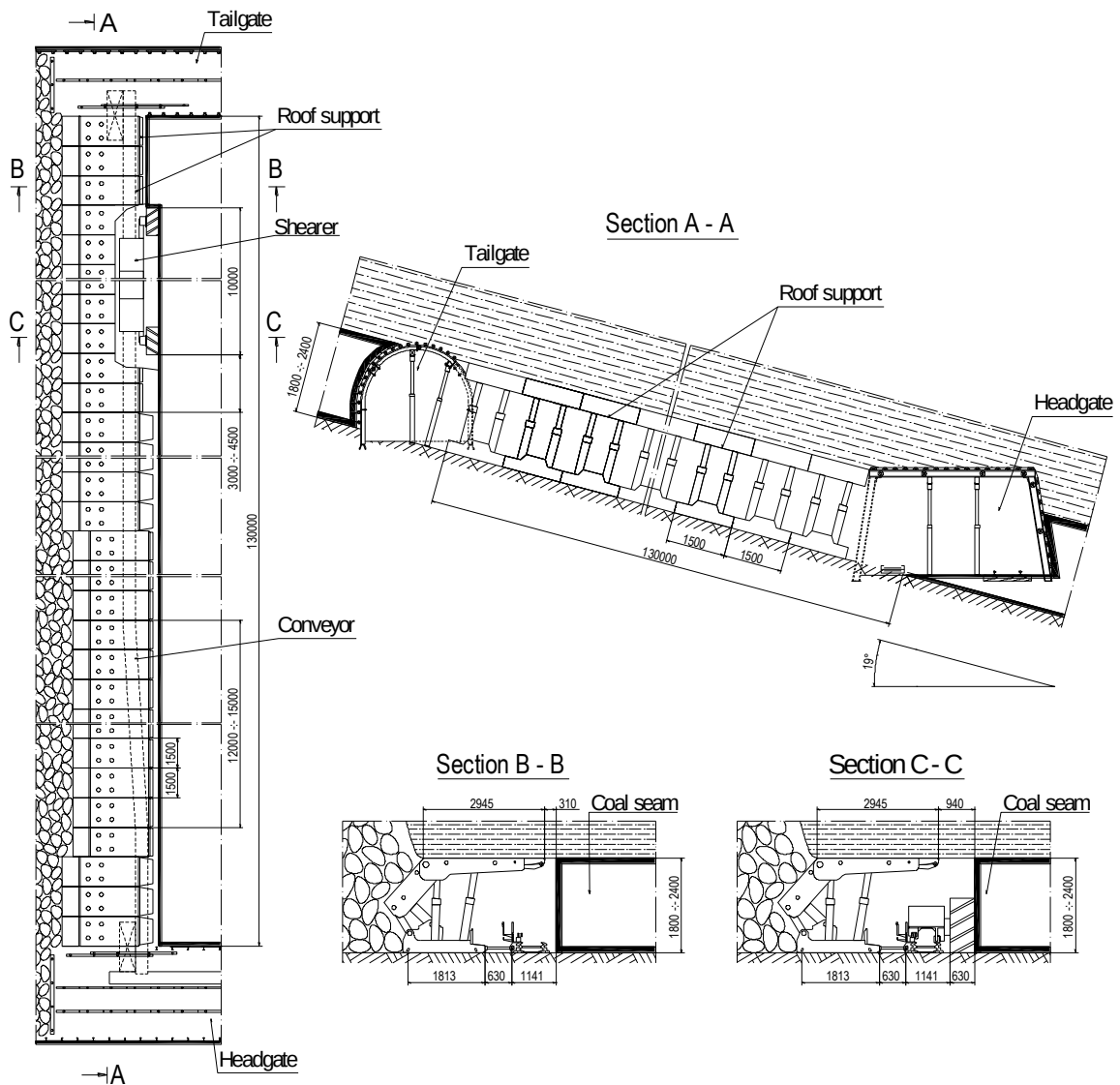


Figure 1-5: Coal winning schema with a coal cutting machine

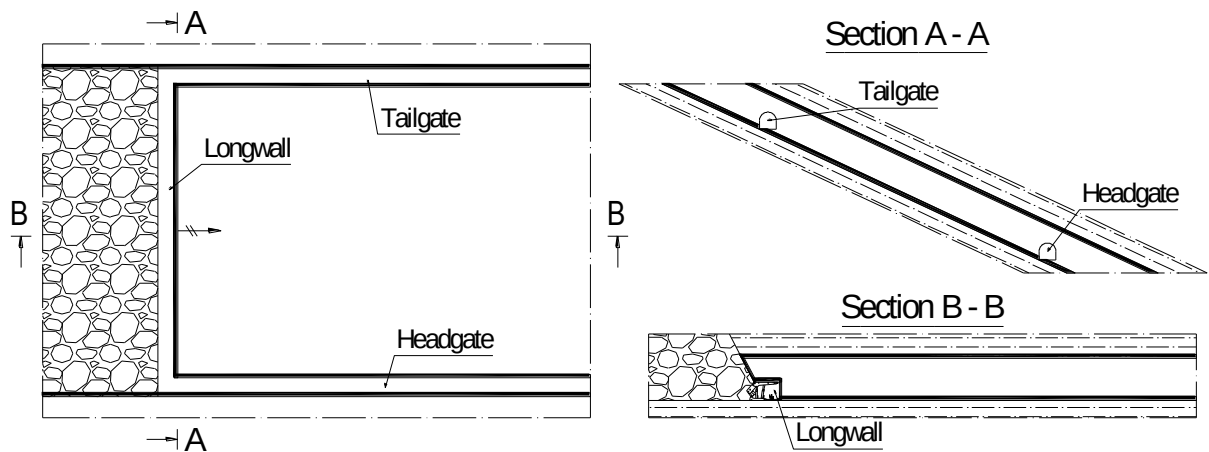


Figure 1-6: Coal winning schema of a long wall face and backward caving system

The system includes a coal cutting machine and the system of self-advancing supports that have two belt conveyors located at both front and behind the support system. Generally, this system is applicable for the coal seam slope of up to 30°, i. e. gentle and medium slopes. However, for slopes greater than 30°, several measures must be taken and the efficiency decreases significantly. Up to now, Vang Danh Coal Mine, Khe Cham Coal Mine, Quang Hanh Coal Mine and Ha Lam Coal Mine have been using this system at coal seam that have gentle slope and thick to very thick coal seams sometimes up to 20 m (Figure 1-7).

As mentioned above, the geological conditions of coal seam have strong effects on the efficiency of this method. At many long wall faces, the mining machine have to be moved sometime because a hard rock mass appears in the range of the long wall coal face. The advancing speed of the long wall becomes slower than expected. Also, the ratio of coal recovery behind the support system is not so high as the long wall coal producing plan. From two reasons above, prevention of coal spontaneous combustion in the goaf area for the mechanized method should be considered.

(5) Slicing mining method for a thick coal seam

A coal seam is divided into slices depending on the kind of the frame system and mining method. The mining method in each layer is the same as (1). The upper layer and lower layer can be exploited at the same time with the distance between two long-walls at each layer is 25 - 30 m (Figure 8).

(6) Long wall panel with a long wall face perpendicular to the strike

Most of above mentioned coal wining panels are set to develop the coal toward the strike direction of the coal seam including some false slant development. This schema is designed to develop the coal seam toward the dip direction of the coal seam (Figure 9).

(7) Sub-level stoping for a thicker and higher inclination coal seam

Recently, this method is widely applied to almost coal mines in Quang Ninh area that is suitable for a coal seam with thickness of 2-5 m and slope greater than 45°. When this method is applied, the coal seam is divided into blocks that have a strike length of 100m. A coal panel is divided into blocks with the strike length from 80 to 100 m. This block is divided into the sub-level by driving in seam roadways (sub-level roadway). The distance between those sub-level roadways is from 6 or 10 m, which depends on the coal seam characteristics of weakness enough for falling down. Coal is extracted by using a blasting method at roof part of the sub-level roadway. The depth of blasting boreholes is 3 m each. Coal extracting area is supported by using square-timber frame chocks or hydraulic support system. The ratio of coal residual inside the goaf area is around 25 or 35 % (Figure 1-10).

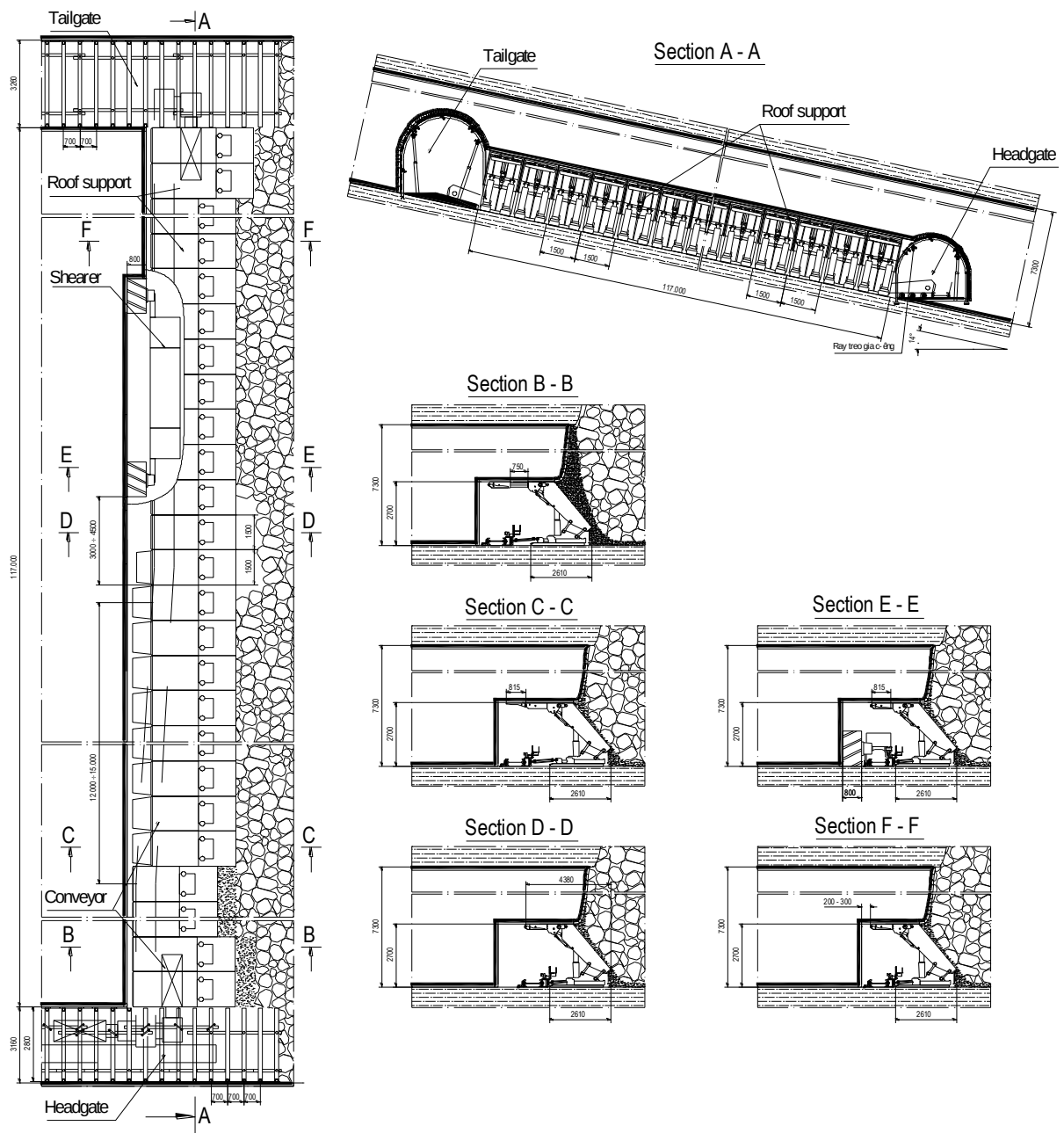


Figure 1-7: Coal winning schema for thicker coal seam

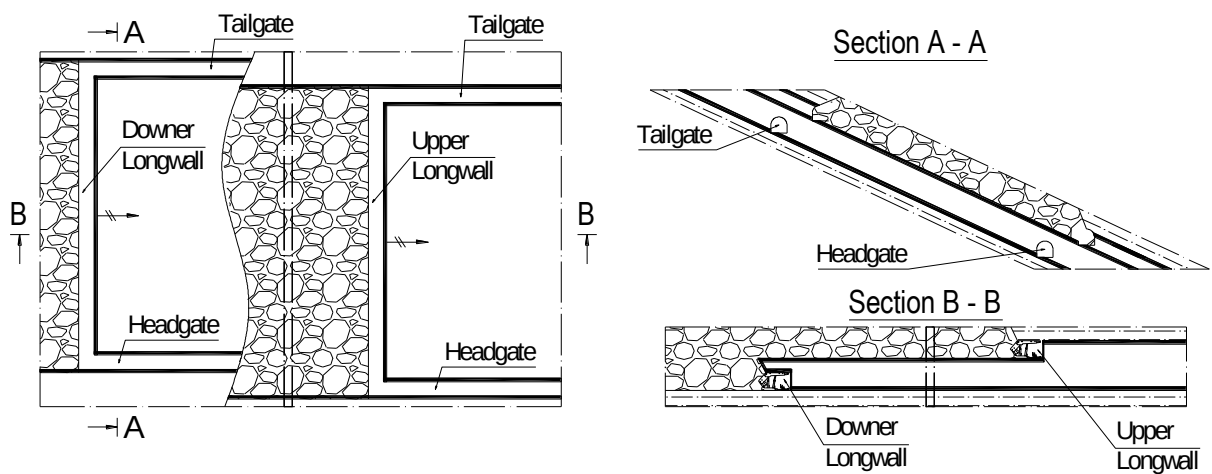


Figure 1-8: Coal winning schema of slicing long wall faces

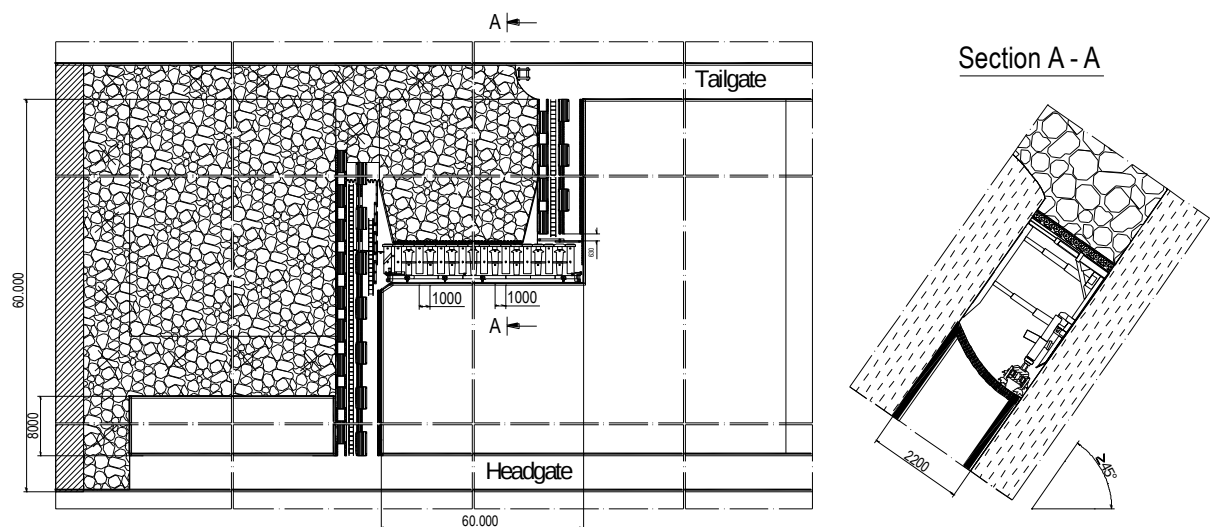


Figure 1-9: Coal winning schema of a long wall face which is perpendicular to the strike

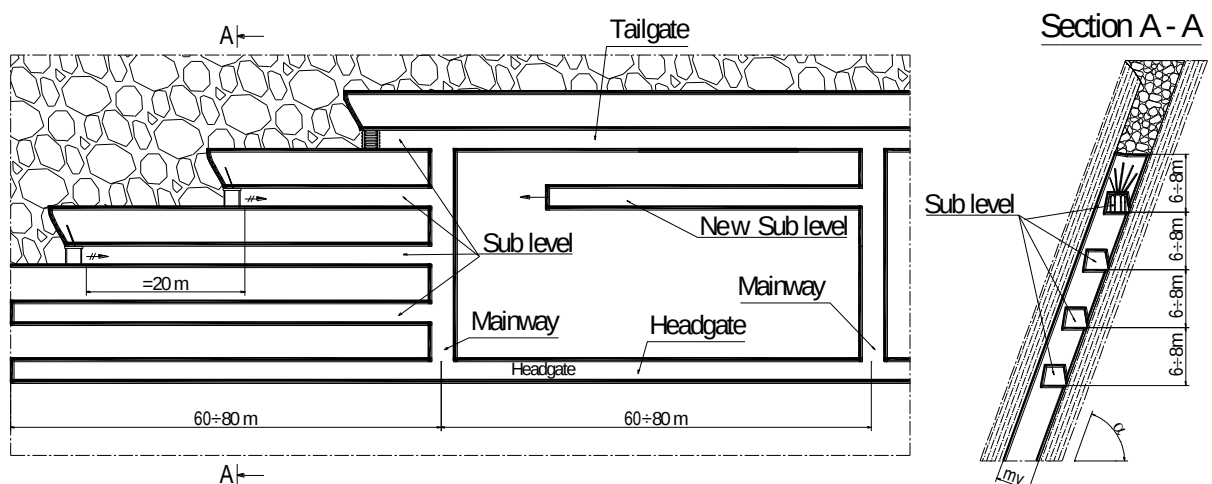


Figure 1-10: Coal winning schema of sub-level stopping

1.4 Coal production in Vietnam

Recently, in Quang Ninh Coal Field, there are around 20 open pit coal mines and 30 underground coal mines, in which top five open pit coal mines produce more than two million tons per year and top ten underground coal mines produce more than one million tons per year. According to the annual report ¹⁻⁵⁾, total coal production is summarized in Table 1-3.

Table 1-3: Coal production in Quang Ninh Coal Field from 2003 to 2017

Year	Open pit		Underground		Total (million ton)
	Production (million ton)	Ratio (%)	Production (million ton)	Ratio (%)	
2003	12.4	64.9	6.7	35.1	19.1
2004	16.2	62.9	9.6	37.1	25.8
2005	20.6	63.0	12.1	37.0	32.8
2006	24.4	63.1	14.3	36.9	38.7
2007	25.0	61.4	15.7	38.6	40.7
2008	23.6	57.8	17.2	42.2	40.8
2009	23.8	56.9	18.0	43.1	41.7
2010	24.6	54.8	20.3	45.2	44.9
2011	24.2	53.4	21.1	46.6	45.4
2012	21.2	51.4	20.1	48.6	41.3
2013	18.8	46.0	22.1	54.0	40.9
2014	19.8	52.0	18.3	48.0	38.1
2015	17.5	43.4	22.8	56.6	40.3
2016	18.5	42.2	25.3	57.8	43.8
2017	20.8	44.2	26.3	55.8	47.1

As shown in Table 1-3, coal production in Quang Ninh Coal Field has been rapidly increased by around 2.4 times from around 19 million tons in 2003 to 47 million tons in 2017 during around one and half decade. The ratio of coal production from underground coal mines has been increased from 35 % in 2003 to 56 % in 2017, some of which have problems of spontaneous combustion. During the years of 2014, 2015 and 2016 the total coal production has been slightly reduced; however, the ratio of coal production from underground coal mines has been increased in spite of reduction of total coal production. According to the Master Plan of Coal Production¹⁻⁶⁾, the requirement of coal for economic development would be increased to 60-65 million tons in 2020, 66-70 million tons in 2025 and 75 million tons in 2030. The plan of coal production up to 2030 in Quang Ninh Coal Field is shown in Figure 1-13.

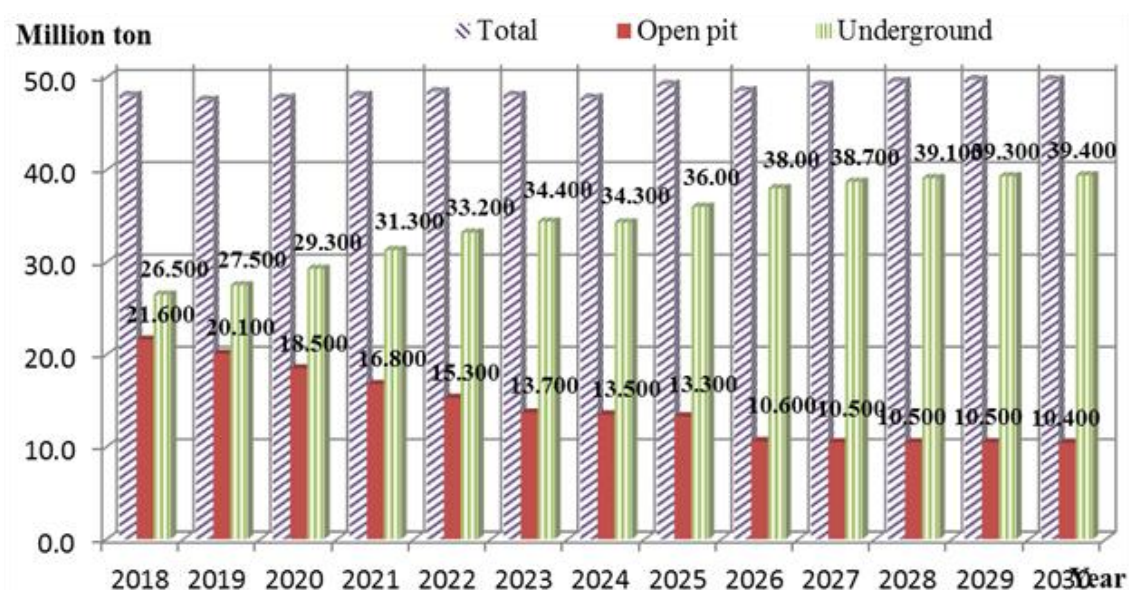


Figure 1-13: Forecast of coal production up to 2030

This figure shows an increase trend of coal production in Quang Ninh Coal Field. However, in comparison with the requirements of the Master Plan, the coal production in Quang Ninh Coal Field might not be sufficient for the requirement of coal for economic development. However, the Quang Ninh Coal Field is still the main coal resources of Vietnam coal production. The ratio of coal from the underground coal mine would be increased from 56% in 2018 (26.5/48.1) to 80% in 2030 (39.4/49.8).

1.5 Trend of coal mine safety

Increase of coal production as well as increase of the ratio of underground coal production tends to increase accidents or increase victims due to accidents. Coal production, fatality number and fatality rate per one million tons of coal production from 1995 to 2016 are summarized in Table 1-4¹⁻⁷⁾. Up to around 2010, the fatality rate per one million tons of coal production stays around 1.0 or sometimes higher than one and then after it was reduced to around 0.5. From such a trend, the safety record in coal mine has been improved recently.

In addition to the statistical data of coal mine safety, serious big accidents have to be overcome to improve the safety records. Major serious accidents in underground coal mines in Vietnam are shown in Table 1-5, from which it is clear that gas explosions are the main causes of serious accidents in Vietnamese coal mine¹⁻⁸⁾.

If we compare Table 1-4 with Table 1-5, it becomes clear that the reason of prominence of the value of fatality per one million tons of coal production such as 1999 (2.9) in the period with less coal production. Fortunately, there is no fatality and injured due to outbreak of spontaneous combustion up to date.

Table 1-4: Coal production and fatality in coal mines of VINACOMIN

Year	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005
Production (million ton)	10.9	12.5	14.1	14.4	12.1	13.9	15.8	19.9	19.1	25.8	32.8
Fatality	9	20	26	11	35	18	15	38	16	25	37
Fatality/one million ton	0.8	1.6	1.8	0.8	2.9	1.3	0.9	1.9	0.8	1.0	1.1
Year	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016
Production (million ton)	38.8	40.7	40.8	41.8	45.0	45.4	41.3	41.0	38.1	40.3	43.8
Fatality	50	36	34	29	35	18	29	23	17	20	25
Fatality/one million ton	1.3	0.9	0.8	0.7	0.8	0.4	0.7	0.6	0.4	0.5	0.6

Table 1-5: Major coal mine accidents in Vietnam

Date	Coal Mine	Accident	Fatality
11-Jan-99	Mao Khe	Gas explosion	19
6-Mar-06	Thong Nhat	Gas explosion	8
8-Dec-08	Khe Cham	Gas explosion	11
31-Jul-13	Dong Vong	Oxygen deficiency	3
15-Jan-14	Dong Vong	Underground fire	6

Reference

- 1-1) Decision No.563/1994/ TTg of the Prime Minister of Vietnam.
- 1-2) Decision No.345/2005/TTg of the Prime Minister of Vietnam.
- 1-3) 財団法人石炭エネルギーセンター、「世界の炭田概要」、1999 年 3 月、pp. 411 – 435 [Japan Coal Energy Center, “Outline of World Coal Fields”, March 1999, pp. 411 - 435]
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Chapter 2: Spontaneous Combustion at Anthracite Coal Mines in Vietnam²⁻¹⁾

2.1 History of Coal Spontaneous Combustion in Vietnam

Different from other coal production countries, reports of coal spontaneous combustion in Vietnamese coal mines were not so many up to the middle of 2000s, which might be thought to support the conventional idea that the susceptibility of anthracite coal to spontaneous combustion is very low. As shown in the previous Chapter, most coal produced in Vietnam, especially in Quang Ninh is anthracite coal. Besides that, coal in Vietnam is mostly produced from open pit coal mines in the past time. The cause of coal fire, therefore, is considered from other factors, such as forest fire or human activity sources.

Actually, some several coal spontaneous combustions had occurred at coal mines in other area of northern part of Vietnam since 1970s at Khe Bo Coal Mine (Nghe An province, sub-bituminous), Phan Me Coal Mine and Lang Cam Coal Mine (Thai Nguyen province, sub-bituminous). However, the causes and solutions for those cases were not described from the view point of the coal susceptibility to spontaneous combustion. Besides that, the research on coal spontaneous combustion in Vietnamese coal has not been conducted not so much as the inherent opinion that anthracite coal has much less proneness to spontaneous combustion.

Some causes of spontaneous combustion have been considered such as volatile content or pyrite content in coal. The causes of those coal fires have been considered from external factors such as methane explosion, use of explosives in coal seam, belt conveyer fire, electrical cable or heating of electrical equipment, etc.

Recently, VINACOMIN has announced eighteen (18) coal mine fires at five underground coal mines, which have been considered as coal spontaneous combustion at Ha Lam, Mao Khe, Hong Thai, No. 91 Enterprise and Khanh Hoa coal mines. In addition, Phan Me coal mine has also been facing problems of spontaneous combustion. Former four coal mines all produce anthracite are located in the western part of Quang Ninh Coal Basin. Two other latter coal mines Khanh Hoa and Phan Me are located in Thai Nguyen province. Different from the former five coal mines, Phan Me Coal Mine produces sub-bituminous. At the east part of Quang Ninh Coal Basin, no outbreak of spontaneous combustion has been reported up to date. The locations of these mines are shown in Figures 2-1-1 and 2-1-2. A history of coal spontaneous combustions in Vietnamese anthracite coal mines are summarized in Table 2-1. The first outbreak of spontaneous combustion in Vietnamese anthracite coal mines was in May 2004. In the case of the sub-bituminous Phan Me Coal Mine (fuel ratio: around 2.3), there might be spontaneous combustions earlier than that period; however, detailed information is not available because that coal mine belongs to Thai Nguyen Steel Company that does not belong to VINACOMIN.

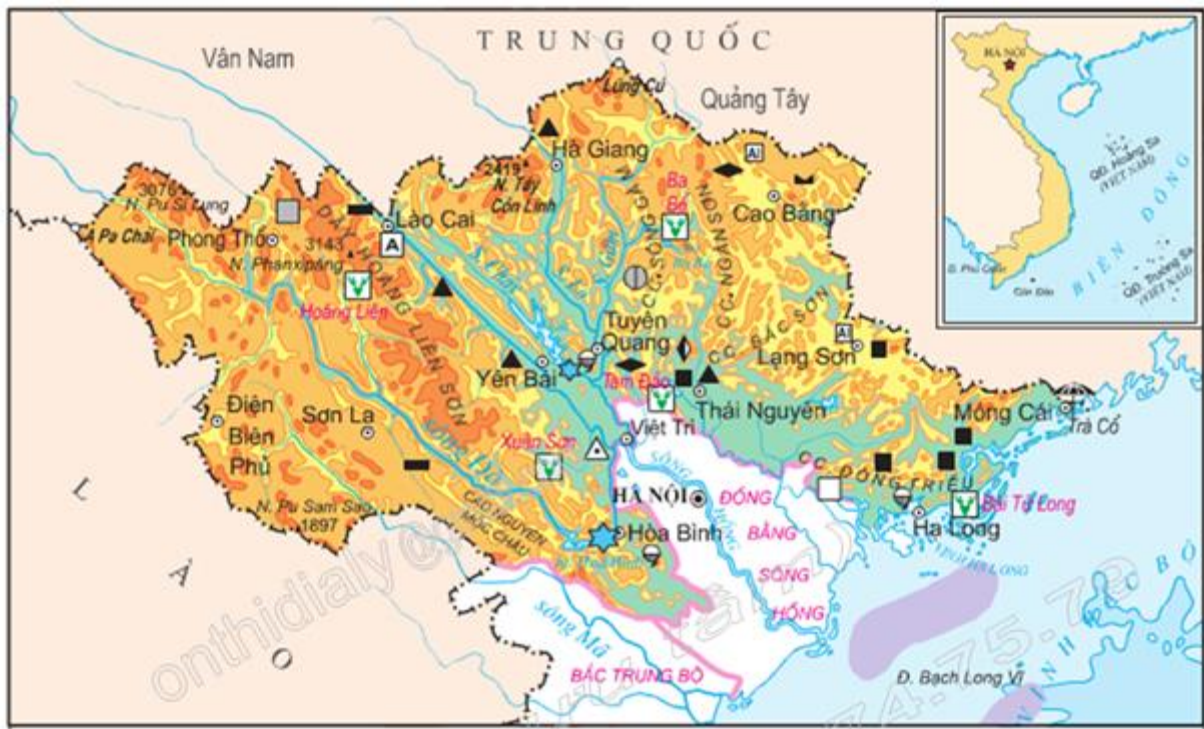


Figure 2-1-1: Coal Mines in Northern Part of Vietnam including Quang Ninh Coal Field

■: Anthracite ▲: Sub-bituminous

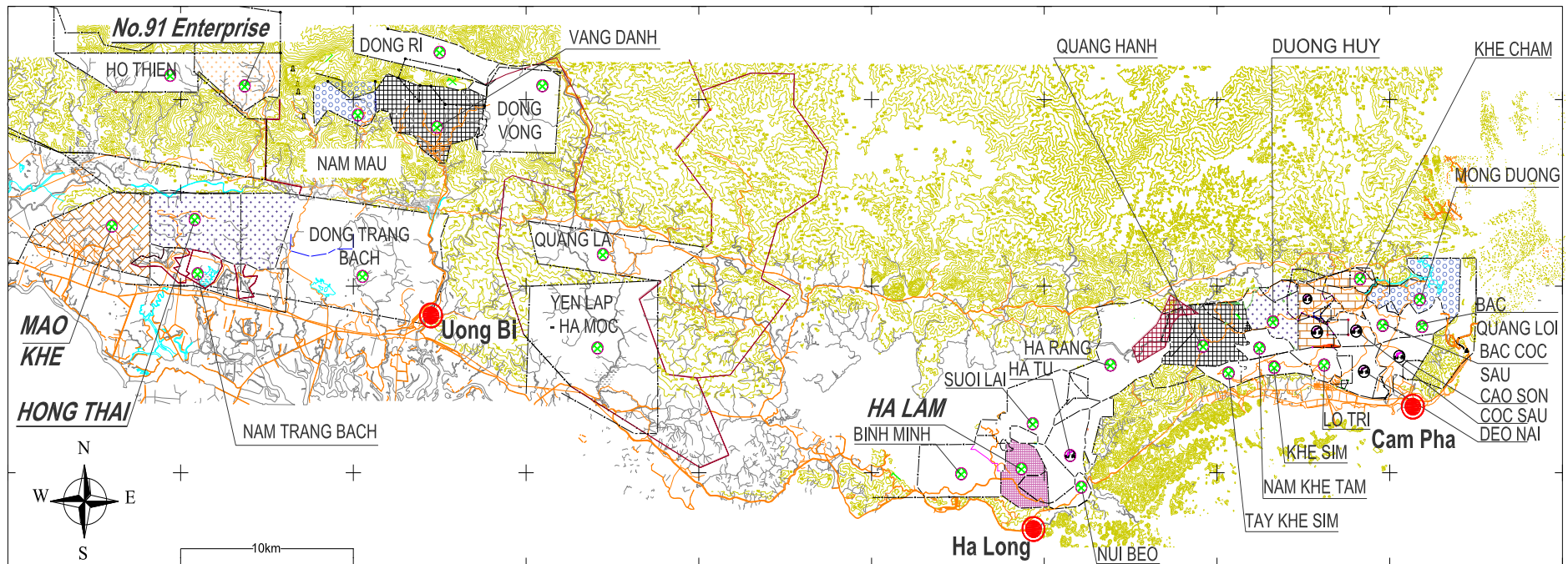


Figure 2-1-2: Location of major anthracite coal mines in Quang Ninh Coal Field of Vietnam

---- Coal mines shown by *capital italic* letter appear in this paper and small character are other anthracite coal mines.

Table 2-1: History of spontaneous combustions in Vietnamese anthracite coal mines

Coal Mine	Coal Seam	Date of outbreak	Remarks
Hong Thai*	No. 24	(1)* May 24, 2004	Fire started at a starting part of the long-wall coal face panel. [Mining method: (1) (see Chapter 1)]
		(2)* May 16, 2005	Fire started after two months since the goaf exposed to air leakage from the gate road. [Mining method: (1) (see Chapter 1)]
		(3)* June 4, 2005	
		(4)* May, 2009	Fire started after two months since the long-wall with hydraulic frame started. [Mining method: (1) (see Chapter 1)]
	No. 12	(5) September 3, 2013	Fire started after one month since a rock entry penetrated the Coal Seam No. 12 in Trang Khe Area.
No. 91 Enterprise	No. 5	(1) Jan, 2007	Fire started in the goaf of Coal Seam No. 5 into which ventilation air might leak in. [Mining method: (1) (see Chapter 1)]
		(2) August, 2007	
		(3) May, 2008	
	No. 4	(4) June 2012	Fire started after two weeks since a rock entry penetrated the Coal Seam No. 4.
Khanh Hoa**	No. 16	(1) May 4, 2011	Fire started within one month after coal exposed to the air at caving place along a gallery in the same Coal Seam No. 16. [Mining method: (70 (see Chapter 1)]
		(2) May 29, 2011	
		(3) March 2012	
		(4) August , 2017	New water drainage in-seam road way (-183 m level)
		(5) August, 2017	
		(6) September 23, 2017	
Mao Khe	No. 10	(1) Jan 13, 2017	Fire started at the goaf of sub-level roadways [Mining method: (7) (see Chapter 1)]
Ha Lam**	No. 10	(1) June 03, 2017	Fire started at the coal pillar between the goaf area and ventilation roadway
	No. 7	(2) September 14, 2017	Fire started in the goaf of the Coal Seam No. 7 into which ventilation air might leak in. [Mining method: (6) (see Chapter 1)]

* (1), (2), (3) and (4) correspond to No. 1, No. 2, No.3 and No.4 in Figure 4, respectively.

** All the galleries are excavated inside the coal seam including permanent galleries.

As a countermeasure for suppressing such coal spontaneous combustions, the author has suggested the relevant coal mines to install sealing walls at all the intake and exhaust galleries so as to cut air supply into the area of spontaneous combustion. An example layout of a mining panel

of Hong Thai Coal Mine where spontaneous combustion occurred in May 2004 is shown in Figure 2-2 and the picture of sealing No.1 is shown in Figure 2-3. And, we had monitored the situations inside the sealing walls by analyzing gas, measuring difference in air pressure both sealing sides, and monitoring air leakage through the sealing wall by smoke tubes as shown in Figure 2-3. Four locations of spontaneous combustions in Coal Seam No. 24 of Hong Thai Coal Mine are shown in Figure 2-4. As shown in Table 2-1, this Coal Seam No. 24 had three spontaneous combustions in one year from May 2004 to June 2005 and another one in 2009, namely from these facts, this coal seam must have a higher susceptibility to spontaneous combustion.

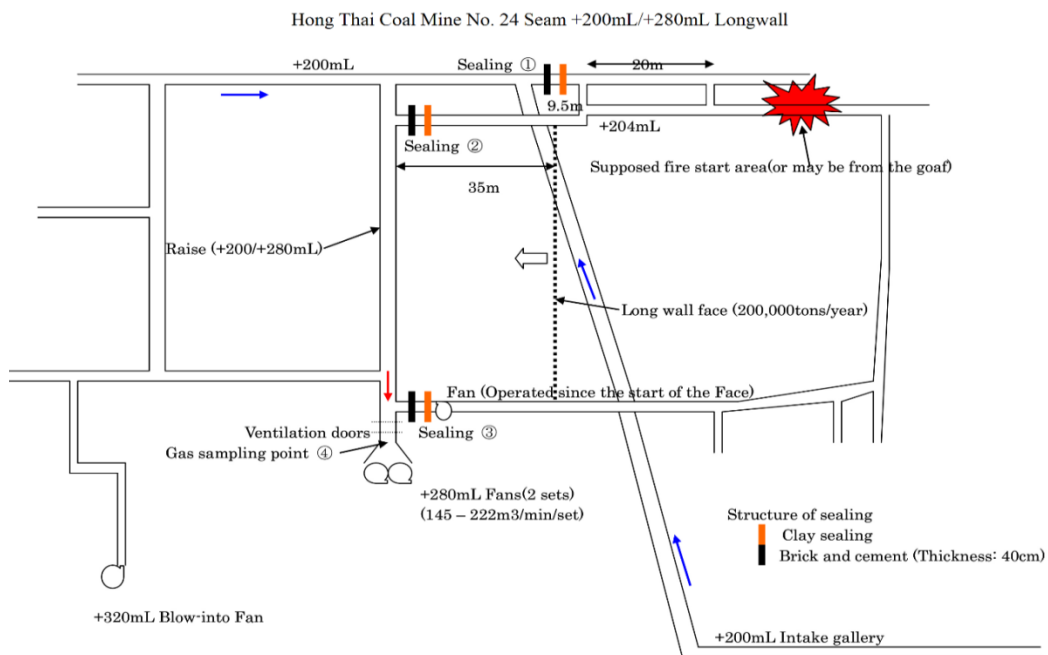


Figure 2-2: Schematic view of sealing walls of the mining panel of Hong Thai Coal Mine for the first spontaneous combustion in May 2004 (No. 1 in Figure 2-4)



Figure 2-3: Sealing wall No.1 in the mining panel shown in Figure 2-2.

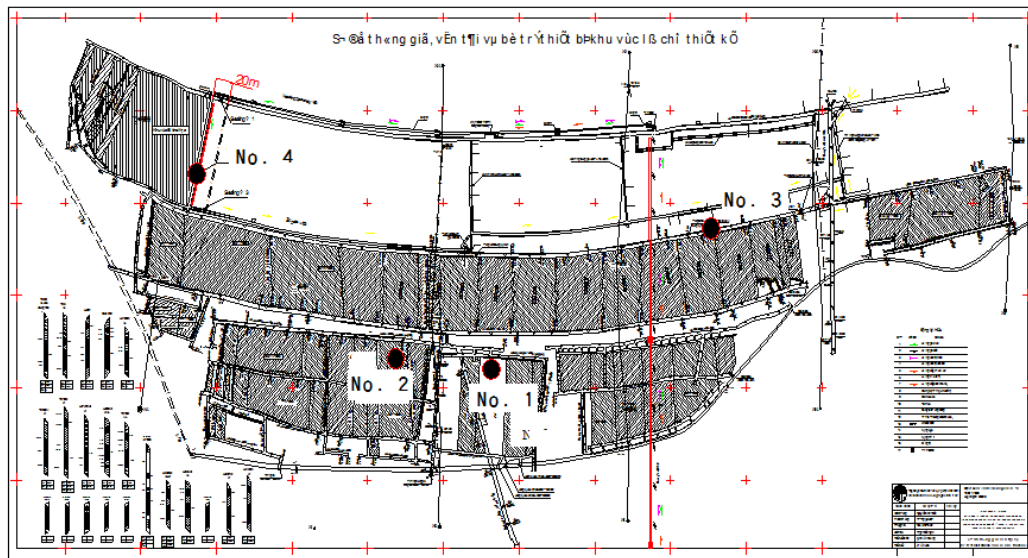


Figure 2-4: Locations of four spontaneous combustions in Coal Seam No. 24 of Hong Thai Coal Mine



Figure 2-5: Narrowing the return gate roadway due to floor heave at Hong Thai Coal Mine

2.2 Characteristics of Spontaneous Combustion of Anthracite in Vietnam^{2-1), 2-2)}

In cases of coal spontaneous combustions at Hong Thai (including Trang Khe Area) and Khanh Hoa coal mines shown in Table 2-1, gas samples were taken from inside the sealing walls by using a vacuum pump and analyzed by gas chromatographs. The gas chromatographs with two detectors of FID (flame ion detector) and TCD (thermal conductivity detector) were used, which can detect H_2 , O_2 , N_2 , CO , CO_2 and hydrocarbon gasses (C_1 - C_4) at concentration level of ppm or less (ppb). Beside the gasses of CH_4 , CO , CO_2 and H_2 , no other hydrocarbon gas was detected in those anthracite coal mines in Vietnam in contrast with spontaneous combustion of bituminous coal mines in other countries.²⁻²⁾ Some extraction gas analysis results of the cases in Coal Seam

No. 24 of Hong Thai Coal Mine in May 2004 and Coal Seam No.12 in Trang Khe Area of Hong Thai Coal Mine in September 2013 are shown in the Table 2-2, and Table 2-3, respectively. As listed in Table 2-1, these two spontaneous combustions occurred in different coal seams of No. 24 and No. 12, respectively.

Table 2-2: Gas analysis results inside the sealing No. 3 of Hong Thai Coal Mine

Date (2004)	Gas concentration (%)				
	O ₂	CH ₄	CO ₂	CO	H ₂
1-Jun	6.00	0.40	9.89	1.09	2.40
2-Jun	7.70	0.33	9.82	0.61	1.58
3-Jun	11.29	0.52	6.03	0.61	1.06
4-Jun	5.01	0.35	12.47	0.44	0.31
5-Jun	18.08	0.10	0.85	0.11	0.09
6-Jun	15.02	0.12	NA	0.11	0.08

Table 2-3: Gas analysis results inside the sealing of Trang Khe Area of Hong Thai Coal Mine

Date (2013)	Gas concentration (%)				
	O ₂	CH ₄	CO ₂	CO	H ₂
4-Sept	15.88	0.20	1.72	0.02	0.14
5-Sept	9.85	0.06	2.82	0.30	0.20
6-Sept	9.63	0.04	2.33	0.19	0.10
7-Sept	9.36	0.02	1.66	0.15	0.07

In the cases of coal spontaneous combustion at Ha Lam Coal Mine (coal seam No. 10 and coal seam No. 7) and Mao Khe Coal Mine (coal seam No. 10), the gas analysis results are shown in Table 2-4, Table 2-5, and Table 2-6, respectively.

Table 2-4: Gas analysis results inside the sealing No. 5 - coal seam No. 10 of Ha Lam Coal Mine²⁻³⁾

Date (2017)	Gas concentration (%)					
	O ₂	CH ₄	CO ₂	CO	H ₂	N ₂
4-June (Before N ₂ injection)	16.41	0.15	3.72	0.18	0.20	79.87
5-June	9.75	0.31	8.42	0.22	0.32	81.23
6-June	8.34	0.38	8.32	0.29	0.48	81.77
7-June	8.06	0.38	8.40	0.20	0.21	81.67
8-June	7.59	0.33	8.09	0.082	0.042	83.59
9-June	7.10	0.30	8.35	0.029	0.0084	83.86
10-June	8.67	0.23	7.50	0.0066	0.0015	83.57
11-June	8.48	0.19	7.41	0.0021	0.0002	83.63
12-June	7.35	0.20	8.80	0.0076	0.0006	82.27

Table 2-5: Gas analysis results inside the sealing area - coal seam No.7 of Ha Lam Coal Mine²⁻³⁾

Date (2017)	Gas concentration (%)					
	O ₂	CH ₄	CO ₂	CO	H ₂	N ₂
14-Sept (Before N ₂ injection)	17.81	0.17	2.76	0.67	0.38	77.56
9-Oct	1.11	29.15	3.36	0.0051	0.011	66.04
13-Oct	2.41	26.45	2.53	0.0022	0.010	68.29
18-Oct	1.75	29.23	3.00	-	0.0097	65.52
25-Oct	2.59	15.04	3.53	0.0008	0.014	78.72
8-Oct	1.22	11.69	3.69	0.0005	0.017	82.74

Table 2-6: Gas analysis results inside the sealing at -14 m level in coal seam No. 10 of Mao Khe Coal Mine²⁻⁴⁾

Date (2017)	Gas concentration (%)					
	O ₂	CH ₄	CO ₂	CO	H ₂	N ₂
19-Jan	14.89	5.53	1.33	0.14	0.15	77.92
20-Jan	12.90	6.97	1.97	0.24	0.32	77.29
21-Jan	11.28	8.75	2.18	0.18	0.25	76.63
22-Jan	10.42	9.54	2.53	0.17	0.21	76.33
23-Jan	10.87	8.75	2.82	0.092	0.087	77.23
2-Fer	9.62	7.06	4.47	0.24	0.064	78.04
4-Fer	3.31	9.99	7.89	2.79	2.79	72.68
6-Fer	1.39	9.34	11.62	4.86	4.32	68.21
Start of injection of N ₂						
13 - Fer	0.91	18.04	9.83	0.45	0.80	72.17
21 - Fer	0.32	30.98	9.33	0.012	0.0078	60.62
24 - Fer	0.42	31.04	8.80	0.0064	0.0050	60.16
7 - March	0.89	29.05	7.49	0.0046	0.0048	63.18
8 - March	1.55	26.90	6.92	0.0055	0.0050	65.26
9 - March	1.70	27.36	7.13	0.0058	0.0055	63.08
10 - March	1.10	28.43	7.27	0.0055	0.0048	63.40
11 - March	0.95	29.35	7.30	0.0035	0.0050	63.35

In case of Hong Thai Coal Mine in May 2004, the combustion area was isolated from roadway ventilation system by installing three sealing walls of No. 1, No. 2 and No. 3 of which No. 1 and No. 2 were installed in the intake galleries and No. 3 was installed in the exhaust gallery as shown in Figure 2-2. From Table 2-2, higher concentration H₂ and CO gasses without hydrocarbon gasses except for the lowest rank CH₄ gas were detected.

In case of Coal Seam No. 12 in Trang Khe Area of Hong Thai Coal Mine in September 2013, the fire started at a place where a coal seam was penetrated by a rock entry heading so that only one sealing wall was not enough to suppress the spontaneous combustion. Same as Table 2-2,

higher H₂ and CO gas concentrations were observed and no hydrocarbon gas except for the lowest rank CH₄ gas was observed through all the analyses.

In case of Ha Lam coal mine, the coal spontaneous combustion occurred twice in June and September of 2017. In the first time, the fire started at place of coal pillar of Coal Seam No. 10, which isolated a ventilation roadway from the goaf area beneath the block. Lately, two sealing walls were made in order for isolating the fire area. The gas analysis results are shown in Table 2-4.

In the second time, coal spontaneous combustion occurred at the goaf area of a mechanized long wall of Coal Seam No. 7. The cause of coal fire might be air leakage through the sealed wall behind the goaf area. A series of countermeasures were taken including nitrogen injection through a borehole and constructing of sealing walls. The results of gas analysis are shown in the Table 2-5.

For both cases of Ha Lam coal mine, the gasses in the fire area showed higher concentrations of H₂ and CO gases. Along with the time sequence, these concentrations were reduced due to the injection of nitrogen gas into the sealed area. However, the results showed the same tendency of hypothesis of only H₂ and CO gasses in the sealed area.

In case of Mao Khe coal mine in January 13th in 2017, the coal spontaneous combustion occurred at the fourth sub-level roadways of -14 m, -38 m, -48 m and -58 m levels. Sealing walls were made at each sub-level by wooden plates and clay. Lately, the whole area is isolated by three sealing walls at the level of -25 m, -80 m and -150 m cross-cut roadways. The gas samples were periodically collected and analyzed. The results of gas analysis from the isolated areas were shown in Table 2-6. The main gases from the fire in the isolated area were also composed of mainly H₂, CO and CO₂.

Taking account for the effect of nitrogen injection into the fire area, the results from three tables (Tables 2-4 to 2-6) show the change of gas concentrations inside the sealed wall. Since the coal seam No. 10 had low gas methane content at Ha Lam coal mine, the oxygen concentration was reduced in proportion with the increase of nitrogen concentration. The level of coal spontaneous combustion also was reduced along with reduction of CO and H₂ gas concentrations.

In contrast with the lower methane gas content of coal seam, the results of gas analysis for coal seams No. 7 (Ha Lam Coal Mine) and No. 10 (Mao Khe Coal Mine) showed their differences in the gas concentrations. In case of coal seam No. 7 (Ha Lam Coal Mine), the methane gas concentration inside the sealed area increased up to around 30%; the CO₂ concentration also increases up to around 3.7%. During the nitrogen injection period, the methane gas concentration varies from normal level to higher level, especially, sometimes in the range of methane gas explosion (5-15%).

In case of coal seam No. 10 (Mao Khe Coal Mine), the effect of nitrogen injection was observed by the reduction of CO gas. However, the concentrations of methane and hydrogen

gases increased. In the period of nitrogen injection, the methane gas concentration stayed in the range of explosion and the concentration of hydrogen also stayed in the range of explosion (on Feb. 6). This fact indicates the risks of coal spontaneous combustion not only for fire of coal but also a hazard of gas explosion.

2.3 The site mining condition and the cause of spontaneous combustion

Beside the susceptibility to spontaneous combustion of anthracite itself, some site factors might promote this phenomenon. For all those coal mines that experienced spontaneous combustions, the spontaneous combustions started around the rainy season of northern part of Vietnam. Among eighteen (18) reported spontaneous combustions shown in Table 2-1, fifteen (15) [83 %] of them occurred from May to September, when sudden squall showers within sunny days were repeated, and others occurred twice in January, one time in March, respectively.

Mining skeleton design also has an important role for preventing spontaneous combustion. Most coal mines have poor safety coal pillars between the worked out panel and the newly developing panel, which has to protect the return gate roadway for the next coal winning panel. By the rock pressure effect of the former coal winning panel, the return gate cross section size is reduced, which results in easier introduction of intake air into the goaf area and outbreak of spontaneous combustion. An example of a narrowed return gate road is shown in Figure 2-5, in which narrowing the gallery and floor heave can be found by the lifting up of the rail of the gallery floor up to near the ceiling of the return gate. Figure 2-5 was taken at the return gate of the long wall face behind of which No. 4 spontaneous combustion started in the goaf as shown in Figure 2-4 and Table 2-1.

At some coal mines, even the permanent galleries are located inside the coal seam to be developed. The air can be penetrated from the gallery to coal seam through crack system around galleries as well as water penetration from surface, which might cause introduction of fresh air after dripping or drying the penetrated water. Such shifted skeleton of the mine often results in spontaneous combustion. (See Figure A2-1 and Figure A2-6 in Annex 2.)

From the above facts, systematic countermeasures have to be developed for preventing spontaneous combustion at anthracite coal mines in Vietnam.

2.4 Conclusion

Regardless the common knowledge that anthracite coal hardly starts spontaneous combustion, 18 time spontaneous combustions have been reported in anthracite coal mines in northern Vietnam since May 2004 up to date. Some of these are different from the spontaneous combustion of bituminous or sub-bituminous coal. Site data of spontaneous combustions at

anthracite coal mines can be summarized as follows:

- (1) Even anthracite coal mine starts spontaneous combustion easily. Eighteen spontaneous combustions have occurred at seven coal seams of six coal mines up to date since May 2004.
- (2) In the shortest case, a coal seam started spontaneous combustion within two weeks after exposure to air. (Less than two weeks after a rock entry heading penetrate the coal seam.)
- (3) Spontaneous combustions started at both goaf area and freshly exposed coal wall.
- (4) Higher concentration of hydrogen gas (H_2) was observed in gas samples from inside the sealing.
- (5) Except for methane gas (CH_4), no higher rank hydrocarbon gases such as ethane (C_2H_6), ethylene (C_2H_4) or others were found. This feature is completely different from the cases of bituminous or sub-bituminous.
- (6) The more coal mines go to fresh coal seam area, the more spontaneous combustions they are facing. Namely, for anthracite coal mines, the increase of depth gives more chances for occurrence of coal spontaneous combustion due to the increase of strata pressure, ventilation pressure, etc.

Reference

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- 2-3) Ha Lam Coal Company. Report of coal spontaneous combustion. June and September 2017.
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Chapter 3: Gas Generation from Heated Coal^{3-1), 3-2)}

3.1 Introduction

As shown in Chapter 2, the results of gas analysis from several coal spontaneous combustions at Vietnamese anthracite coal mines showed special features totally different from the ones of bituminous coal spontaneous combustion³⁻²⁾. Research on gases generated from heated coal is one of the key points for earlier detection and determination of coal spontaneous combustion at underground coal mines.

As reported by Banerjee³⁻³⁾, there are many kinds of gases produced by coal spontaneous combustion. Most research were considered carbon monoxide (CO) and carbon dioxide (CO₂) as main gases generated from coal heating process. Hence, there are several gas indices for determination of the coal mine fire stage. One of the most widely used gas indices for assessing of coal mine fire situation is Graham's ratio³⁻⁵⁾. The Graham's ratio is a ratio between CO production and O₂ consumption. In the Young's ratio³⁻⁵⁾, CO₂ was considered as one of main gases generated during mine fire. The latter ratio was established by using the relation between CO generation and O₂ consumption. Willet's ratio³⁻⁵⁾ is a relation between CO product and the sum of N₂ excess, CO₂ production and combustible gas. Jones and Trickett ratio³⁻⁵⁾ is shown by the ratio between the sums of (CO₂ + 0.75 CO – 0.25 H₂) and O₂ deficiency. Chosh and Banerjee³⁻⁵⁾ also introduced the C/H ratio in order to determine mine gases. Then, the gas indices have been used to assess the intensity characteristics of fire. L. Yuan and A.C. Smith³⁻⁶⁾ reported their research on the gas emission from spontaneous heating of coal. This research showed that the main gases generated at initial stage of coal spontaneous combustion were CO and CO₂. Hence, they proposed the CO/CO₂ deficiency ratio for early detection of coal spontaneous combustion at coal mines.

Based on the above results and theories, gas indices of coal spontaneous combustion are divided into two groups of gases. One is the group of gases generated at initial stage and low temperature of coal spontaneous combustion. This group includes CO, CH₄, CO₂ and O₂, which are sometime called a basic group. The other group is H₂ or trace gases including C₂H₆, C₃H₈, iso-C₄H₁₀, n-C₄H₁₀, C₂H₄, C₃H₆, C₂H₂, and H₂³⁻⁷⁾. As the requirements by Czech Republic of State Mining Authority, both groups of gases generated from coal mines should be monitored through the results of gas samples from underground coal mines. China also requires coal mines to monitor some kinds of gas at underground coal mine such as CH₄, C₂H₆, C₃H₈, iso-C₄H₁₀, n-C₄H₁₀, C₂H₄, C₃H₆, C₂H₂, CO, and CO₂. As shown in the research of Lu et al., the heating tests were carried out by using bituminous coal samples from Long Gu coal mine³⁻⁸⁾. In their experiment, several gases were detected during coal heating process such as CO, C₂H₄,

and C_2H_6 at the ambient temperature. These kinds of gases generated depended on the phase of coal heating steps. Finally, the experiment showed that using CO and other hydrocarbon gases used depends on the stages of rising temperature. The other research on coal spontaneous combustion for bituminous by Wang et al.³⁻⁹⁾ showed the formation of CO_2 and CO at low temperature oxidation of coal. The direct burn off reaction was the results of carbon oxide production. The research also proposed to use the ratio of production rate of CO_2 and CO for assessing the oxidation level of coal.

In the report of gases generated during coal heating experiments by Kim³⁻¹⁰⁾, the gases generated from the experiment are determined by using the combination of differential thermal analysis and crossing-point temperature method. Coal samples were heated in a tube furnace with the air flow going through the tube. These experiments were conducted in two cases, one was wet air flow and the other was dry air flow. In both cases, the temperature of coal samples was increased until it became equal or more the furnace temperature. From the experiments, Kim made clear of the relationship between the amount of O_2 consumed and the amount of CO_2 produced during the experiments. Both of these factors had the tendencies to increase with the increasing temperature for each sample. Generally, the production of CO_2 varied directly with the consumption of O_2 . In detail, at 200 °C, the average amount of CO_2 produced was less than 25% of O_2 consumed. At low temperature, the relationship between them became more random. The results showed that CO_2 was a primary product of coal oxidation. In the case of analysis of mine fire gas also indicated that CO_2 was the product from coal heating process. CO_2 production is directly related to O_2 depletion. The relation was shown more clearly by the rate of CO_2 at mine fire increase by 0.7% for every 1% decrease of O_2 concentration. In case of O_2 concentration decreased to 2%, the CO_2 concentration increased up to maximum of 15%. Beside the generation of CO_2 , the generations of other gases have been reported. CH_4 desorption was shown to be related with the increase of temperature. The production of CO in mine fire is usually evident only at higher temperatures (>50 °C) and lower O_2 concentration (< 12%).

Different from the above mentioned methods, Polish mining industries is concerned with more gases generated from coal heating process. Recently, a research method of the Central Mining Institute (GIG) of Poland was transferred to Vietnamese coal mines through the cooperation program. In accordance with the Polish method, the gases generated from coal mines are determined from coal heating test results. About 400 g of coal sample are subjected for the heating test by using a heating oven that has a temperature control program, and the air flow is introduced to a coal sample's container at a constant flow rate. Coal sample's temperature is received from the oven for every 25 °C per step from ambient temperature and sampling gas sample is obtained at each step. Hence, gas sample is analyzed by using a gas chromatograph with the detection sensitivity of 0.01 ppm for hydrocarbons. Data from gas analysis are collected and plotted on a graph between temperatures of coal sample versus generated concentration of

each gas. The relationship between those gasses was built by using the ratio between gases, for example, concentration of CO, CO₂, C_nH_m/CO₂ and so on.

The purpose of those steps is to simulate the situation of gases generated during coal heating process in the laboratory. Taking this method for Vietnamese coal, several coal samples have been tested by sending those coal samples to GIG for analysis. As shown in the analysis results, the hydrocarbon gases were found with very small amounts (less than ppm order) during coal heating tests. The reason might come from the test condition such as flow rate of air, speed of temperature rise step and coal sample requirements. One of the important aspects of this test method is the differences between test results and site analysis results. In order to use this method for Vietnamese coal samples, the condition as well as the requirement for the test should be modified to appropriate conditions.

All of those methods above showed their advantages and disadvantages for use in the field situation. However, all of those methods showed the interesting points that coal heating produce several kinds of gases such as CO, CO₂, H₂, and hydrocarbon group. The results from the above mentioned research showed the characteristics of gas generation from most kinds of coals except anthracite coal.

In order to find out the characteristics of gas generated from coal heating, an experiment method of coal heating has been established. As shown by several results, the gases generated from coal heating were varied depend on the coal type and coal components. The evidences of the gas analysis results clearly showed that anthracite coal did not generate hydrocarbon gases during the oxidation process. The reason might be deduced from the facts that much less methane gas content as well as much less volatile matter in anthracite coal was obtained. In addition, anthracite contains much less hydrocarbon molecule or hydrogen element in itself comparing with bituminous as shown in Table 3-2 of elementary analysis. From these facts, hydrocarbon gases might hardly appear even through oxidation heating process such as spontaneous combustions at mining sites.

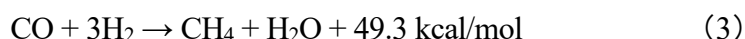
The results in Table 3-1 show lower hydrogen content in those anthracite coal samples. It might be difficult to explain that the source of hydrogen in Tables 2-2 and 2-3 has come from coal itself. Then another source of H₂ might be the “water gas reaction” at the fire area with high temperature. Those might be the reactions (7) or (8) as shown in the provable reactions in which carbon (C), water (H₂O), and other relevant gases are involved:

(1) *Methane gas generation:*

Heat decomposition:



Reaction with hydrogen:



(2) *CO gas generation*

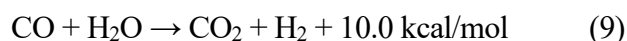
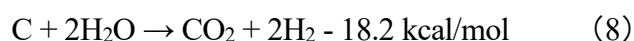
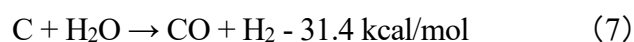
Reaction with Oxygen:



Reaction with Carbon Dioxide:



(3) *Hydrogen gas generation:*



Some of those reactions might only occur at the higher temperature around several hundred degrees (°C). Full scale big underground fire experiments were carried out under the condition of “fuel rich” and much H₂ gas was observed in the burned gas of the fire up to 5% depending on the fire condition. From those experiences, the concentration of H₂ gas might be a good indicator of the combustion temperature.³⁻⁴⁾

From these facts of gases generated from anthracite coal spontaneous combustion, a series of experiments were carried out with the aim to find out the feature of spontaneous combustion of anthracite coal.

3.2 Coal analysis

Coal samples were collected from coal faces of three underground coal mines. Thereafter, coal samples were crushed to sieve out to the requirements of particle size for proximate analysis. From proximate analysis results shown in Table 3-2, all coal samples can be defined as anthracite because of their higher fuel ratios, but depending on the criteria of classification, coal sample from Khanh Hoa Coal Mine might be regarded as “semi-anthracite”³⁻¹⁾.

Table 3-1: Proximate analysis of coal from underground coal mine in Vietnam

Coal Mine	Seam No.	Moisture (%)	Ash (%)	Volatile Matter (%)	Fixed Carbon (%)	Fuel Ratio
Hong Thai	12	0.73	39.03	4.07	56.17	13.8
	18	0.56	14.72	3.29	81.43	24.8
No. 91 Ent.	5	3.14	23.20	6.78	66.88	9.9
	4	1.32	9.12	2.47	87.09	35.3
Khanh Hoa	16	0.85	16.44	9.40	73.31	7.8

Table 3-2: Elementary analysis of coal from underground coal mine in Vietnam

Coal Mine	Seam No.	Total Sulfur (%)	Carbon (%)	Hydrogen (%)	Nitrogen (%)	Oxygen (%)	Ash (%)	Sulfur in Ash (%)
Hong Thai	12	2.12	55.31	1.73	0.69	0.42	39.32	2.14
	18	1.54	79.49	2.27	0.78	0.67	14.80	1.55
No. 91 Ent.	5	3.49	66.48	0.42	0.47	3.01	23.20	3.28
	4	2.35	85.19	1.28	0.69	0.22	9.24	2.38
Khanh Hoa	16	1.89	75.73	2.92	2.42	1.10	16.58	1.91

3.3 Heating experiment of Vietnamese anthracite coal³⁻¹⁾

3.3.1 Equipment and experimental procedure

Because of differences of generated gases during heating process of anthracite and other kind coals, for further discussion on the results shown by Tables 2-2 and 2-3 as well as on above discussion, coal sample heating experiments were carried out by using a devise shown in Figure 3-1.

In these experiments, a coal sample of around 100 g with particle size of less than 500 μm was sealed into a cylindrical steel made container, which has two holes at both side ends for introducing constant flow of oxygen or air. This container was located at the center of an electric furnace that had a temperature control program with a constant temperature increase rate at 0.5 $^{\circ}\text{C}/\text{min}$ and a constant air supply system with a flow rate at 100 ml/min, and generated gases were analyzed by gas chromatograph. During this process, supplied air was saturated with water vapor by introducing the air through a closed water container by bubbling. This equipment has two temperature measuring systems; namely, one is used for controlling the temperature of the heating furnace that controls the temperature increase rate and the other one is used for measurement of the coal sample temperature during heating test.

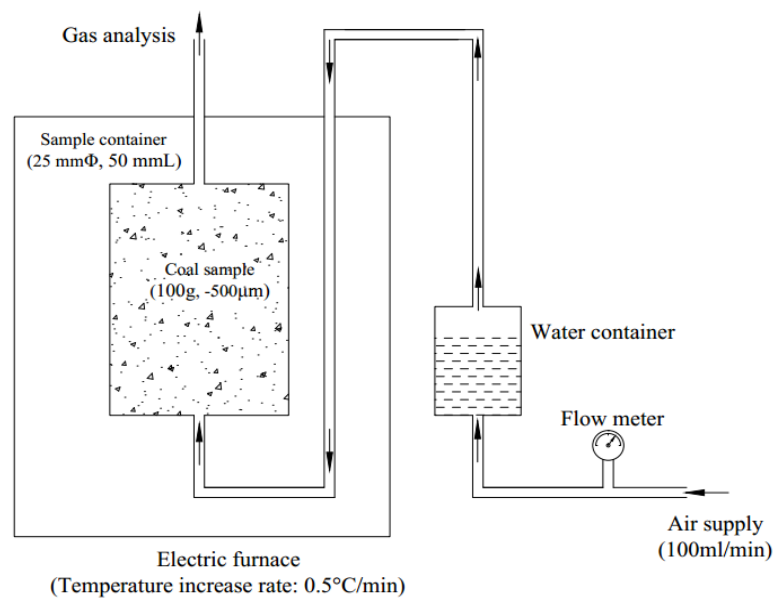
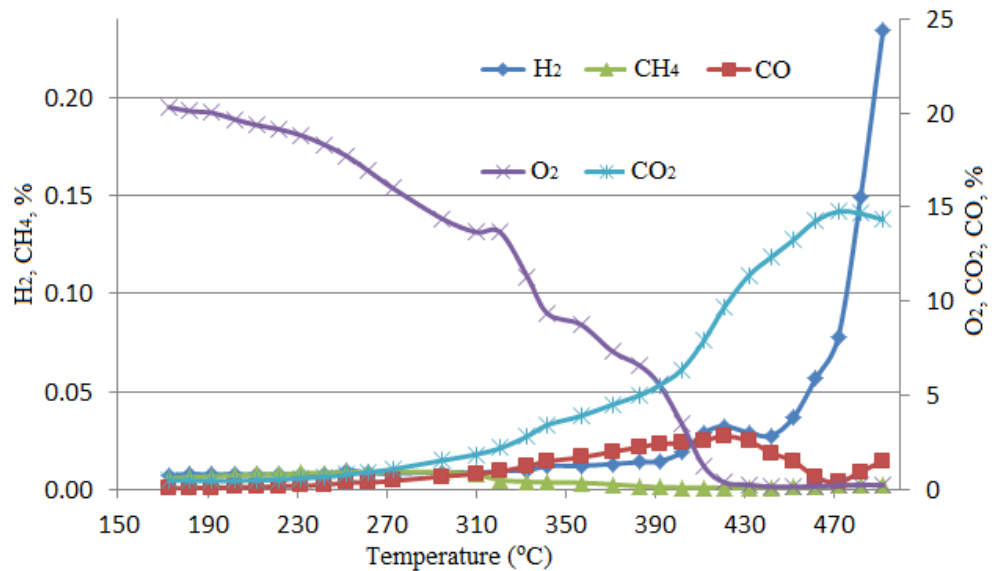


Figure 3-1: Schematic diagram of experimental setup for analyzing gases from heated coal

3.3.2 Results and discussion

Three anthracite coal samples from Hong Thai Coal Mine (Coal Seam No. 24), Khanh Hoa Coal Mine and No. 91 Enterprise Coal Mine, and one sub-bituminous from Phan Me Coal Mine were used. Experimental results of anthracite are shown in Figure 3-2 and Table 3-3 whereas experimental results of sub-bituminous are shown in Table 3-4.



**Figure 3-2: Gas generation from anthracite coal during heating process with air supply
- Coal Seam No. 4 of No. 91 Enterprise Coal Mine**

In case of anthracite coal samples from Coal Seam No. 4 of No. 91 Enterprise Coal Mine as shown in Figure 3-2, O₂ gas started to decrease gradually from nearly 200 °C, and CO₂, CO and H₂ started to increase from around 250 °C. On the other hand, CH₄ generated from much lower temperature at an almost constant concentration and no other hydrocarbon gas came out through all the temperature range. These tendencies were the same as those even in case of other anthracite coal samples from other three coal mines as shown in Table 3-3. In these tests, H₂ gas appeared from around 200 °C or 250 °C and no higher grade hydrocarbon gases (C₂ or higher) was observed throughout the experiments. From these facts, it might be possible to deduce that CH₄ gas might come from desorption due to increase of the temperature of coal itself whereas other gases such as CO₂, CO and H₂ might appear by some chemical reactions during heating process.

As shown in the temperature range greater than 450 °C of Figure 3-2, H₂ gas increased quite rapidly along with an increase of the temperature of coal samples, and CO gas decreased in an inverse proportion with H₂ gas. CO₂ gas also showed maximum at the same temperature range and in addition, O₂ gas became almost “zero” after 400 °C. From these data, the environment inside the coal sample container became “complete reduction atmosphere”, so that reactions such as reactions (7), (8) or (9) might be easy to occur.

Table 3-3: Gas generation from anthracite coal during heating process with air supply

Coal Mine (Seam No.)	Temperature (°C)	O₂ (%)	CO₂ (%)	CO (%)	CH₄ (%)	H₂ (%)
Hong Thai (Seam No. 24)	270 °C	3.85	3.47	1.46	0.002	0.019
	283°C	1.16	9.40	1.79	0.003	0.033
	295 °C	0.30	7.70	2.07	0.002	0.029
No. 91 Enterprise (Seam No. 5)	240 °C	12.92	1.79	0.43	0.005	0.039
	270 °C	4.72	4.34	0.91	0.007	0.077
	306 °C	0.00	9.28	1.22	0.005	0.089
Khanh Hoa (Seam No. 16)	240 °C	8.11	2.62	0.98	0.002	0.028
	266 °C	0.00	9.53	2.80	0.003	0.025
	302 °C	0.00	14.12	4.58	0.003	0.022

Table 3-4: Gas generation from Phan Me sub-bituminous coal during heating process with air supply

Temperature (°C)	O₂ (%)	CO₂ (%)	CO (%)	CH₄ (%)	C₂H₆ (%)	C₂H₄ (%)	H₂ (%)
182	15.94	0.62	0.20	0.038	0.002	0.0004	0.015
230	0.18	3.47	1.56	0.088	0.017	0.013	0.043
282	0.33	7.27	2.72	0.058	0.007	0.010	0.023

Even in the other experiments of anthracite as shown in Table 3-3, it may be possible to understand that (1) no hydrocarbon gas except for CH₄ gas comes out and (2) H₂ gas appears during heating process. Although it is not clear that how much extent of site conditions of spontaneous combustion are represented in the series of coal sample heating tests by using the set-up shown in Figure 3-1, it might deduce qualitatively that the analyzed results of in situ sampled gases have similar characteristics to gases generated from artificial heating tests of coal samples with some air flow.

Apart from these tests of anthracite, an example result of artificial heating tests with sub-bituminous from Phan Me Coal Mine is shown in Table 3-4. In this test, some discriminative points were detected such as: (1) hydrocarbons of C₂H₄ and C₂H₆ with concentrations around hundreds ppm were detected together with CH₄ from lower temperature around 200 °C or less, (2) rapid oxidation processes might occur from much less temperature than the case of anthracite from around 190 °C and then O₂ gas became almost zero at around 230 °C, which was 200 °C lower than the case of anthracite and (3) after the atmosphere inside the coal sample holder became “complete reduction atmosphere” at the temperature range greater than 230 °C, there was not so much change in generations of H₂, CO, CO₂, C₂H₆ and C₂H₄. The other gases of CO and CO₂ showed not so much differences between the cases of anthracite in all the tested temperature range.

3.4 Proposal for an indication gases of spontaneous combustion of anthracite

The first gas indices for spontaneous combustion were proposed by Graham who focused mainly on CO and CO₂ released and O₂ concentration decreased during coal oxidation process and these have been widely used worldwide under the name Graham’s ratios³⁻⁵). Depending on the applied method and coal rank in each country, many researches have focused on the other

hydrocarbon gases generated during coal oxidation process (C_2H_6 , C_2H_4 , C_2H_2 , C_3H_8 , C_3H_6 , etc.)³⁻⁶). In the researches done by an Indian researcher, the mine fire gas indicators were proposed such as production of CO, disappearance of CO, oxygen consumption, CO/CO₂ ratio, Willet's ratio, C/H ratio and CO/CO₂ deficiency³⁻⁷). Most researches on gas indicators for coal spontaneous combustion rely on the generation of hydrocarbon gases as well as CO, CO₂ and O₂ deficiency at the temperature range less than 100 °C. In a Japanese reference handbook on coal spontaneous combustion of bituminous coal mines, generation of many kinds of hydrocarbon gases such as C_2H_6 , C_2H_4 , C_3H_8 , C_3H_6 and C_2H_2 in addition to CO and CO₂ are introduced³⁻⁷).

From the results of in situ sampled gas analysis from inside the sealing of spontaneous combustion as well as from the tests of anthracite coal oxidation, it was shown that the tendency of gas generation during spontaneous combustion of anthracite coal was quite different from the other types of coal. One important feature is generation of only two gases of CO and H₂ during heating process, both of which do not exist in the ambient ventilation air, and another important point is no generation of hydrocarbon gases except for CH₄ gas that might be released into ventilation even under the mine operation ambient temperature. From such situation, it might be possible to use the relationship between CO and O₂ generated during coal heating as one of the indicators for coal heating process of anthracite coal; however, as shown in Figure 3-3, blasting is conducted quite often and some heavy diesel engine machines are used at the area of nearby intake galleries so that CO gas might not be used as an indicator gas.

Hence, for detecting a spontaneous combustion of anthracite coal mines in Vietnam, H₂ gas in exhaust ventilation might be only one important indicator.

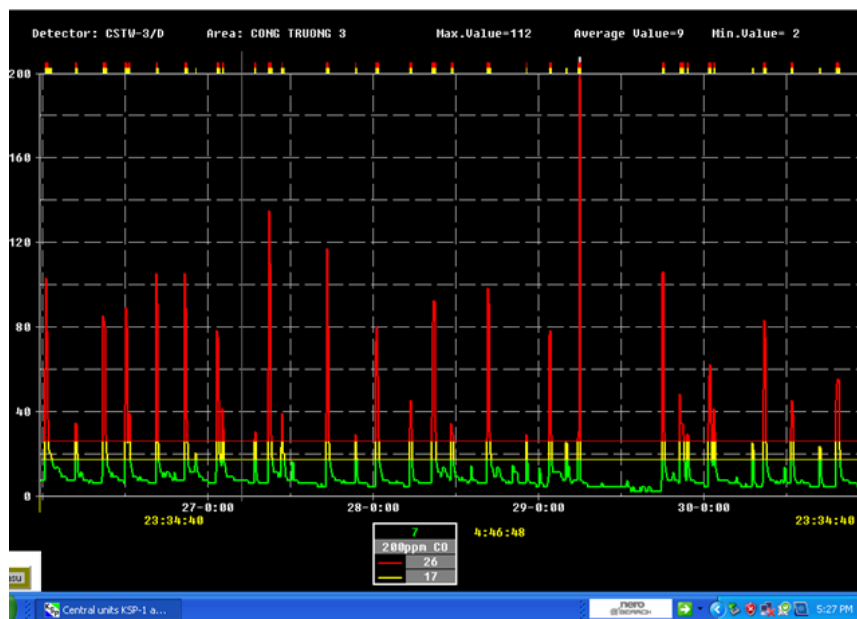


Figure 3-3: CO gas concentration change due to blasting at coal seam No. 4 of No. 91 Enterprise Coal Mine (From display of the central monitoring system)

3.5 Conclusion

In cases of spontaneous combustion of anthracite, only H_2 and CO, not contained gases in usual ambient air were observed in the sample gas from inside the sealing in contrast with bituminous coal that generates various hydrocarbon gases. Heating tests of several anthracite coal samples with some amount of fresh air flow supported the actual site phenomena shown above, namely, it was also shown that H_2 and CO gases were observed together with trace CH_4 gas as well as no hydrocarbon gas from C_2 or higher grade. Hence, H_2 might be only one important indicator for spontaneous combustion of anthracite coal.

The proximate and elementary analysis results showed that the characteristics of Vietnamese's anthracite coal had less amount of sulfur as well as hydro carbon groups. From these points, the cause of coal spontaneous combustion does not result from the oxidation of pyrite in coal samples. It is also useful to analyze the gases generated from heated coal samples. The cause of anthracite coal spontaneous combustion in Vietnam might also come from the low temperature oxidation of coal when it is exposed to the air. This is a basic hypothesis for the processing of investigation on the cause of spontaneous combustion of Vietnamese anthracite coal.

Besides the characters of spontaneous combustion of Vietnamese anthracite coal mines, there are many points that have to be improved from the viewpoints of prevention of spontaneous combustion. Because H_2 is inflammable gas with wide range explosive concentrations, the concentration of H_2 at underground coal mine should be considered as dangerous not only for coal spontaneous combustion but also for gas explosion. Hence, the research on the H_2 generation should be further concentrated on the phase of gas release depending on the temperature stages of coal oxidation process. Even if the feature and susceptibility to spontaneous combustion of anthracite coal differ from those of other types of coal, the site conditions resulting in outbreak of spontaneous combustion are not different from the others. Then the experiences in the prevention measures of spontaneous combustion of bituminous coal can also be useful for anthracite coal mine in Vietnam.

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Chapter 4: Conventional Methods for Evaluating the Susceptibility of Coal to Spontaneous Combustion⁴⁻¹⁾

4.1 Introduction

Because of lack of understanding on coal spontaneous combustion, the Vietnamese coal industry does not have its own test method for classification of coal on its proneness to spontaneous combustion. Since the first outbreak of coal spontaneous combustion, three foreign proposed methods have been examined if they are applicable to Vietnamese anthracite coal or not to evaluate the susceptibility to spontaneous combustion. Since 1970s, the Russian method⁴⁻²⁾ or U_{25} index has been used in the cases of coal spontaneous combustion at Khe Bo, Phan Me coal mines. The second method is the Japanese one⁴⁻³⁾ proposed by Idemitsu Co. The third one is Polish method⁴⁻⁴⁾ or Olpinski method.

4.2 Russian method⁴⁻²⁾

The Russian method was developed by Vasselovsky (IGD – Institute of Mining) to determine the susceptibility to spontaneous combustion of coal, which is based on an idea of isothermal absorption of oxygen by coal sample, which is confined into a flask with temperature of 25 °C. Based on the IGD proposal, the method has been developed by a researcher group of Russian Research Institute of Mine Rescue. Recently, it has become the method corresponding to the requirement in Article 476 and 477 of Russian Safety Rule for Underground Coal Mine. Outline of this method is as follows (Figure 3-1):

A coal sample is collected from a depth of 0.5 m of an active coal face in underground coal mine. The coal sample is sealed into a closed container to bring it from the coal mine site to the laboratory. The size of coal sample is adjusted in a range from 1 mm to 3.175 mm and the coal sample of 40 g to 100 g is put into a flask with the volume of 600 ml, which is filled with normal air and kept the temperature at 25 °C and the valves are closed for 24 hours. After 24 hours the gas inside flask is extracted and the concentrations of CH₄, O₂ and CO₂ are analyzed. After extraction of gases, the valves of the flask are opened to get fresh air into the flask and closed again for 24 hours for additional oxygen absorption by the coal sample and the inside gas is analyzed again. This procedure is repeated again and again for continuous ten days. From analyzed results and measured inside pressure of the flask, oxygen absorption index, $U_{25(n)}$ (ml/g·hr) is calculated based on the equation shown in the standard⁴⁻⁵⁾.

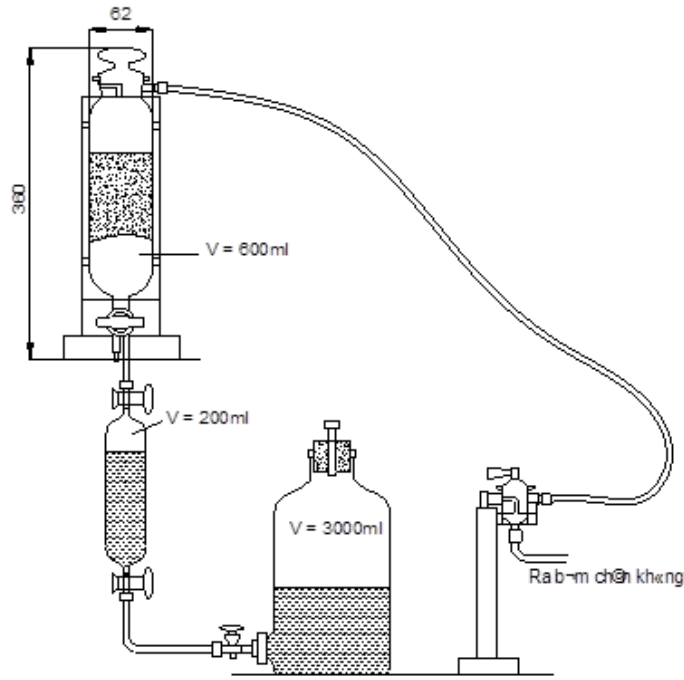


Figure 3-1: Concept of Russian method to derive U_{25} values

$$U_{25} = \frac{V(B-P)}{W.t.760} \ln \frac{(1-CO).Ca}{Co(1-Ca)} \quad (\text{ml/g.hour})$$

where:

- V – Air volume in closed flask [cm^3]
- P – Pressure of saturated vapor at 25 °C [23.8 mm Hg]
- B - Barometric pressure [mm Hg]
- W - Weighed portion of coal [g]
- t - Time of absorption [hour]
- C_O - oxygen concentration in the air [%]
- C_a - oxygen concentration in the air of flask after the elapse of time 24 hours [%]
- 760 - Barometric pressure of the air on normal conditions
- V - Air volume in closed flask [cm^3]

$U_{25(n)}$ is a value of oxygen absorption (ml) per unit weight (g) of coal and unit time (hour) of n^{th} day measurement ($n = 1$ to 10). The average U_{25} for ten days measurements is calculated. ($U_{25} = [U_{25(1)} + U_{25(2)} + \dots + U_{25(10)}]/10$)

Based on the Russian Method, if U_{25} is greater than 0.025 ml/g·hr, the coal is regarded as “higher susceptibility to spontaneous combustion”.

Based on the Russian Method, coal samples of Vietnamese anthracite were tested to get U_{25} indexes.⁴⁻⁶⁾ An example of ten days measurements of $U_{25(n)}$ and U_{25} for a coal sample from No. 91

Enterprise Coal Mine that has problems of spontaneous combustion is shown in Table 4-1. The results of U_{25} for coal samples from various coal mines are shown in Table 4-2.

From Table 4-1, $U_{25(n)}$ index after 24 hour elapsed time became smaller and smaller day by day from the first day measurement, $U_{25(1)} = 0.0146$ ml/g·hr decreased to $U_{25(8)} = 0.0014$ ml/g·hr, which were kept constant afterwards. Average U_{25} index was 0.0037 ml/g·hr, which was much smaller than the Russian criterion, 0.025 ml/g·hr in spite of spontaneous combustion problems at No. 91 Enterprise Coal Mine.

As shown in the previous our paper⁴⁻⁶⁾, Khanh Hoa, No. 91 Enterprise and Hong Thai, Mao Khe and Ha Lam coal mines have had spontaneous combustions for several times. The U_{25} index for their coal samples shown in Table 4-2 was all much smaller than the Russian criterion, however, U_{25} index of the coal samples from those coal mines that had spontaneous combustion problems showed higher values than the values of the coal samples from coal mines without problem of spontaneous combustion. Perhaps, U_{25} index might show some tendency to spontaneous combustion of coal sample. If the criterion of U_{25} for anthracite is set around 0.0020 or 0.0025 ml/g·hr, it might be able to use it as an index for susceptibility to spontaneous combustion of Vietnamese anthracite coal regardless Russian criterion, 0.025 ml/g·hr.

However, if we take account for short term start of spontaneous combustion after exposure of coal seam into fresh air flow⁴⁻⁶⁾ ten days period for deriving U_{25} by Russian Method is too long for practical application for Vietnamese coal mines. (Minimum starting time of spontaneous combustion was less than two weeks after a coal seam was penetrated by a rock entry.⁴⁻⁶⁾) This method also needs many batch operations with skillful techniques.

Table 4-1: Results of Russian method for No. 91 Enterprise coal sample

Day (n)	C_a (%)	$U_{25(n)}$ (ml/g·h)
1	11.6	0.0146
2	15.2	0.0067
3	17.3	0.0037
4	18.6	0.0022
5	18.7	0.0021
6	18.7	0.0021
7	19.3	0.0015
8	19.4	0.0014
9	19.4	0.0014
10	19.4	0.0014
Average U_{25}		0.0037

C_a : Oxygen concentration after 24 hours exposure of coal sample and air in the flask

Table 4-2: Summary of Russian method for other coal sample

Sample No.	Coal mine	Coal seam No.	U_{25} index (ml/g·hr)
2	Khanh Hoa*	16	0.0063
3	No. 91 Enterprise*	5	0.0037
4	Hong Thai*	10	0.0052
5	Vang Danh	8	0.0025
6	Ha Lam*	10	0.0011
	Khe Cham	12	0.0012
9	Thong Nhat	6	0.0007
10	Quang Hanh	7	0.0018
12	Duong Huy	5	0.0016

(The coal mines shown with * mark have problem of spontaneous combustion.)

Data of (1) Mao Khe, (8) Nam Mau and (11) 86 Ent. are not available.

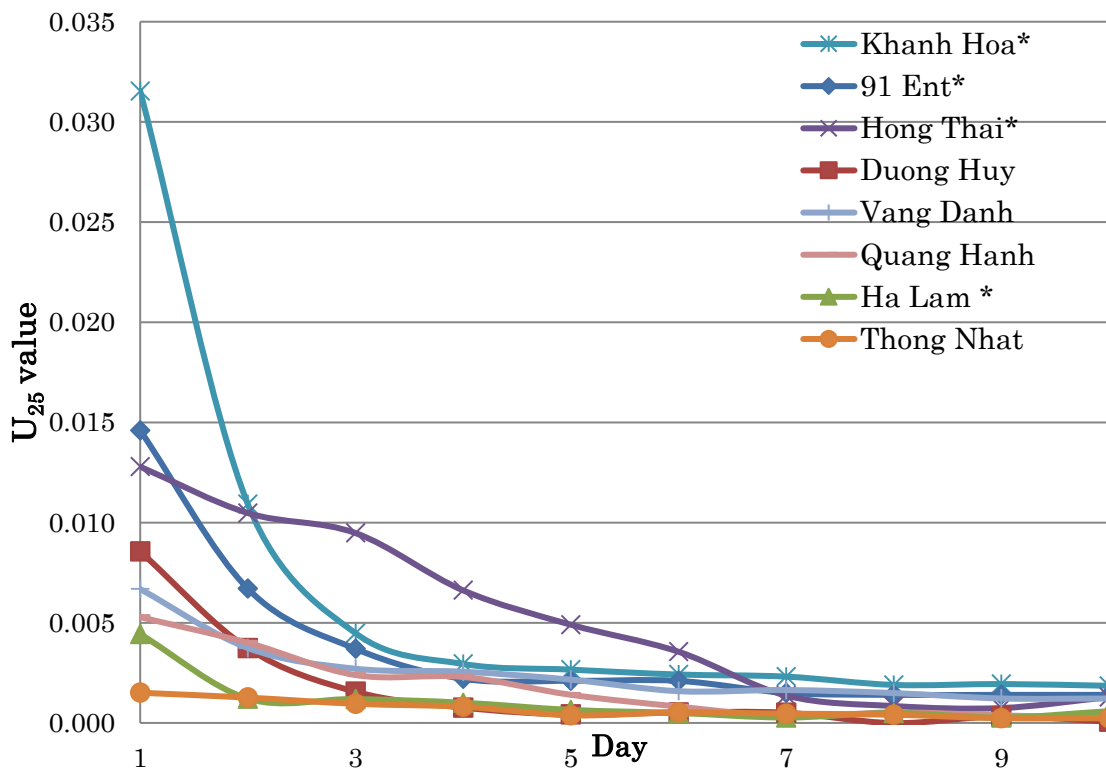


Figure 4-2: Daily change of $U_{25(n)}$ of coal samples shown in Table 4-2

The coal mines with * mark have spontaneous combustion problems.

In case of the Russian method, 10 day's average of $U_{25(n)}$ is compared with the criterion 0.025 ml/g·hr. As shown in Table 4-1, the value of $U_{25(n)}$ decreased in an exponential manner

day by day. Based on the idea that if this behavior of $U_{25(n)}$ is useful for estimating the susceptibility of anthracite to spontaneous combustion or not, all the data of change in $U_{25(n)}$ shown in Table 4-2 are plotted as shown in Figure 4-2.

Except for Hong Thai coal sample data, most of $U_{25(n)}$ values showed some exponential attenuation day by day and most of them showed clear attenuation up to the third day. In case of Hong Thai coal, it showed almost linear attenuation up to the 7th day. In cases of three coal mines of Khanh Hoa, No. 91 Enterprise and Hong Thai coal mines' samples showed much higher $U_{25(n)}$ values in the first two or three days; however, in case of Ha Lam Coal Mines sample showed the second lowest values even in the first two days.

From such data, application of the idea of Russian method to the spontaneous combustion might need further discussion. However, it is very clear that in case of anthracite coal samples, all of them show much higher oxygen absorption capacities within the first three days.

4.3 Japanese (Idemitsu Co.) Method⁴⁻³⁾

Another estimation method has been proposed for bituminous or sub-bituminous coal and used for evaluating the rank of susceptibility to spontaneous combustion of coal by Idemitsu Kosan Co., Ltd.⁴⁻³⁾ of Japan. This method adopts the value, T_{180} , which is the time needed to increase the coal sample temperature from 110 °C to 180 °C in an adiabatic oxidation equipment (Shimazu SIT-2: see Figures 4-3 and 4-4), details of which will be explained in the following chapter. The rank of susceptibility to spontaneous combustion is classified into 5 depending on the value of T_{180} as follows:

Rank A ($T_{180} > 115$ min): Hard to start spontaneous heating

Rank B ($T_{180} =$ from 85 to 115 min): Normal

Rank C ($T_{180} =$ from 70 to 85 min): Be careful

Rank D ($T_{180} =$ from 50 to 70 min): Danger of spontaneous combustion

Rank E ($T_{180} < 50$ min): Start self-heating in a short term

In this method, the initial coal sample temperature is set at 110 °C by introducing inert N₂ gas until the temperatures of coal sample and outside oven become equal and then N₂ gas is alternated to O₂ gas to start the oxidation of the coal sample. In case of a spontaneous combustion occurring at actual coal mine sites, the oxidation phenomenon starts from ambient temperature around 30 °C or 35 °C or less, which is much less than the method shown above.

Comparison between this Japanese method and the results of anthracite will be discussed in the following chapter.



Figure 4-3: SHIMAZU Ignition Tester (SIT-2) at Idemitsu Co. to determine T_{180} value
(<http://www.idemitsu.co.jp/rd/laboratory/environment/natural.html>)

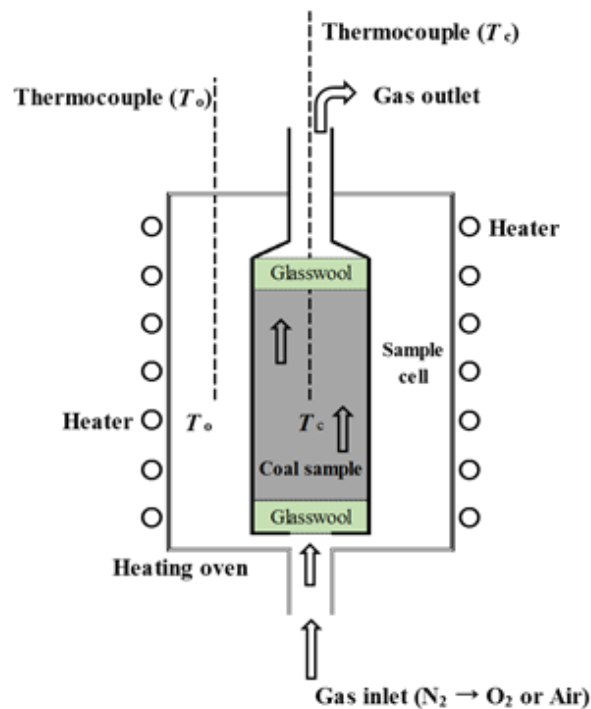


Figure 4-4: Conceptual diagram of an “adiabatic spontaneous ignition tester”
(SHIMAZU SIT-2)

In Figure 4-5 test results of No. 91 Enterprise coal sample (Coal Seam No. 5) based on Idemitsu Co. Method are shown. Three curves shown in Figure 4-5 were derived by using the same coal samples from Coal Seam No. 5. T_{180} values of curve (1), (2) and (3) were 207 minutes, 542 minutes and ∞ (no temperature increase), respectively. All these data were classified into rank “A” [Hard to start spontaneous heating]; however, 91 Enterprise Coal Mine has had four (4) time spontaneous combustions up to date.

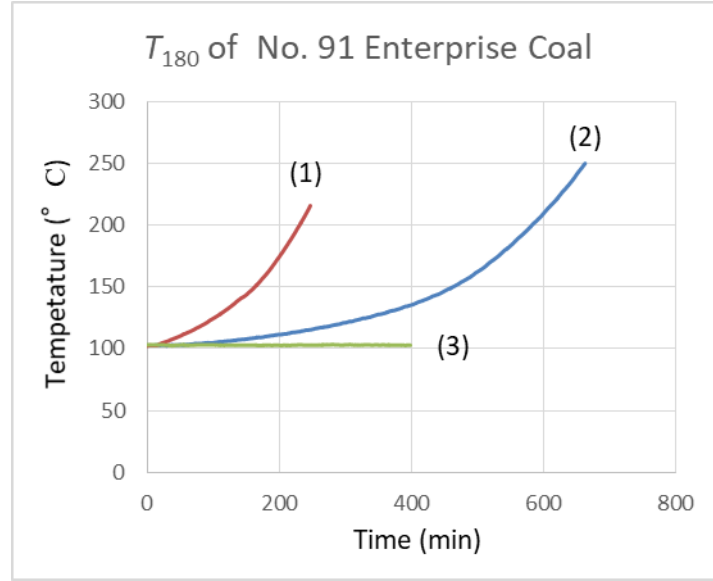


Figure 4-5: Measured results of T_{180} of No. 91 Enterprise coal sample (Coal Seam No. 5) based on Idemitsu Co. Method

From the data, the Japanese (Idemitsu Co.) method is applied to bituminous coal or sub-bituminous coal, and not to anthracite. Thus, it is also shown that “because anthracite has much less site for absorption of oxygen, anthracite hardly starts oxidation”.

4.4 Polish method⁴⁻⁴⁾

The Polish method was developed by Polish scientists, Olpinski and Struminki, and it is also called as Olpinski method sometimes. Their idea is to use the activation energy, E , which is introduced in Arrhenius' equation to show the relation between chemical reaction rate and temperature for evaluating the susceptibility to spontaneous combustion of coal. The “Arrhenius' Equation” shows the “chemical reaction rate constant”, k as follows:

$$k = A \exp (-E/RT) \quad (1)$$

where, A and E are “frequency factor” and “apparent activation energy”, respectively, both of which might be expressed as a function of temperature. R and T are “gas constant” ($R = 8.31451 \text{ J/K} \cdot \text{mol}$) and “absolute temperature” of the reaction sample, respectively.

To get the activation energy, E of coal sample, there is an empirical equation as shown below:

$$E = 96.79 \log_{10} (S_{za}/S_{za'}) \quad (2)$$

Where S_{za} and $S_{za'}$ are temperature increase ratio due to oxidation of the coal sample per an unit of time ($^{\circ}\text{C}/\text{min}$) at 237°C and 190°C , respectively. The value of S_{za} ($^{\circ}\text{C}/\text{min}$) is called as an “index

of spontaneous combustion”. To keep these temperatures constant, they used chemicals such as quinoline (boiling point = 237 °C). For their measurement, coal sample of 3 g with the size between 0.06 and 0.075 mm is shaped into a hollow cylindrical shape briquette, one end of which is closed. This briquette is inserted into an oven with the constant temperature of 237 °C or 190 °C and air is supplied at a rate of $25 \pm 3 \text{ dm}^3/\text{hr}$ ($= 41.7 \text{ l/min}$) into the oven from the open side. The temperature increase rate of coal sample briquette is measured when coal sample briquette temperature becomes equal to the oven temperature of 273 °C or 190 °C.

From such experimental conditions, the derived the activation energies are the average ones from 190 °C to 237 °C. By using those measured activation energy, E and index of spontaneous combustion, S_{za} , the susceptibility to spontaneous combustion of coal samples is classified into five categories from I (Too Low) to V (Too High) as shown in Table 4-3.

Recently, the Olpinski method is still used in Poland coal industry under the National standard of PN-93 G-04558. However, the device for the measurement was modified as shown in the Figure 4-6. The preparation for coal sample and the steps of measurements are the same as the above mentioned procedure. The oven temperature is controlled to keep temperatures constant before test by using the heating device instead of quinolone. The data of temperature rise per time inside an oven is recorded to draw the curve of temperature rise.

More than ten Vietnamese anthracite coal samples were sent to the Central Mining Institute of Poland (GIG-Poland) and asked them to evaluate the susceptibility to spontaneous combustion of those samples based on the Olpinski method, the results of which are shown in Table 4-4. The results vary from Category III (Medium) down to Category I (Too Low); however, Hong Thai Coal Mine has had spontaneous combustions in both Coal Seams No. 12 (Sample No. 4-1) and No. 18 (Sample No. 4-2) in spite of evaluation of Category I (Too Low), and 91 Ent. Coal Mine (Coal Seam No. 4) (Sample No. 3) has also had spontaneous combustion regardless evaluation of Category II (Low). Mao Khe Coal Mine (Category III: Medium) has never had spontaneous combustion regardless their disadvantageous mining method of advancing coal mining in all the coal faces up to the end of 2016 in their very long history, but it had a spontaneous combustion in January 2017. Namely, all the coal samples of anthracite have been classified into less susceptibility to spontaneous combustion by Polish Method in spite of actual problems of those coal mines.

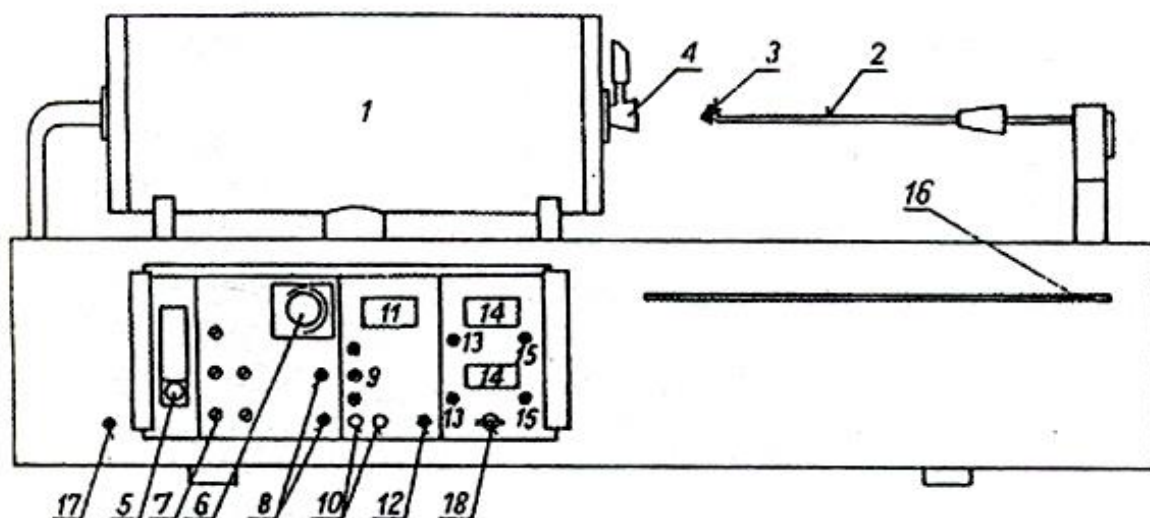


Figure 4-6: The device based on Olpinski method for determination of an index of spontaneous combustion

1-Oven; 2-Temperature probe; 3-Coal sample; 4-Mouth of Oven chamber; 5-Flow meter; 6-18: Indicator lamp and operation buttons;

Table 4-3: Classification of coal susceptibility to spontaneous combustion according to Polish (Olpinski) Method

S_{za} Index (°C/min)	Activation Energy, E (kJ/mol)	Classification	Susceptibility to spontaneous combustion
Up to 80	> 67	I	Too low
	46 to 67	II	Low
	< 46	III	Medium
> 80 to 100	> 42	IV	High
	< 42		
> 100 to 120	> 34	V	Too High
	< 34		
> 120	Abnormal		

Table 4-4: Determination of susceptibility of Vietnamese anthracite coal to spontaneous combustion by Polish method

Sample No.	Sample name	Combustion index		Activation Energy (kJ/mol)	Spontaneous Combustion Rank
		S_{za}	$S_{za'}$		
		°C/min			
1	Mao Khe*	7	3	39.16	III: Medium
2	Khanh Hoa*	26	6	61.3	II: Low
3	91 Ent.*	14	3	64.4	II: Low
4-1	Hong Thai (Seam 12)*	26	5	68.9	I: Too Low
4-2	Hong Thai (Seam 18)*	22	4	71.2	I: Too Low
5	Vang Danh	20	4	67.2	I: Too Low
6	Ha Lam	27	7	56.4	II: Low
7	Khe Cham	30	7	60.8	II: Low
8	Nam Mau	16	5	48.6	II: Low
10	Quang Hanh	15	3	67.2	I: Too Low
12	Duong Huy	21	6	52.3	II: Low
13	Thong Nhat	3	2	20.32	III: Medium
14	86 Ent.	4	2	31.74	III: Medium

(The coal mines shown with * mark have problem of spontaneous combustion.)

4.5 Conclusion

In case of the Russian method, all anthracite coal samples obtained from coal mines with problem of spontaneous combustion showed their U_{25} values from 28% (maximum) to 4% (minimum) of Russian criteria, $U_{25} = 0.025$. Namely, all those Vietnamese anthracite coals were evaluated as “Less or no susceptibility to spontaneous combustion”. However, if we compare U_{25} values within Vietnamese anthracite coal samples, the coal samples with higher U_{25} values comparing with the coal samples that did not have spontaneous combustion up to date.

In case of the Japanese method, the minimum T_{180} value of No. 91 Enterprise coal showed 1.8 times of the criteria, $T_{180} > 115$ min for “Hard to start spontaneous heating”.

In case of the Polish method, all the anthracite coal samples shows “III: Medium” to “I: Too

Low”

From the data shown above, it can be concluded that the three conventional methods are not suitable to evaluate the susceptibility of Vietnamese anthracite to spontaneous combustion. Namely, new methods should be discussed for spontaneous combustion of Vietnamese anthracite.

Reference

- 4-1) Le Trung Tuyen, K. Ohga, T. Isei. Susceptibility to spontaneous combustion of Vietnamese anthracite. *Journal of Mining and Minerals Processing Institute of Japan*, Vol. 133, No. 6, pp. 140 - 150, 2017.
- 4-2) Ministry of Fuel and Energy of Russia, Russian Safety Rule for Underground Coal Mine, Ministry of Fuel and Energy of Russia, Russian Technical Standard Association. Method for evaluation of coal susceptibility to spontaneous combustion (МЕТОДИКА ОЦЕНКИ СКЛОННОСТИ ШАХТОПЛАСТОВ УГЛЯ К САМОВОЗГОРАНИЮ)”, May 29th 1997.
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Chapter 5: Susceptibility of Anthracite Coal to Spontaneous Combustion⁵⁻¹⁾

5.1 Oxidation process of coal from lower temperatures

At coal mine sites, all spontaneous combustion start from ambient temperature of the coal mine site. From such a premise, it might be essential to examine the oxidation process of coal samples from lower temperature including ambient temperature around 25 °C or 30 °C.

For the purpose, an adiabatic oxidation process analyzer (SHIMAZU Ignition Tester: SIT-2), which is the same equipment used for Idemitsu method (Japanese method)⁵⁻²⁾, was used by setting initial oven temperature less than 100 °C in contrast to 115 °C for the Idemitsu method.

5.2 Introduction of testing equipment and its principle and function

As a definition, “spontaneous ignition” or “spontaneous combustion” is defined as following: “a substance starts to produce heat due to self-oxidation even from much lower temperature than the ignition temperature and the heat of it is accumulated for a longer time until the temperature reaches to the ignition temperature and finally it starts combustion by itself”. One method to examine self-oxidation of a substance is a way so called “adiabatic oxidation test” in which no heat exchange occurs between the subjected sample and outer test system. In such system, all the heat generated by the self-oxidation accumulates inside the subjected sample, which turns to temperature increase of the sample. One of such system is realized by SHIMASZU Co. as “Spontaneous Ignition Tester, SIT-2”, essential part of which is illustrated in Figure 5-1.

SIT-2 can be used for identify the “susceptibility (liability) of spontaneous combustion” of a sample material by using small amount (approximately 1g). The schematic diagram of the central functional part is shown in Figure 5-1. This equipment has two temperature controllers; namely, one is used for controlling the temperature, t_2 of the “oven” that adjusts the initial test environmental temperature and the other one is used for controlling the adiabatic condition when the sample generates the heat due to slow oxidation not so as to transfer the generated heat to outside the sample. By those two functions, the temperature of the oven, t_1 and the temperature of the sample, t_2 are kept same ($t_1 = t_2$) always during the test. The temperature of the sample, t_2 is displayed on the CPU monitor screen of the data treatment device time by time.

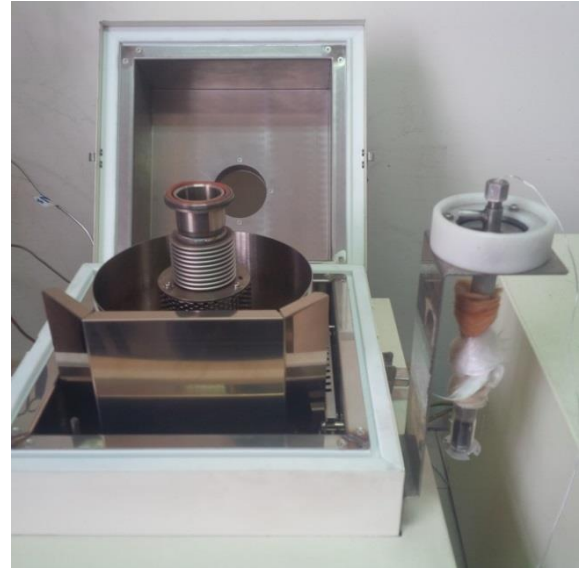
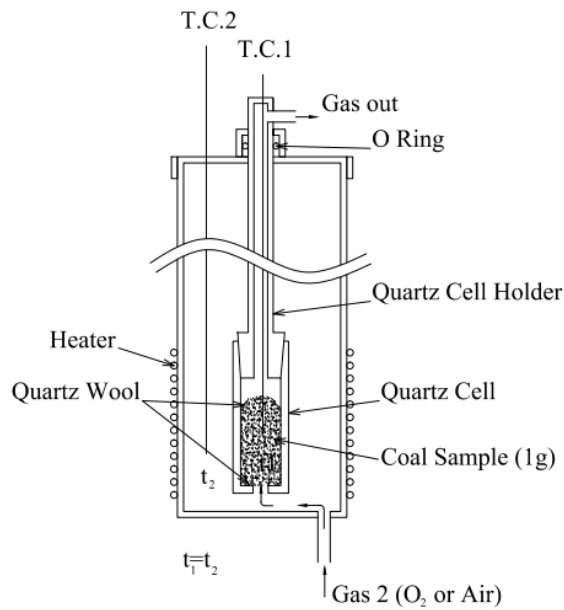


Figure 5-1: Adiabatic oven chamber and sample cell for oxidation reaction of the sample

(Right photo shows a status that the quartz sample cell is taken out from the oven.)

To keep adiabatic (no heat exchange) condition, oven temperature, t_2 is kept equal to coal sample temperature, t_1 ($t_1 = t_2$) always by the temperature control system. Before introduction of oxygen or air into sample cell, inert gas (N_2 gas) is introduced until “ $t_1 = t_2$ ” is achieved so as to keep “adiabatic” condition from the first of oxidation process of the sample.

Simple explanation of the measuring method is as follows: at first, the sample is put into the quartz made cell set at the temperature of the “oven” by introducing Gas 1 (N_2 gas) into the oven at a constant rate (1.5 ml/min) and this introduced Gas 1 (N_2) passes through the “sample” from the bottom hole of the quartz cell. This temperature is called as the “initial temperature”. After an enough suitable time (a half or one hour), the both temperatures of the sample, t_1 and the “oven”, t_2 become equilibrium state ($t_1 = t_2$). Then, the adiabatic heat controller is started and the introducing gas is changed from Gas 1 (N_2) to Gas 2 (O_2 gas or O_2 containing gas such as air or mixed gas of oxygen and nitrogen) into the “oven” chamber, when starting measurements. By the introduction of Gas 2, oxidation process might start. After some introduction time period, the sample might start to increase the temperature itself due to generating heat by oxidation. By measuring temperature increase trend of the sample, original oxidation reactions of the sample occur. This oxidation temperature increase curve might change due to many factors such as “initial temperature” of the test, Gas 2 (oxygen or air) introduction flow rate, characteristic of the sample itself (kind of coal, coal mine, coal seam, place of sampling, size of coal sample, moisture of coal sample, storage period of coal sample, etc.).

5.3 Experiment conditions

For this study, many anthracite lump coal samples from wide range Quang Ninh Coal Field were used. Among them coal samples from five coal mines with spontaneous combustions up to date are included. Those coal mines are Hong Thai Coal Mine, No. 91 Enterprise Coal Mine, Mao Khe Coal Mine, Ha Lam Coal mine and Khanh Hoa Coal Mine, etc.

In addition to those anthracite coal samples, one bituminous coal sample from Kushiro Coal Mine of Japan and one sub-bituminous coal sample from Phan Me Coal Mine in Thai Nguyen Province in Vietnam were examined in the same manner of anthracite coal samples. Both Kushiro Coal Mine and Phan Me Coal Mine have problems of spontaneous combustion. In case of Phan Me Coal Mine, they have ongoing spontaneous combustion problems.

Coal samples were kept in a closed container in which N_2 gas was filled just after brought back to the laboratory to prevent oxidation of coal samples under storage room temperature condition. These samples were subjected to the oxidation tests by SIT-2 within two or three days after sampling at coal mine sites. Effects of storage period after sampling were also examined by using the same sample.

According to the original procedure of the Idemitsu Kosan Company, the testing condition as well as coal sample is described as follows:

- Sample requirement: Putting 1g sample crushed to -0.25 mm into a quartz sample cell, which is set into an adiabatic condition of spontaneous heating test apparatus.
- Initial temperature: The sample is heated up to 110 °C under inert gas flow of N_2 gas.
- After stabilizing the sample temperature, the gas is alternated from N_2 to O_2 . And the time needed to increase up to 180 °C is measured under an adiabatic condition.

Based on this procedure, most coal samples of Quang Ninh coal mines were subjected to the test. Apart from the test conditions of the Idemitsu Kosan's procedure, some parameters such as initial oven temperature were changed for the purpose of simulating the in situ condition at the mining site. The experiments carried out up to date are summarized in Table 5-1. This table shows the number of experiments by the denominator of each column and the numerator of each column shows the number of temperature increase of coal sample in the adiabatic oven of SIT-2 as shown in Figures 5-2 to 5-5.

5.3.1 Representative temperature increase curves

As shown in Table 5-1, many coal samples were subjected for the test by changing the test conditions. Along with the change of the experimental conditions, the results showed differences. In some tests it took very long time for increasing the temperature and in some tests the temperature of coal sample did not go up even these coal seams had problem of coal spontaneous combustion. In these experiments, the conditions were varied from room temperature to 110 °C.

The purpose of those changes was to find out analogous oxidation condition of coal sample as coal mine site conditions.

Derived some representative temperature increase curves are shown in Figure 5-2 (No. 91 Enterprise coal), Figure 5-3 (Phan Me coal) and Figure 5-4 (Kushiro coal).

Table 5-1: Summary table of experiments by SIT-2 (1) -Effect of initial temperature

No	Initial Temperature	26 °C	30 °C	35 °C	40 °C	50 °C	60 °C	70 °C	80 °C	105 °C	110 °C	Total
1	Kushiro					1/8	1/1	1/1	3/4	1/1	1/1	8/16
2	Phan Me			0/1		0/7	1/1	2/3	4/10		1/2	8/24
3	Khanh Hoa	3/5				19/22			0/5			22/32
4	No. 91 Ent.	10/22			0/1	35/39	1/1	1/3	9/13		2/3	58/83
5	No. 618 Ent.								3/3			3/3
6	Hong Thai					7/9	1/1		6/11			14/21
7	Mao Khe	14/18				17/18			1/1			32/37
8	Uong Bi		1/1						0/3			1/4
9	Vang Danh					2/2			2/3			4/5
10	Nam Mau	0/2				2/3		1/1	5/9			8/15
11	Ha Lam	0/2		4/4		6/6			0/6			10/18
12	Giap Khau					4/4						4/4
13	Ha Rang					2/2						2/2
14	Duong Huy					3/3			2/2			5/5
15	No. 86 Ent.								1/1			1/1
16	Quang Hanh	24/31				23/24			3/4			50/59
17	Thong Nhat					4/5			1/1			5/6
18	Cam Thanh								0/1			0/1
19	Mong Duong					4/4			0/2			4/6
20	Khe Cham					5/6			0/4			5/10
	Total	51/80	1/1	4/5	0/1	134/162	4/5	5/8	40/83	1/1	4/6	243/351

Dominator: Number of experiments. Numerator: Number of temperature increase

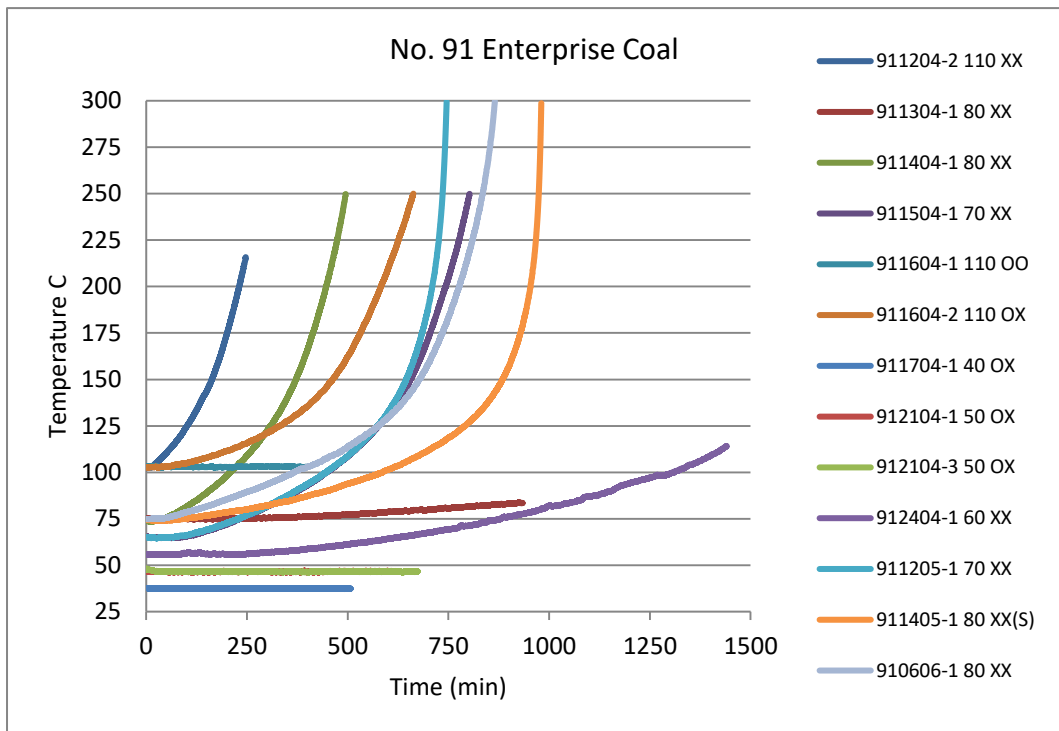


Figure 5-2: Test results of No. 91 Enterprise Coal

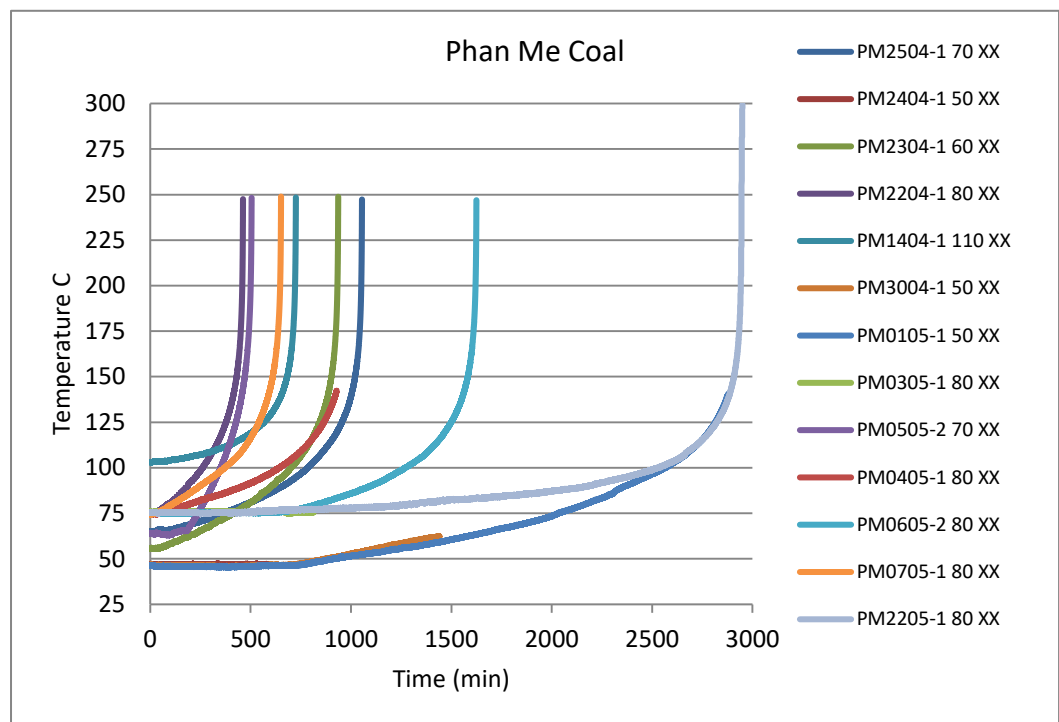


Figure 5-3: Test results of Phan Me Coal

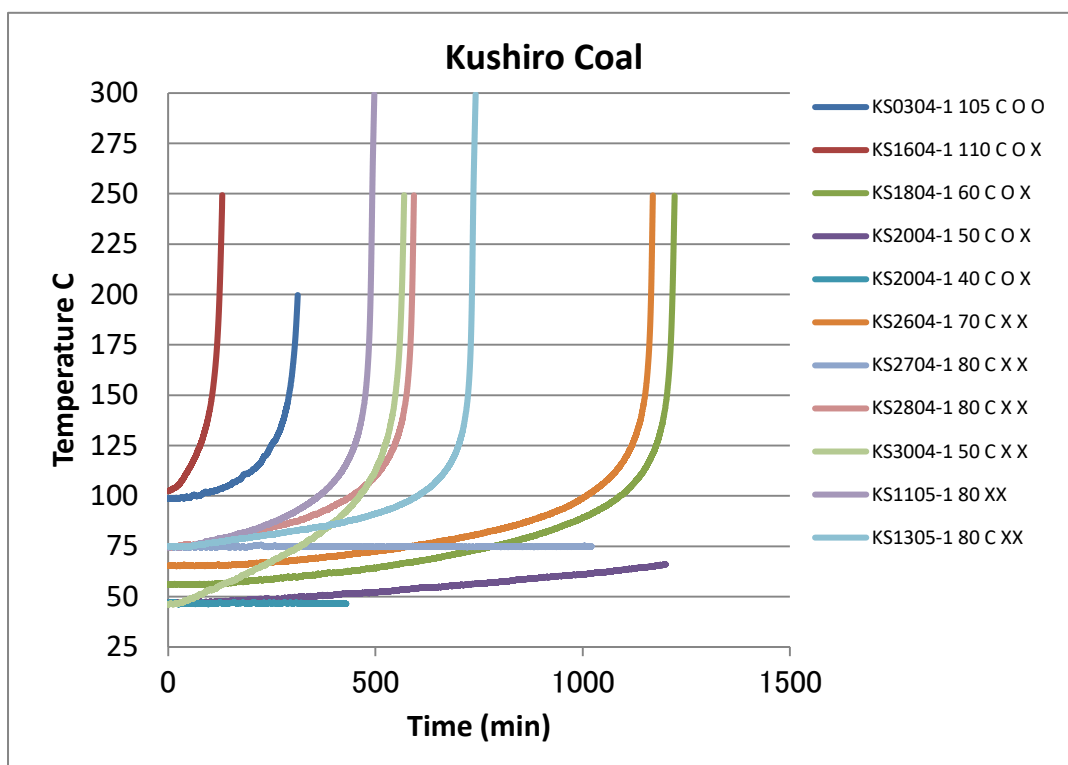


Figure 5-4: Test results of Kushirol Coal

Experimental conditions of SIT-2 for the tests shown in Figures 5-2, 5-3 and 5-4 are as follows:

- Temperature stabilizing time before starting the oxidation measurements: from 1 to 2 hours in N₂ gas (1.5 ml/min). (After starting the oxidation measurement, N₂ gas is cut off from SIT-2.)
- Initial oven temperature: from 26 °C to 110 °C (shown in the top column of Table 5-1.)
- Gas 2 (oxidation gas): oxygen (2.5 ml/min [first 30 minutes] then after 1.5 ml/min)
- Measured temperature range: Up to 250 °C or 300 °C.
- Moisture of coal sample: Most samples were used without drying process.
- Particle size of coal sample: -500 μm except for “911405-1” (-150 μm and +65 μm)

All three Figures show the test results of two coal mines in Vietnam and one of Japan, all of which have problems of spontaneous combustion.

From the file names in the figures you may be able to understand some test conditions. For example, first two characters show the coal mine such as “91” for No. 91 Enterprise Coal Mine, “PM” for Phan Me Coal Mine and “KS” for Kushirol Coal Mine, and next four figures show “Date” and “Month”, and next two figures with hyphen show “Experiment No. of the date”. Another two or three figures show the “Initial Temperature” of the test. The following characters show setting condition of SIT-2, which have no important meaning for understanding the test results except for “(s)” which means the particle size of coal sample is “Small” (-150 μm and +65

μm). For example, “911204-2 110 xx” shown on the top of the Figure 5-2 means “Experiment No. 2” of “No. 91 Enterprise Coal” on “April 12” with the “Initial Temperature = 110 °C”.

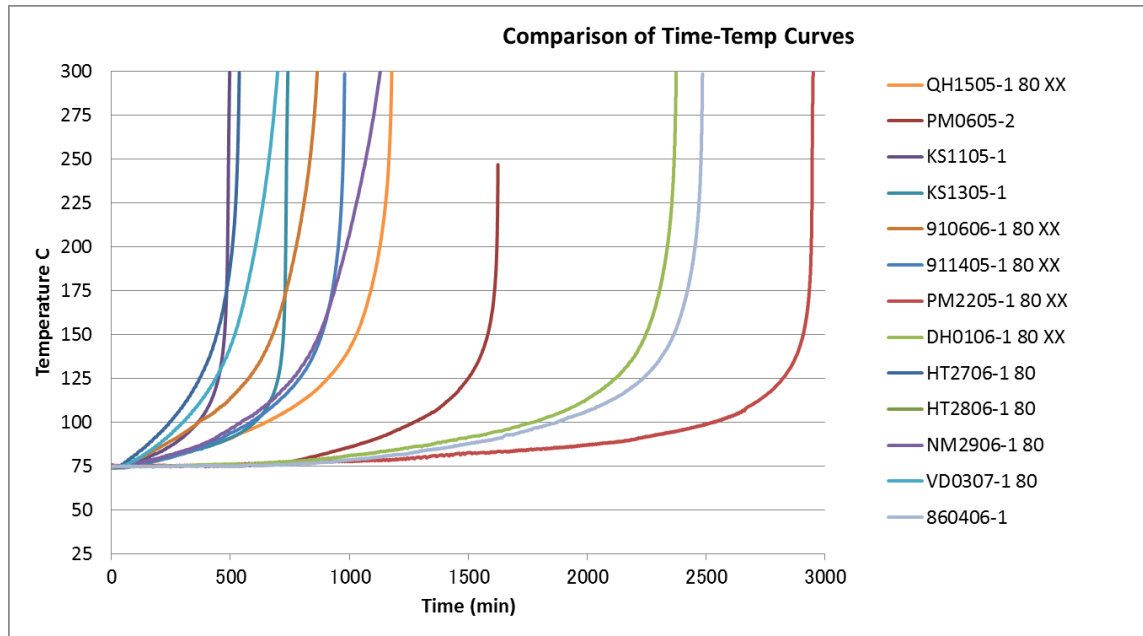


Figure 5-5: Comparison of Test Results of some coal mines

In Figure 5-6 test results of “Time-Temperature Curves” of different coal mine samples are compared with each other including coal samples of Quang Hanh Coal Mine and Duong Huy Coal Mine, Hong Thai Coal Mine, Nam Mau Coal Mine, Vang Danh Coal Mine and No. 86 Enterprise Coal Mine together with No. 91 Enterprise, Phan Me and Kushiro coal samples shown in Figures 5-2, 5-3 and 5-4, respectively. All the curves shown in Figure 5-5 are the results of same “Initial Temperature (= 80 °C)” and “Sample Particle Size (-500μm)”.

From those curves shown in Figures 5-2, 5-3, 5-4, and 5-5, we can recognize as follows:

- 1) Some coal samples need longer time to increase the temperature up to 100 °C or 125 °C. ---- “Incubation time” is longer.
- 2) Most of coal samples show temperature increase at an “adiabatic condition” of SIT-2 due to oxidation process.
- 3) Many of samples show very steep temperature increase after around 150 °C.
- 4) Some samples show milder temperature increase even after around 150 °C.
- 5) In case of bituminous coal from Kushiro Coal Mine, it shows much steeper temperature increase after around 125°C than anthracite coal.

5.3.2 Effect of “Initial Temperature”

From the test results shown from Figure 5-2 to Figure 5-5 and Table 5-1, the effects of the initial temperature are discussed. In Figures from 5-2 to 5-4, the test results of initial temperatures

from 40 °C to 110 °C are shown. From the comparison of the data of the same sample as well as from the comparison between different coal mine samples, it might be possible to summarize the following:

- 1) In cases of lower initial temperature, it takes a longer time for starting to increase the temperature of coal sample. This tendency is notable in the cases for the initial temperatures are 40 or 50 °C.
- 2) Even in cases of higher initial temperature, some samples do not show increase of the temperature after several hundred minutes (4 to 17 hours). Namely, as shown in Table 5-1 and other figures, even under very high initial temperature of 110 °C, Phan Me coal and No. 91 Enterprise coal show probability of 1/2 and 2/3 temperature increase, respectively.
- 3) Some coal sample show faster temperature increase even it is started from lower initial temperature.
- 4) Once the temperatures increase up to 125 °C or so, the temperature increase rates are very similar because the temperature increase curves have in parallel each other especially in case of Phan Me coal and Kushiro coal.
- 5) In case of No. 91 Enterprise coal, there may be two or more groups of different characteristics, namely, group of “911205-1” and “911405-1” shows different temperature increase rates than others at higher temperature range more than 125 °C.
- 6) Some coal samples such as No. 91 Enterprise coal show immediate temperature increase with much less “incubation time” even from lower temperature. On the contrast, some coal samples show much longer “incubation time” such as Phan Me coal and some times more than 600 minutes (10 hours) or more.
- 7) From Figure 5-6, the coal samples from coal mines without problem of spontaneous combustion at the present time show some similar temperature increase rate with those from coal mines with the problems of spontaneous combustion.

There were so many differences in the incubation time for coal samples in the case of different initial temperature. Even in the case of the same coal sample, the incubation time was also different. In order to set up the initial temperature for the next experiment, the initial temperature could be selected at 75 °C for all coal samples. However, the effect of the initial temperature should be considered more by comparing the succession of each experiment on the same coal sample in the future experiment.

5.3.3 Effect of Particle Size of the Coal Sample

As shown in Table 5-2 we carried out experiments by changing the particle size of No. 91 Enterprise coal samples for several times as well as we conducted three experiments by using coal samples of Kushiro and Phan Me at the size between -125 μm and + 65 μm .

In Figure 5-6, “time-temperature curves” of each coal sample particle size of No. 91 Enterprise coal are shown. Except for coal sample particle size, other experimental conditions were the same. From the results shown in Figure 5-6 and Table 5-2, it might be impossible to find out some particular effect of particle size of coal sample. Namely, significant effects of the particle size are not recognized, for example, even smaller particle size sample needed longer “incubation time” and even coarse larger size samples showed faster start of temperature increase. In case of Phan Me coal, temperature increase at even very small particle size between -125 μm and + 65 μm was not observed as shown in Table 5-2.

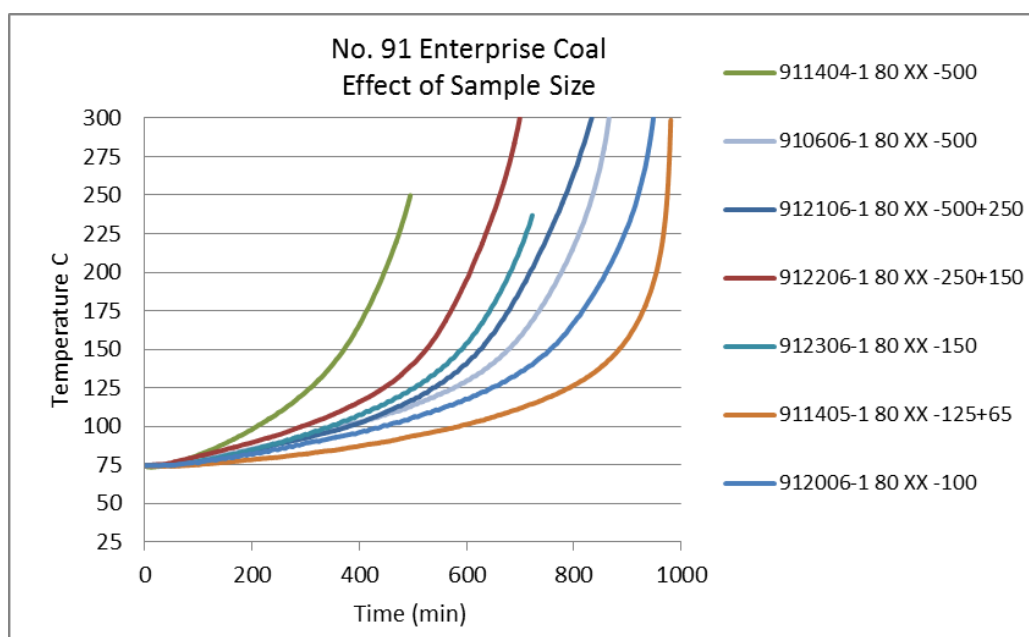


Figure 5-6: Temperature – Time curves of No. 91 Enterprise coal samples of different particle size

From the common sense, it might be thought that “smaller article size coal sample shows higher reactivity than coarser samples with oxygen because the relative specific surface area becomes higher, inversely proportion with the particle size”; however, some other factors might have a significant role for coal particle oxidation such as moisture of coal that affects oxygen contained gas penetration through the gap of sample particle.

From the experimental results and consideration above, it might be possible to conclude that “within the range of particle size -500 μm , there might not be so much effect on the oxidation process of coal sample.” (This hypothesis might be also supported by the analysis based on the Arrhenius’ theory shown in the next part.)

Table 5-2: Summary table of experiments by SIT-2 (1) --- Effect of coal sample size

No	Size	<500	500-250	250-150	>150	<150	<120+65	<500+150	Total
1	Kushiro	7/11			1/3	0/2			8/16
2	Phan Me	8/17			0/2	0/1	0/2	0/2	8/24
3	Khanh Hoa	22/29					0/3		22/32
4	No. 91 Ent.	53/73	1/2	1/3		1/1	1/2		57/81
5	No. 618 Ent.	1/1	1/1					1/1	3/3
6	Hong Thai	14/21							14/21
7	Mao Khe	32/37							32/37
8	Uong Bi	1/4							1/4
9	Vang Danh	4/5							4/5
10	Nam Mau	8/15							8/15
11	Ha Lam	10/18							10/18
12	Giap Khau	4/4							4/4
13	Ha Rang	2/2							2/2
14	Duong Huy	5/5							5/5
15	No. 86 Ent.	1/1							1/1
16	Quang Hanh	50/59							50/59
17	Thong Nhat	5/6							5/6
18	Cam Thanh	0/1							0/1
19	Mong Duong	4/6							4/6
20	Khe Cham	5/5							5/5
Total		236/320	2/3	1/3	1/5	1/4	1/7	1/3	243/345

5.3.4 Selection of Reasonable Test Conditions

Of course, the spontaneous combustion at a coal mine site starts from ambient temperature of 30 °C or 40 °C so that it might be better to examine the susceptibility of spontaneous combustion of coal sample from usual room temperature; however, as shown in Figures 5-2 to 5-4, it takes much longer time to get the results of the tests.

On the contrary for some coal samples such as No. 91 Enterprise coal showed earlier oxidation reaction even at the range less than 100 °C as shown in Figure 5-2.

As one of the other factors of the relevant experiments, we have to think about the time for completion of each experiment, because there are many coal samples to be examined from all the coal mines as well as from all the coal seams.

From the above mentioned three factors, we have to make a reasonable compromise for the experimental conditions by using SIT-2. Together with the considerations above, it might be possible to set the experimental conditions as shown in the Table 5-3 for the purpose of evaluation of “susceptibility of spontaneous combustion of coal” from every coal mine:

Table 5-3: Suggested experiment condition of SIT-2

Item	Experiment condition
Initial Temperature	75 °C (= Oven temperature: 80 °C)
Particle size of coal sample	-500 μm
Measuring temperature range	Up to 250 °C or 300 °C
Waiting time for “incubation”	10 hours (600 minutes)
Moisture of coal sample	No dry process (natural moisture)
Gas 2 of SIT-2 (oxidation gas)	Oxygen
Oxygen flow rate of SIT-2	2.5 ml/min (first 30 min) then 1.5 ml/min

5.4. Discussion on susceptibility of coal to spontaneous combustion

5.4.1 Arrhenius’ Equation

As discussed in 5.3.1, all the temperature increase curves shown in Figure 5-6 look like exponential due to “chemical oxidation reaction of coal”; however, from the comparison of those curves, it might be difficult to compare the susceptibility to spontaneous combustion of each coal sample, because most of the curves look very similar except for “incubation (retention) period” at lower temperature and overall shape of the curve.

In addition, even the coal sample that comes from the coal mine with the problem of spontaneous combustion such as Phan Me coal has a very long incubation (retention) period. In case of two Phan Me coal samples shown in Figure 5-6, it took 21 hours (1,268 min) and 42 hours (2,527 min) for increasing the temperature by 25 °C from the initial temperature, 75 °C to 100 °C.

To analyze those data, it might be very useful in introducing the idea shown by the “Arrhenius’ Equation” ---- Equation (5-1) shown below, because this equation can be applied to the general chemical reactions including oxidation process of coal.

$$k = A \exp (-E/RT) \quad (5-1)$$

where, A and E are so called as “frequency factor” and as “apparent activation energy” respectively, both of which might be expressed as a function of temperature. R and T are “gas constant” ($R = 8.31451 \text{ J/k}\cdot\text{mol}$) and “absolute temperature” of the reaction sample respectively. If we take natural logarithm of the both side of Equation (5-1), we can derive Equation (5-2):

$$\ln(k) = \ln(A) - E/RT \quad (5-2)$$

From this consideration, the experimental data of oxidation reaction temperature increase shown in Figure 5-6 might be analyzed by taking “differential gradient” of the temperature increase curves. Some part of this idea (at only 190 °C and 237 °C) was introduced by a Polish scientists, Olpinski and Struminki, which is adopted into Polish Standard.⁵⁻³⁾

5.4.2 Reaction rate Constant, k

Through the test by SIT-2, it is possible to get a continuous temperature increase curve under the adiabatic oxidation reaction from a lower initial temperature up to 250 or 300 °C so that we can derive continuous reaction rate represented by the time needed to increase a constant temperature. From the practical purpose and easier data handling, we calculated the time (min) needed for increasing the temperature by each 25°C from 75 °C up to 250 or 300 °C and based on the idea of Equation (5-2), we took the natural logarithm of those values, which represent the value “ $\ln(k)$ ” in Equation (5-2).

So, if we assume that all these oxidation processes follow the Arrhenius’ Equation, we may be able to analyze those data by the following:

- (1) “Chemical reaction rate constant”, k can be expressed by the temperature increase rate (°C/min) of the curves shown in Figure 5-6.
- (2) For the convenience of practical analysis, we get the temperature increase rate (°C/min) from the time, dT (min) needed to increase the coal temperature by 25 °C.
- (3) “Frequency factor”, A is constant within the temperature range of 25 °C.

From the assumption above, “Chemical reaction rate constant”, k can be regarded empirically as shown in Equation (5-3):

$$k \approx 25/dT \text{ (°C/min)} \quad (5-3)$$

Namely, $\ln(25/dT)$ may be able to represent $\ln(k)$ analogously in Equation (5-2). From the discussion on Arrhenius’ equation as well as assumptions above, by plotting a graph between $\ln(k)$ versus temperature at each corresponding temperature, we could get graphs shown in Figures 5-7 and 5-8. From Figure 5-7, coal samples from Mao Khe Coal Mine [Sample (1)], No. 91 Enterprise Coal Mine [Sample (3)], Hong Thai Coal Mine [Sample (4)] and Vang Danh Coal Mine [Sample (5)] had higher values of $[\ln(k)]$ than others in the temperature range less than 150 °C. Taking account for each coal mine situation, it might be possible to conclude that the coal sample from a coal mine with spontaneous combustion problem shows higher value of $\ln(k)$ at lower temperature range than others, namely, such coal samples had faster and easier oxidation reaction from ambient temperature.

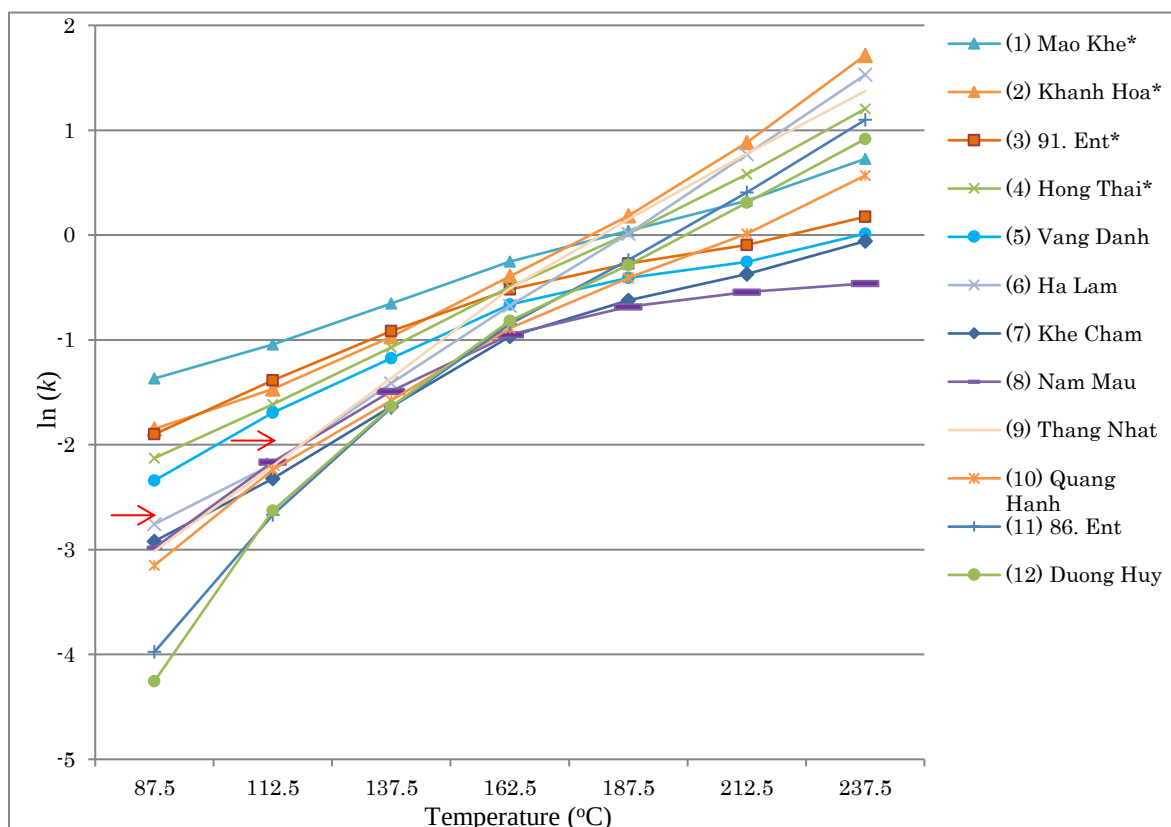


Figure 5-7: $\ln(k)$ versus temperature of anthracite coal samples
Red arrows show average value of $[\ln(k)]$ at 87.5 $^{\circ}\text{C}$ and 112.5 $^{\circ}\text{C}$.

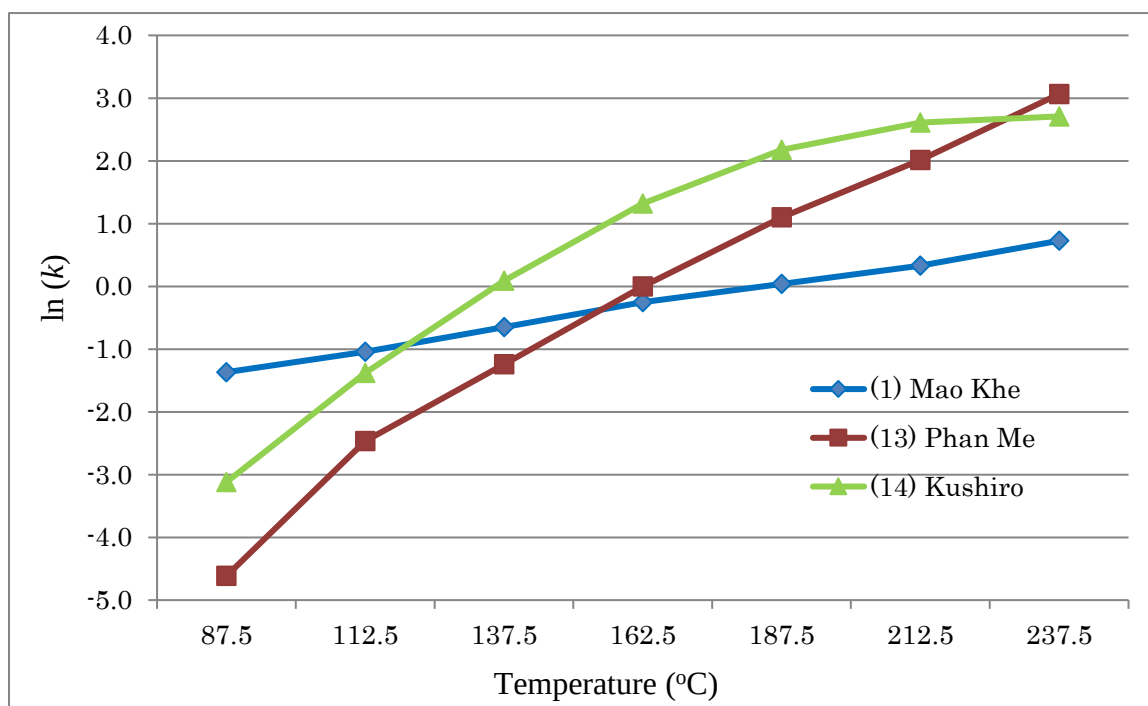


Figure 5-8: $\ln(k)$ versus temperature ---- comparison between anthracite [(1) Mao Khe] and sub-bituminous [(13) Phan Me] and bituminous [(14) Kushiro]

Besides that, the oxidation of anthracite coal was quite different from bituminous coal when comparing the results in Figure 5-8. In the case of anthracite coal, the rate of oxidation reaction was much lower at higher temperature range more than 150 °C comparing with bituminous or sub-bituminous coal as shown by the curves of Kushiro [Sample (14)] and Phan Me [Sample (13)] coal samples.

5.4.3 Proposal for Classifying the Susceptibility of Spontaneous Combustion

In Figure 5-7 examples of analysis of the test results of 12 coal samples by SIT-2 are shown. Among these 12 coal mines, five coal mines have spontaneous combustion problems up to date as shown in Table 5-4.

Table 5-4: Used coal samples for experiments

Sample No.	Coal mine	Coal Seam No.**	Kind of Coal	Spontaneous combustion
1	Mao Khe	No. 10	Anthracite	Yes*
2	Khanh Hoa	No. 16	Anthracite	Yes*
3	91 Ent.	No. 5	Anthracite	Yes*
4	Hong Thai	No. 12, No. 18	Anthracite	Yes*
5	Vang Danh	No. 8	Anthracite	No
6	Ha Lam	No. 7	Anthracite	Yes*
7	Khe Cham	No. 12	Anthracite	No
8	Nam Mau	No. 7	Anthracite	No
9	Thong Nhat	No. 6	Anthracite	No
10	Quang Hanh	No. 7	Anthracite	No
11	86 Ent.	No. 8	Anthracite	No
12	Duong Huy	No. 5	Anthracite	No
13	Phan Me	-	Sub-bituminous	Yes*
14	Kushiro (Japan)	-	Bituminous	Yes*

Note 1 *: “Yes” means that the coal mine has spontaneous combustion problem up to date.

Note 2 **: Because the coal mines shown in the Table 5-4 is widely spread within the Quang Ninh Coal Field, even if shown by the same coal seam No., those are completely different coal seam each other.

Namely, the coal mines shown by “Yes” in Table 5-4 have problems of spontaneous combustion up to date. Taking account for the facts shown in Table 5-4 and the discussion above, it might be possible to set a boundary tentatively by which we can divide the susceptibility (liability) to spontaneous combustion of the coal sample. This boundary may be set empirically by the concept shown below:

- All the data of the coal samples from coal mines without spontaneous combustion problem belong in the area of “Less liability”.
- At least some of the data of the coal samples from coal mines with spontaneous combustion problem belong in the area of “Higher liability”.
- Boundaries can be set at a lower temperature zone less than 150 °C in which coal samples from coal mines with spontaneous combustion show higher values of $\ln(k)$ [higher oxidation rate].

This tentative boundary is shown by red arrows in Figure 5-7, which are derived from average $[\ln(k)]$ values of all measured values [$\ln(k) = -2.72$ at 87.5 °C, and $\ln(k) = -1.97$ at 112.5 °C]. From the shown boundary (two arrows) of Figure 5-7, it may be possible to understand that if the plotted data of a coal sample are locate at “Higher Value” zone than the places of two arrows, the test coal sample has “higher susceptibility (liability) to spontaneous combustion”, otherwise, the test coal sample has “less susceptibility (liability) to spontaneous combustion”. If this tentative proposal of “the boundary” is acceptable, we may classify the coal seam into three groups of “susceptibility (liability) to spontaneous combustion” as shown in Table 5-5.

Table 5-5: Examples of proposed classification of “susceptibility to spontaneous combustion”

Class	Rank of Susceptibility to Spontaneous Combustion	Example of relevant coal mines
I	High	(1) Mao Khe, (2) Khanh Hoa, (3) 91 Ent., (4) Hong Thai, (5) Vang Danh
II	Medium	(6) Ha Lam, (7) Khe Cham, (8) Nam Mau, (9) Thong Nhat, (10) Quang Hanh, (11) No. 86 Enterprise, (12) Duong Huy, etc.
III	Low	Cam Thanh, etc.

Because, every coal has potential of “spontaneous combustion” ultimately as well as there might be no clear boundary, it might be better to classify the “susceptibility (liability) to spontaneous combustion” into three groups of “high”, “medium” and “low” as shown in Table

5-5. In this case we classified the coals into Class III, “Low” “susceptibility (liability) to spontaneous combustion” that did not show any temperature increase at the tests by using SIT-2 at least for the period of more than 10 hours like Cam Thanh. Namely, all the group that are located less values than the average values of $[\ln(k)]$ (two red arrows) are classified onto “medium”.

In summary, our proposal can categorize the susceptibility into three as follows:

- (1) Class I - “High”: Oxidation rate, $[\ln(k)]$ are higher than the average values of all measured oxidation rate, $[\ln(k)]$ at lower temperature range less than 150 °C (at 87.5 °C and 112.5 °C).
- (2) Class II - “Medium”: Oxidation rate, $[\ln(k)]$ are lower than the average values of all measured oxidation rate, $[\ln(k)]$ at lower temperature range less than 150 °C (at 87.5 °C and 112.5 °C).
- (3) Class III - “Low”: No temperature increase is observed through the test by using an adiabatic oxidation process analyzer (SIT-2) at least for a period of 10 hours.

5.5 Conclusion

In relation with spontaneous combustion properties of anthracite coal from the analysis of oxidation reaction data of anthracite under adiabatic condition and comparison of those data with the site data from coal mines with spontaneous combustion problems, we derived the following conclusions:

- (1) Anthracite coal samples from coal mines with problems of spontaneous combustion showed higher oxidation reaction rate at lower temperature range less than 150 °C than other coal samples come from coal mines without spontaneous combustion problems.
- (2) If those anthracite data with higher susceptibility to spontaneous combustion are compared with the data of bituminous coal and sub-bituminous coal with higher susceptibility to spontaneous combustion, the oxidation reaction rate reverses at the temperature range around 150 °C. Namely, in case of anthracite, oxidation reaction rate is much higher than that of bituminous or sub-bituminous at lower temperature range; however, those data reverse completely at higher temperature range.
- (3) At higher temperature range more than 150 °C, there is not so much difference in oxidation reaction rate of anthracite regardless of higher susceptibility to spontaneous combustion of coal sample.
- (4) The relative classification of coal based on its susceptibilities to spontaneous combustion is influenced by the fact of coal spontaneous combustion at mine site of Vietnamese coal mines. For more accurate determination, some effects from geological condition and coal

characteristic should be considered because those parameters have significant roles for initiation of spontaneous combustion.

- (5) From the conclusion shown above, it might be possible to explain why Polish method is not appropriate for estimating the susceptibility of anthracite to spontaneous combustion, because the Polish method is based on the activation energies at 190 °C and 237 °C only and the oxidation reaction rate [$\ln(k)$] of anthracite in such higher temperature range show much less values than those of bituminous or sub-bituminous. In other words, the susceptibility to spontaneous combustion of anthracite can be found at much less temperature range [less than 150 °C] than the standard of the Polish method.

Reference

5-1) Le Trung Tuyen, K. Ohga, T. Isei. Susceptibility to spontaneous combustion of Vietnamese anthracite. *Journal of Mining and Minerals Processing Institute of Japan*, Vol. 133, No. 6, pp. 140-150, 2017.

5-2) Idemitsu Kosan Co., Ltd., “Basis of coal”, 2013 [Japanese: 出光興産(株)、「石炭の基礎」、2013年1月22日、pp. 35 – 38] (http://www.jcoal.or.jp/coaldb/shiryo/material/01_ando.pdf)

5-3) Poland Standard, PN-93/G-04558, 1994.

Chapter 6: Evaluation of Other Effects on the Susceptibility of Anthracite Coal to Spontaneous Combustion⁶⁻¹⁾

6.1 Effect of porosity of coal on oxidation process⁶⁻¹⁾

As shown in the previous paper⁶⁻²⁾, all the coal mines that have problems of spontaneous combustion are located nearly west end part of Quang Ninh Coal Field belt expanding 250 km long from west to east and 30 km wide from south to north. To find out the reason of regionally ubiquitous occurrence of spontaneous combustion, we conducted porosity measurements⁶⁻³⁾ of eight coal samples^{*1} by using an established method of inert gas absorption⁶⁻⁴⁾. By these measurements, pore surface area (t-Plot micropore area⁶⁻⁵⁾) and pore volume of unit weight (t-Plot micropore volume⁶⁻⁵⁾) as well as pore size (BJH adsorption average pore width⁶⁻⁵⁾) of each coal sample were derived. Measured results are shown in Table 6-1 by arranging the data in descending order of numerical values of three measurement items, respectively.

Table 6-1: Porosity measurement results of eight coal samples

Pore surface area (m ² /g)	Hong Thai*	Nam Mau	Khanh Hoa*	91 Ent*	Quang Hanh	Ha Lam*	Thong Nhat	Duong Huy
	2.18	0.96	0.96	0.78	0.72	0.62	0.26	0.16
Pore volume (cm ³ /g x 10 ⁻³)	Hong Thai*	Nam Mau	Khanh Hoa*	91 Ent*	Quang Hanh	Ha Lam*	Thong Nhat	Duong Huy
	0.990	0.427	0.390	0.351	0.321	0.288	0.126	0.083
Pore size (mm)	Hong Thai*	Khanh Hoa*	91 Ent*	Ha Lam*	Nam Mau	Quang Hanh	Thong Nhat	Duong Huy
	25.37	21.13	20.32	16.16	12.08	10.97	7.53	6.91

(The coal mines shown with * mark have problem of spontaneous combustion up to date.)

In case of “pore size”, four coal mines of Hong Thai, Khanh Hoa, No. 91 Enterprise and Ha Lam with problems of spontaneous combustion occupied top four among eight coal samples. In case of “pore surface area” and “pore volume” the three coal mines were within top six; Hong Thai at first, Khanh Hoa at third and No. 91 Enterprise at fourth as well as Ha Lam at sixth. From these results, the pore size might have stronger effects on oxidation process of anthracite coal.

^{*1} Measurements were conducted at Hanoi National University of Education by using an established method for activated carbon. ⁶⁻⁵⁾

On the other hand, coal samples from east end coal mines such as Duong Huy, Thong Nhat and Quang Hanh located near east end of the coal field showed much smaller values of all items. Nam Mau and Ha Lam are located in rather central side of the coal field. From these data, it might be possible to conclude that coal samples from the west part show larger values of three measured items and coal samples from east part showed smaller values. Such tendency agrees well with apparent properties that coals of west part field are soft and easy to crush, different from hard coals from east part, some of which are used for carving for souvenirs.

From such measured results, it might be possible to conclude that easy start of spontaneous combustion of some west part coal mines is caused by higher porosity of the coal itself.

From Table 6-1, it is possible to deduce that higher porosity value coal shows higher susceptibility to spontaneous combustion. From this deduction, correlation between the porosity and oxidation rate [$\ln(k)$], which is discussed in Chapter 5, was examined. The results are shown in Figure 6-1, in which relation between pore size of coal sample and apparent oxidation rate, $\ln(k)$ at the coal temperature 87.5 °C and 112.5 °C are shown.

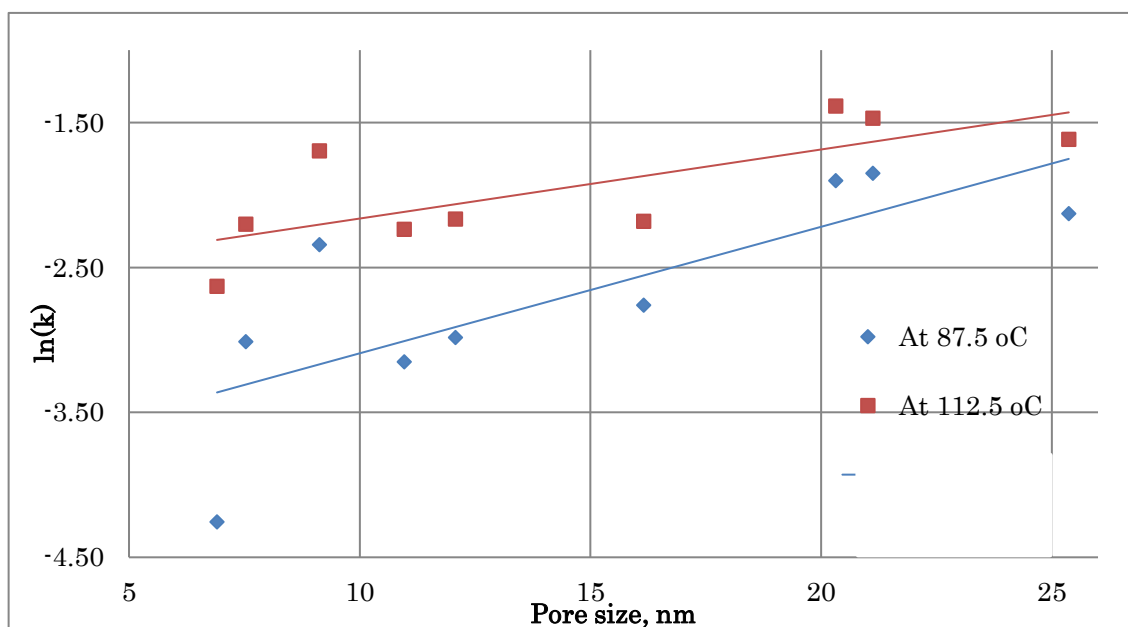


Figure 6-1: Relationship between pore size of coal sample and apparent oxidation rate [$\ln(k)$]

From the results shown in Figure 6-1, there might be positive direct proportion correlation regardless of the variations of data. Namely, higher pore size gave higher oxidation rate. From overall deduction of Table 6-1 and Figure 6-1, higher porosity might give higher susceptibility to spontaneous combustion of anthracite coal.

6.2 Effect of original moisture of coal on oxidation process

6.2.1 Adiabatic oxidation test

Conceptual longitudinal cross section diagram of an “adiabatic spontaneous ignition tester” (SHIMAZU SIT-2) is shown again in Figure 6-2 (see also Figure 5-1 and the explanation in Chapter 5). An essential point of this system is to set temperatures sensors of the sample coal, T_c inside the sample cell and the oven, T_o equal before introducing oxygen or air for oxidation process of the coal sample.

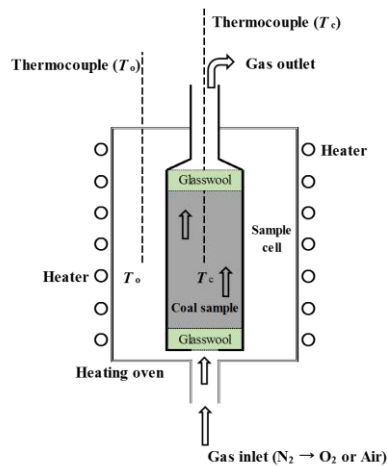


Figure 6-2: Conceptual diagram of an “adiabatic spontaneous ignition tester”

When the sample cell with test coal sample (about 1,000 mg) is mounted inside the oven for the test, both temperatures T_c and T_o are not equal. So, to make coal sample temperature T_c equal to the initial oven temperature T_o (initial test temperature), inert N_2 gas is introduced from the gas inlet at the bottom of the oven with a constant flow rate such as 1.5 ml/min until T_c becomes equal to T_o for around 30 minutes or so. Then after achieving $T_c = T_o$, N_2 gas is alternated to O_2 gas or air at the same flow rate so as to make the sample start oxidation process. From this point, oxidation process of the coal sample can be observed by measuring the change of temperature T_c .

6.2.2 Examples of temperature increase curves due to oxidation process of coal samples

Some experiments were conducted by the following test conditions:

- Coal sample (size: less than 500 μm) = about 1,000 mg
- Initial oven temperature = 80 $^{\circ}\text{C}$
- Temperature stabilization time by introducing N_2 gas = 30 min
- Gas (N_2 or O_2) introduction flow rate = 1.5 ml/min

Extracted some test data are shown in Figure 6-3. Among these seven curves, Nam Mau

Coal Mine [curve (4)] and Duong Huy Coal Mine [curve (6)] had no spontaneous combustions up to date. Other five curves were all derived by the coal samples from coal mines with problems of spontaneous combustion including bituminous [curve (2) – Kushiro Coal Mine in Japan] and sub-bituminous [curves (5) and (7) – Phan Me Coal Mine] as well as anthracite [curve (1) - Hong Thai Coal Mine and curve (3) - No. 91 Enterprise Coal Mine]. Regardless of higher liability to spontaneous combustion of anthracite coal sample, it took much longer time to start active oxidation process in the test system sometimes more than 2,700 minutes (45 hours) as shown by the curve (7) in Figure 6-3. And even in case of the same samples, there were big differences in the active oxidation process as shown curves (5) and (7), namely, when we compare the time needed to increase the coal sample temperature from the start temperature (80 °C) up to 100 °C, it took 21.1 hours in case of curve (5) and 42.1 hours in case of curve (7), respectively.

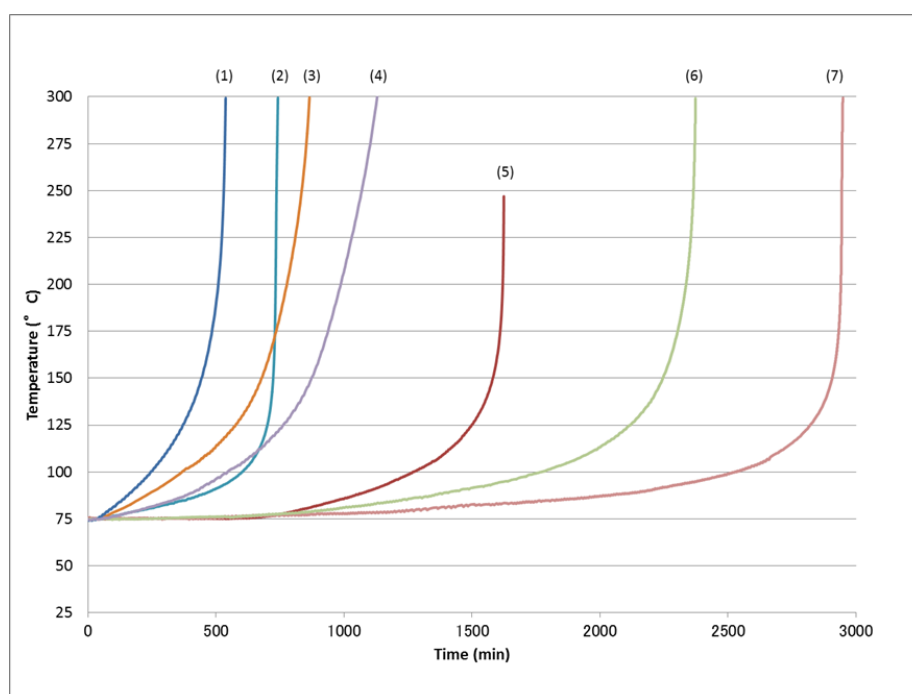


Figure 6-3: Temperature-Time curves of adiabatic oxidation process of coal samples

(1) Hong Thai, (2) Kushiro (Bituminous), (3) No. 91 Enterprise, (4) Nam Mau,
(5) Phan Me (Sub-bituminous), (6) Duong Huy, (7) Phan Me (Sub-bituminous)

All the results shown in Figure 6-3 were observed through the same test conditions shown above. During temperature stabilization process between the coal sample cell and oven by introducing N₂ gas for 30 minutes, in most cases, water condensation at the inner surface of gas outlet plastic tube was observed as shown in Figure 6-4. In all the tests shown in Figure 6-3, the initial oven temperature was kept at 80 °C, so some of moisture of the coal sample inside the sample cell was evaporated and carried by introduced N₂ gas toward the outlet tube kept around room temperature at which water vapor was condensed just at the outlet part from the sample cell.

Because coal sample is subjected to the test of SIT-2 similar to the condition at the mine sites, all the samples might have some extent of higher moisture including adherent free moisture more than the moisture content value derived through proximate analysis as shown in Table 6-2. Namely, coal sample was kept inside a small container or plastic bag just after sampling at mine site and the sample was transferred into a small plastic container to which N_2 gas was injected at laboratory for storage. In other words, the sample was not treated by air dry process subjected to proximate analysis.



Figure 6-4: Water condensation during temperature stabilizing process by introducing N_2 gas

Table 6-2: Examples of moisture content of coal samples derived by proximate analysis

Coal sample	Moisture (%)
Hong Thai	0.73
No. 91 Enterprise	1.32
Nam Mau	1.49
Duong Huy	0.87

When we set coal samples for some period for stabilizing $T_c = T_o$ by introducing N_2 gas, moisture evaporation from coal sample is inevitable at any initial oven temperature. This is because supplied N_2 gas is completely dry basis due to supply from a gas cylinder.

From this fact, the coal sample subjected to the test might be changed in the moisture from the original state before starting oxidation process. From this consideration, some additional experiments were conducted by reducing the N_2 gas introduction period as shown below.

6.2.3 Test results by reducing the time of N_2 gas introduction

Four kinds of anthracite coal samples were used to understand the effects of N₂ gas introduction period on the oxidation process of coal samples in the adiabatic test chambers. Test conditions were the same as the ones shown above to derive Figure 6-3 except for changing the time of N₂ gas from 30 minutes down to 20, 10, 5 and 1 minutes, respectively. Used coal samples are shown in Table 6-3:

Table 6-3: Coal samples used for understanding effects of N₂ gas introduction time

Sample	Coal mine	Coal seam
A	No. 91 Enterprise	No. 5
B	No. 91 Enterprise	No. 4
C	Khanh Hoa	No. 16
D	Quang Hanh	No. 14

Among these four coal mines, only Quang Hanh Coal Mine (Sample D) does not have spontaneous combustion problem up to date. Other two coal mines had experiences of spontaneous combustion for several times up to date.

For each coal sample, the tests were conducted from two to four times to confirm the reliability at each test condition. Some extracted test data of the Coal Sample A are shown in Figure 6-5, in which one representative time-temperature curve for each N₂ gas introduction period is shown. Apparently it is clear from Figure 6-5 that when the N₂ gas introduction period became longer, the time needed to increase the coal sample temperature by oxidation process up to a certain temperature became longer, namely, shorter exposure of coal sample to N₂ gas resulted in earlier time increase of temperature of the coal sample due to oxidation.

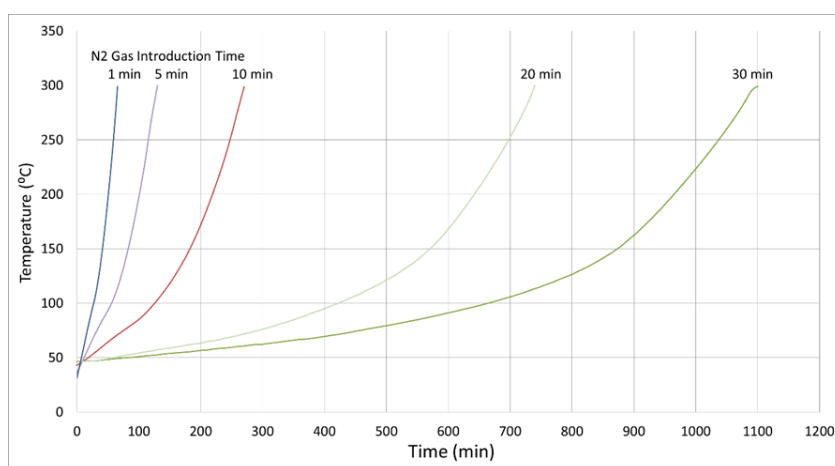


Figure 6-5: Temperature increase curves at each N₂ gas introduction period before introducing O₂ gas: Coal Sample A

In Figure 6-5 only one representative test curve from two or four tests is shown for each N₂

gas introduction period so that it is impossible to check the dispersion of the data obtained in the same test conditions. To confirm the fluctuation of these data under the same test conditions, some extracted data are summarized from all the experiments in Table 6-4, in which time needed to go up the coal temperature up to from 100 to 300 °C by 50 °C step is shown for the same Coal Sample A with the cases of Figure 6-5.

**Table 6-4: Time needed to go up the coal temperature shown in the top column (min)
[Sample A]**

N ₂ Gas Int. Period	Test No.	Coal Sample Temperature				
		100°C	150°C	200°C	250°C	300°C
1 min	Test 1-1	32.3	48.7	59.9	69.3	77.1
	Test 1-2	27.5	45.8	57.1	65.1	71.6
	Test 1-3	27.0	41.2	50.8	58.8	65.5
5 min	Test 5-1	64.4	94.8	115.2	131.7	144.4
	Test 5-2	81.1	118.1	142.3	161.6	175.8
	Test 5-3	55.8	82.8	100.8	115.7	127.0
10 min	Test 10-1	160.4	229.2	271.0	302.8	326.4
	Test 10-2	125.8	182.7	218.9	247.3	269.5
	Test 10-3	141.3	201.9	240.3	268.7	289.4
	Test 10-4	142.3	204.9	243.2	272.0	293.5
20 min	Test 20-1	422.6	570.8	641.9	697.7	741.3
	Test 20-2	460.8	634.3	714.8	784.8	857.4
	Test 20-3	539.8	700.2	772.4	826.6	868.0
30 min	Test 30-1	711.0	929.6	1021.8	1095.2	1154.4
	Test 30-2	663.8	873.1	964.5	1036.9	1094.6
	Test 30-3	877.3	1126.3	1225.9	1305.8	1372.8

In case of N₂ gas introduction period of one minute, Test 1-3 showed the minimum time needed to go up to the specific temperature such as 27.0 minutes up to 100 °C, 50.8 minutes up to 200 °C and 65.5 minutes up to 300 °C, and Test 1-1 showed the maximum time of 32.3 minutes up to 100 °C, 59.9 minutes up to 200 °C and 77.1 minutes up to 300 °C. By comparison of the columns of the same N₂ gas introduction period group with the adjacent columns of next N₂ gas introduction period group, it is clear that the maximum time needed for the shorter N₂ gas introduction period group never exceeds the minimum time needed for the next longer N₂ gas introduction period group. Namely, Table 6-4 showed the same tendency with the results in Figure 6-5, that is, shorter N₂ gas introduction period results in earlier temperature increase of

coal sample due to oxidation process and longer N₂ gas introduction period results in slower temperature increase of coal sample.

Throughout all the experiments conducted by using four coal samples, the same tendencies were observed in Coal Samples of B, C and D without problems of spontaneous combustion up to date. Representative data from all four coal samples are summarized in Table 6-5, in which the time needed to go up 100, 150, 200 and 250 °C for each N₂ gas introduction time is shown. From Table 6-5, it is clear that the longer N₂ gas introduction time turns the longer time needed to increase the coal sample temperature in all coal samples of A, B, C and D.

Table 6-5: Change in required time to increase the temperature due to the change of N₂ gas introduction time

Coal Sample	N ₂ Gas Int. Period	Coal Sample Temperature			
		100 °C	150 °C	200 °C	250 °C
A	1 min	27.0	41.2	50.8	58.8
	5 min	55.8	82.8	100.8	115.7
	10 min	125.8	187.2	218.9	247.3
	20 min	422.6	570.8	641.9	697.7
B	1 min	19.3	30.3	39.6	46.0
	5 min	59.8	88.0	106.8	122.3
	10 min	101.8	146.6	175.6	198.3
	20 min	614.3	789.3	863.9	918.7
C	1 min	24.9	35.8	43.4	48.6
	5 min	43.1	66.8	83.5	93.6
	10 min	112.1	174.1	208.6	223.0
	20 min	281.6	408.2	463.0	483.4
D	1 min	26.4	37.8	45.7	51.8
	5 min	62.9	91.5	110.0	121.6
	10 min	100.4	147.1	175.8	192.8
	20 min	389.3	527.1	586.7	614.7

6.2.4 Discussion

(1) Effects of N₂ gas introduction period

As shown in Figure 6-5, Table 6-4 and Table 6-5, N₂ gas introduction period prior to start of O₂ gas introduction had distinct effects on the oxidation process of the coal sample in the sample cell in the adiabatic oxidation system. From this view point, correlation between N₂ gas introduction period and time needed to go up 100, 200 and 300 °C in case of Coal Sample A is

illustrated in Figure 6-6.

From this figure, both variables of N₂ gas introduction period and time needed to go up 100, 200 and 300 °C had a proportional relation. If we compare all coal samples used in this report in the same way as Figure 6-6, we can obtain Figure 6-7. From this figure, correlation between N₂ gas introduction period and time needed to go up to a certain temperature of coal sample is established clearly.

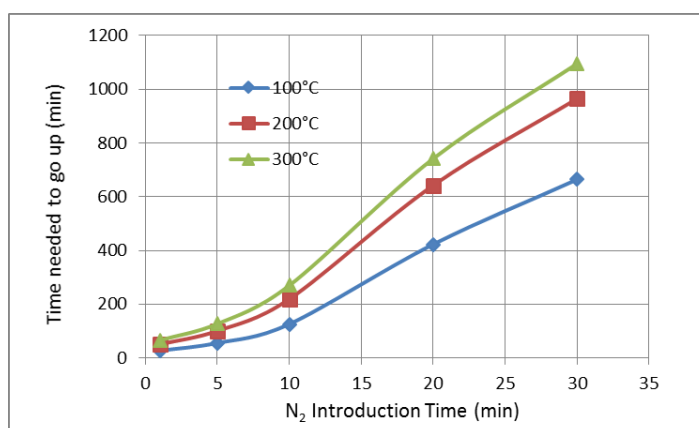


Figure 6-6: Relation between N₂ gas introduction period and time needed to go up 100, 200 and 300 °C (Coal Sample A)

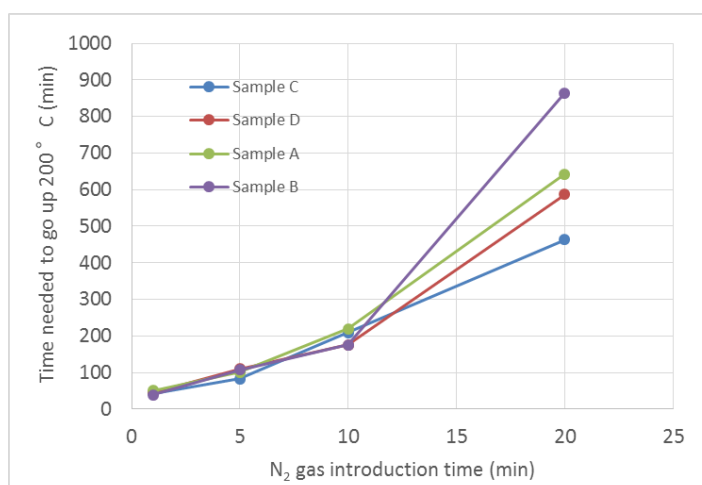


Figure 6-7: Relation between N₂ gas introduction period and time needed to go up 200 °C of each coal sample

(2) Effects of moisture in coal sample

As we discussed in relation with Figure 6-4 and Table 6-2, N₂ gas introduction might encourage evaporation of moisture of the coal sample in the sample cell of an adiabatic oxidation tester, SIT-2 shown in Figure 6-2. From this view point, we examined the weight reduction of coal sample inside the sample cell by the following way:

After measuring the weight of sample cell with coal sample, it was set inside the oven of the

adiabatic oxidation tester kept at a constant temperature at 50 °C as usual oxidation test and N₂ gas was introduced for a fixed period such as 5, 10, 20 or 30 minutes. After introducing N₂ gas at a fixed period, the sample cell was taken out from the oven and the weight of the sample cell with coal sample was measured after cooling it to room temperature. These measurements were conducted in a batch manner for each N₂ gas introduction period by using the same coal sample.

Measured weight loss of each coal sample at each the N₂ gas introduction period is shown in Figure 6-8. From this figure, the weight of the coal sample was reduced in proportion with the period of N₂ gas introduction and weight reduction ratios were different depending on the coal sample. From the results, it might be possible to deduce that the moisture inside the coal sample is evaporated into dry base N₂ gas flow through the coal sample inside of sample cell by receiving the heat from the oven.

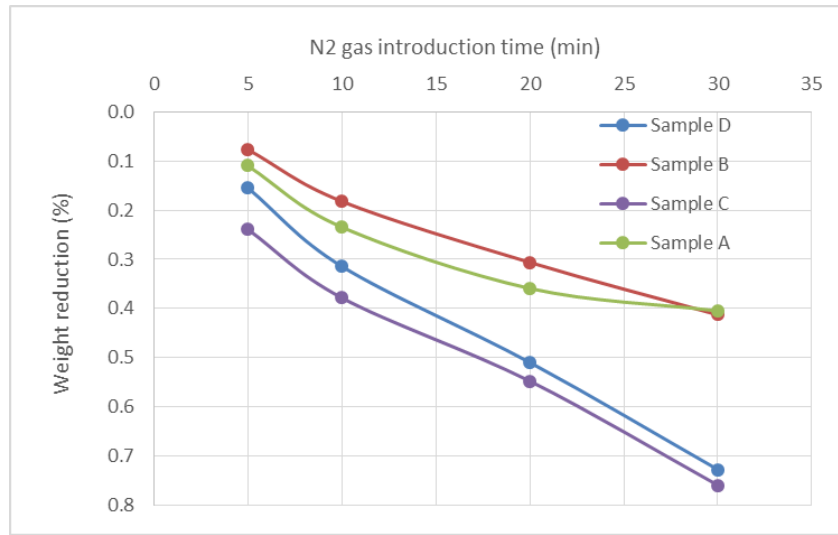


Figure 6-8: Weight reduction of coal sample in the sample cell due to introduction of N₂ gas

(3) Oxidation reaction rate

If we take finite differential inclination of the temperature-time curves shown in Figure 6-3 or Figure 6-5, it is possible to derive the variables that show oxidation reaction rate, which is deeply related with the reaction rate constant k shown by the Arrhenius's equation:

$$k = A \exp (-E/RT) \quad (1)$$

where, T : absolute temperature

R : Gas constant

E : apparent activation energy

A : Frequency factor

To take apparent reaction rate constant k , finite differential inclinations were derived at each 25 °C step from 50 °C to 300 °C for all the tests ($k \approx 25/dT$, dT (min): time needed to increase the temperature by 25 °C)⁶⁻¹⁾. An example of the cases of Coal Sample C for N₂ gas introduction

period of 1, 5, 10 and 20 min are shown in Figure 6-9. Some variations in the same N_2 introduction time group are shown; however, it is clearly shown that shorter N_2 gas introduction period gives larger relative oxidation rate $[\ln(k)]$ and greater differences in relative oxidation rate $[\ln(k)]$ at lower temperature converge to between $\ln(k) = 2$ and $\ln(k) = 3$.

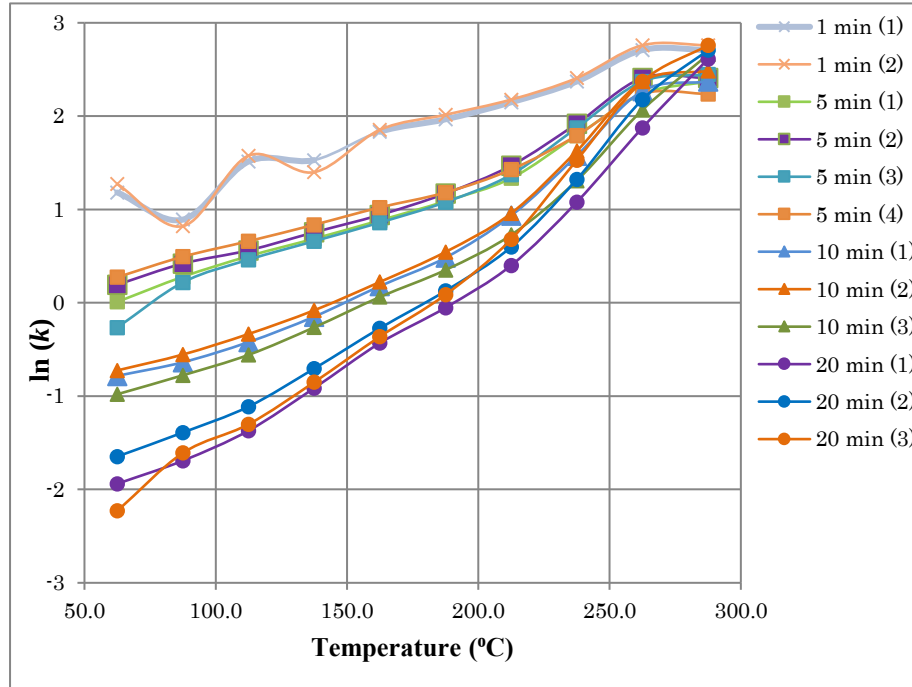


Figure 6-9: Apparent oxidation reaction rate $[\ln(k)]$ of Coal Sample C (Khanh Hoa) for N_2 gas introduction period, 1, 5, 10 and 20 minutes

Deducing the results of Figures 6-5, 6-6, 6-7, 6-8 and 6-9 as well as Tables 6-4 and 6-5, at the test of an adiabatic oxidation tester, SIT-2, it might be possible to conclude that moisture evaporation due to introduction of N_2 gas for stabilizing the temperature coal sample in the test cell and outside oven acts as reducing oxidation of coal sample in the cell, so that the oxidation rate decreases in proportion with the N_2 gas introduction period. This fact suggests that direct replacement from moisture to oxygen at micro pore surface of coal sample reduces the oxidation reaction rather than drying by N_2 gas coal pore surface on which N_2 gas might be absorbed during temperature stabilizing process.

(4) Effects of initial oven temperature

One important premise of the test condition by SIT-2 is to keep “apparent adiabatic condition” between the coal sample in a glass made cell and the oven enclosing the sample cell. For this purpose it is required to set temperature stabilization period by introducing N_2 gas up to get the condition of $T_c = T_o$.

From this premise, if we reduce the introduction period of N_2 gas, the temperature of coal

sample, T_c becomes lower than the temperature of oven, T_o ($T_c < T_o$) in case that the initial oven temperature is higher than the room temperature. From this effect, oxidation process starts under the condition that the coal temperature is lower than the oven temperature, which does not satisfy “apparent adiabatic condition”. In such a case, some heat is transferred from the oven to coal sample, which might encourage oxidation of the coal sample.

From such consideration, we conducted some tests that started from “room temperature” (around 27 or 30 °C) of both coal sample and oven in the test condition. Some examples are shown in Figures 6-10 and 6-11.

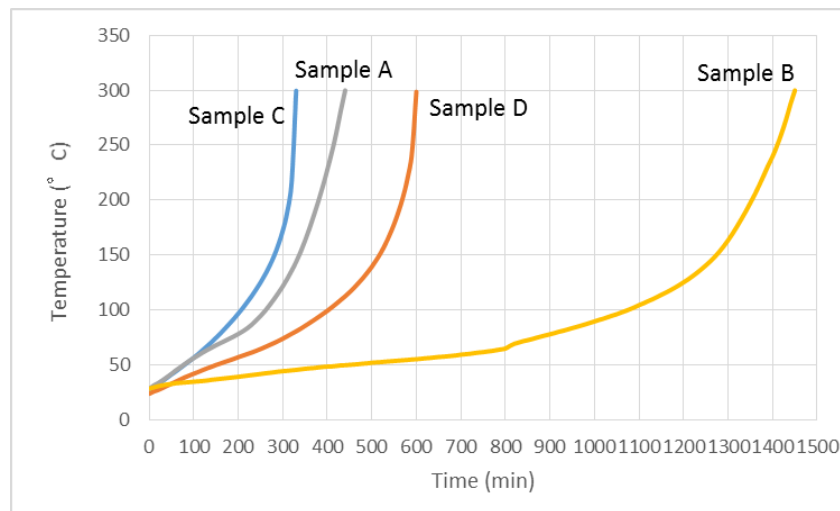


Figure 6-10: Temperature-time curve from room temperature of both coal sample and oven

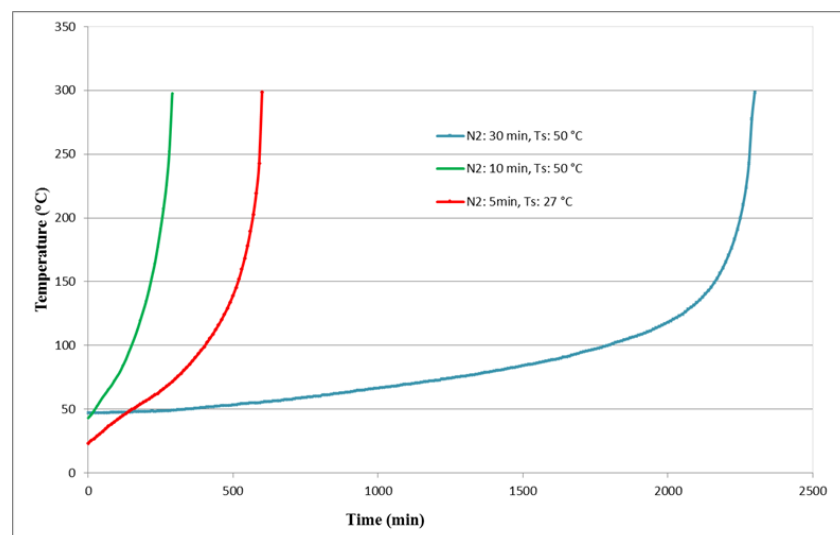


Figure 6-11: Comparison of temperature-time curves of different N₂ gas introduction period of same coal sample (Sample D)

Figure 6-10 shows comparison of temperature increase curves of (from left) Sample C,

Sample A, Sample D and Sample B, respectively. Here again it is clear that not necessarily the coal sample from a coal mine with problems of spontaneous combustion shows higher speed temperature increase rate due to oxidation. Namely, Sample B shows the slowest temperature increase rate than others including Sample D without spontaneous combustion up to date.

Figure 6-11 compares the temperature increase curves of Sample D, two of which are initial oven temperature were 50 °C with N₂ gas introduction periods of 30 minutes and 10 minutes, respectively and the other one was initial oven temperature of “room temperature” with N₂ gas introduction period of 5 minutes. From comparison of these three curves, it is again clear that shorter N₂ gas introduction period (lower evaporation of moisture from coal sample) showed much higher oxidation activities than longer N₂ gas introduction period (higher evaporation of moisture) including even oxidation process starting from room temperature.

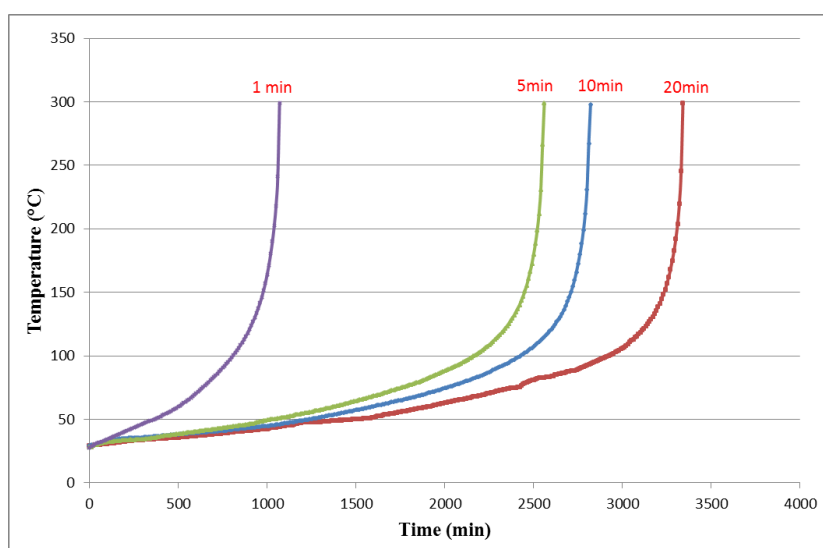


Figure 6-12: Comparison of temperature-time curves from room temperature of both coal sample and oven with different N₂ gas introduction period (Coal sample: D)

To make clear the effect of N₂ gas introduction time on the oxidation process in SIT-2 under the same test conditions, a series of experiments were conducted by using coal sample D from room temperature of both coal sample and the oven. Taking account for the effects of coal sample storage time⁶⁻¹⁾ after sampling at the mine site, experiments were conducted in the order from 20, 10, 5 and 1 minutes by using the same coal sample D, which was sampled on August 16, 2016. Coal sample storage times before subjecting to the experiment were 1, 4, 6 and 8 days after sampling for N₂ gas introduction time, 20, 10, 5 and 1 minutes, respectively. The results of the experiments are illustrated in Figure 6-12.

As shown in Figure 6-12, the time needed to go up to 300 °C became longer in the order of 1, 5, 10 and 20 minutes, respectively. Namely the longer N₂ gas introduction time resulted in longer

incubation time, respectively. Even if the trend that incubation time becomes longer for longer coal sample storage time⁶⁻¹⁾ is taken into account, the data show the shortest incubation time in case of 1 minute N₂ gas introduction, regardless of the longest sample storage time.

From the results shown in Figure 6-12, it might be possible to deduce that longer N₂ gas introduction time induces evaporation of the moisture of the coal sample in the sample cell of SIT-2 and might result in longer incubation time.

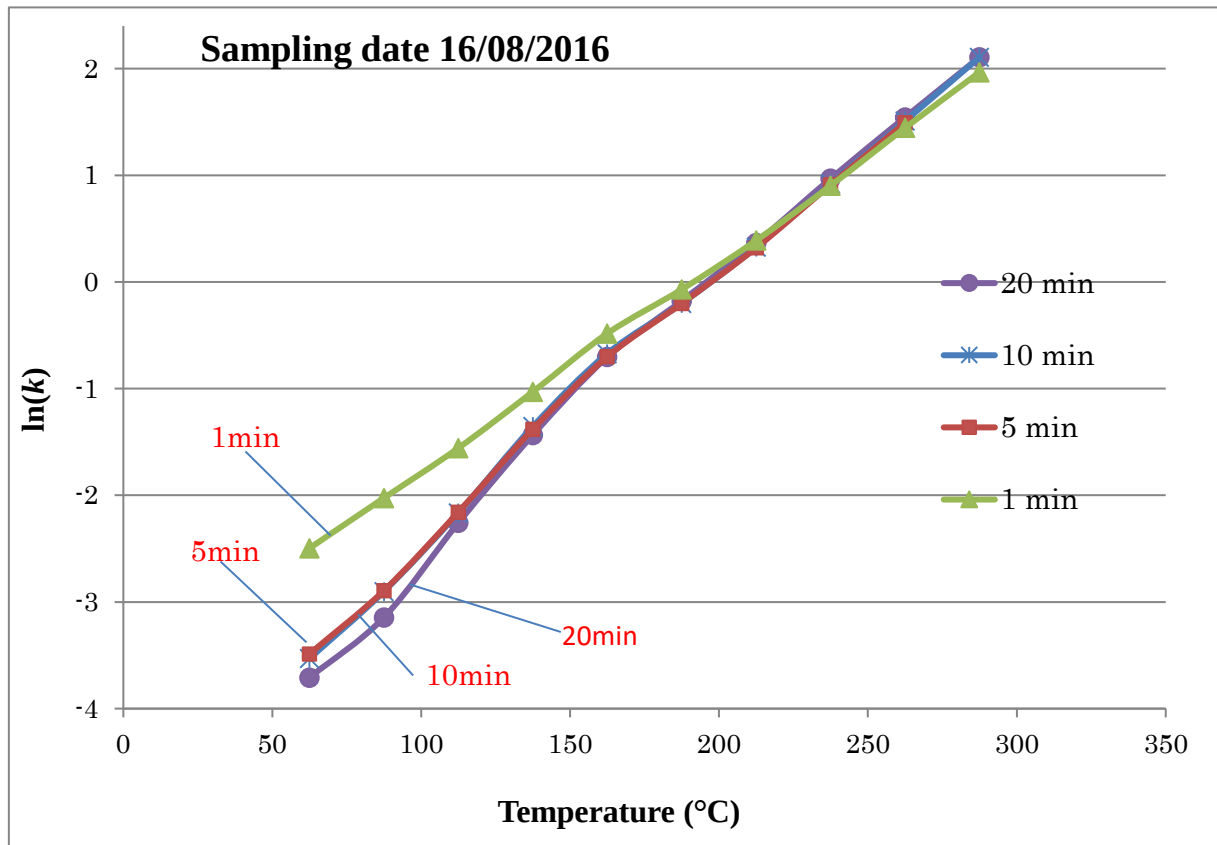


Figure 6-13: Apparent oxidation reaction rate [ln (k)] of Coal Sample D for N₂ gas introduction period of 1, 5, 10 and 20 minutes

The oxidation rate of each curve in Figure 6-12 is compared by using ln (k) like in Figure 6-9, as shown in Figure 6-13. At lower temperature range less than 150 °C, ln (k) for 1 minute N₂ gas introduction time showed much higher value than other three cases of 5, 10 and 20 minutes N₂ gas introduction times. Namely, it is clear that longer introduction of N₂ gas produces faster oxidation of coal sample in the sample cell in SIT-2. In other words, longer introduction of N₂ gas might change the oxidation activity of coal sample from the one collected at coal mine site.

From the results shown from Figure 6-5 to Figure 6-13 as well as Table 6-4 and Table 6-5, it might be possible to conclude that the original moisture of coal contributes to more active coal oxidation at lower temperature less than 150 °C even from ambient temperature where coal exists

in underground. Namely, original moisture of coal might promote oxidation of coal by replacing moisture inside the micro pore structure with oxygen from lower ambient temperature underground.

6.3 Effect of other factors on coal oxidation process

6.3.1 Oxygen absorption by anthracite at ambient temperature

As discussed in Figure 5-8 in Chapter 5, anthracite coal from the coal mine with problems of spontaneous combustion shows higher oxidation rate [$\ln(k)$] than other anthracite coal samples at lower temperature range less than 100 °C. At such lower temperature range less than 100 °C, oxidation rate [$\ln(k)$] of anthracite coal sample from higher proneness to spontaneous combustion is also even higher than that of bituminous or sub-bituminous from coal mines from higher proneness to spontaneous combustion.

From such data, it might be possible to set an assumption that anthracite coal samples from the mine with higher proneness to spontaneous combustion have higher activity of oxidation process even at lower temperature including ambient temperature.

Such specific characteristics can be imagined by the facts of deformation of a PET bottle used for coal sample container. One example is shown in Figure 6-14.

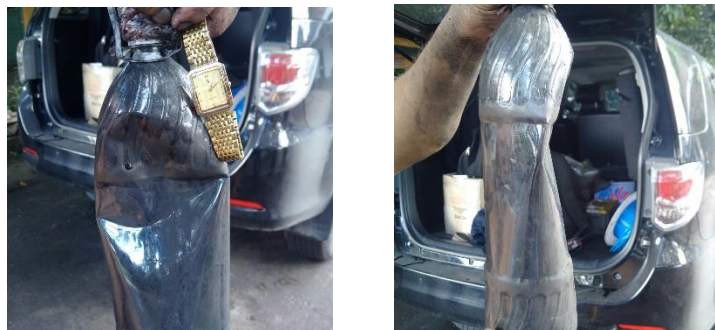


Figure 6-14: Deformation of a PET bottle used for the sample container due to oxygen absorption by sampled anthracite coal

(Khanh Hoa coal after 40 minutes from sampling at the underground site)

Figure 6-14 shows the deformation of the PET bottle only just after 40 minutes the coal sample was filled at underground mine site of Khanh Hoa Coal Mine. Deformation of the PET bottle results from the inside pressure reduction due to oxygen absorption by coal from inside limited volume air.

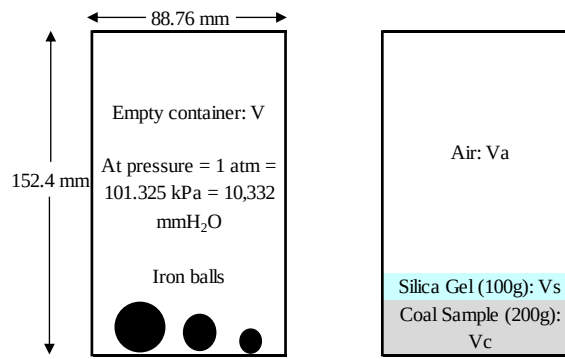


Figure 6-15: Steel container used for oxygen absorption by anthracite coal

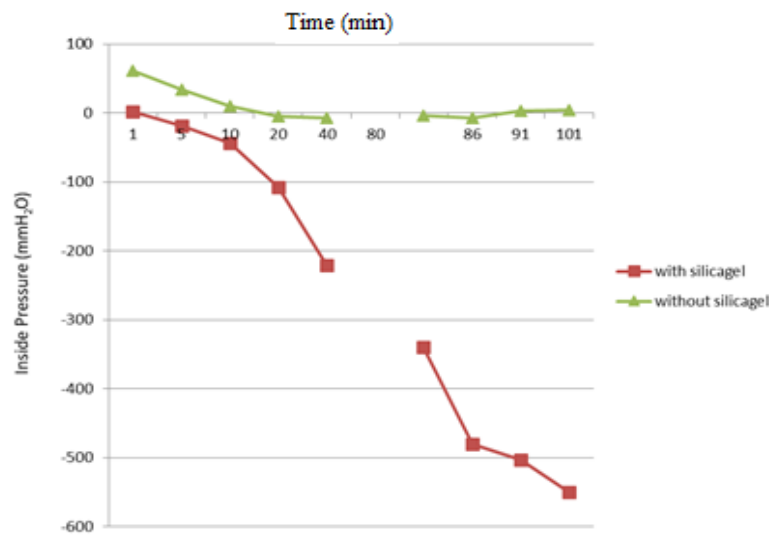


Figure 6-16: Inside pressure change after confining anthracite coal sample (Quang Hanh) into a steel container (Disconnected part is the time for crushing by a vibrating machine.)

Figure 6-16 shows an example of pressure changes after confining anthracite coal sample (Quang Hanh) into a steel cylinder container shown in Figure 6-15. For this test, two same size steel containers were prepared and 200 g of coal sample is confined with steel balls in both containers and 100 g of silica gel was confined in one of those. After confining the coal sample inside pressure was measured by a pressure gauge and some minutes later crushing operation was conducted by using a vibration machine and then after again pressures of both containers were measured, the inside gas was analyzed by a gas chromatograph.

In case of the data shown in Figure 6-16, it is obvious that Quang Hanh coal absorbed much more oxygen when it was confined with silica gel than the case without silica gel. The final gas analysis result showed that the oxygen concentration inside the container was 4.96 % after three days of confinement. These results might show absorption of moisture from coal sample by silica gel encourages oxygen absorption by coal itself. This trend might agree with the results shown in

6.2 in this Chapter.

However, the author could not get yet the reliable quantitative data of oxygen absorption by anthracite coal, because the data vary depending on the confining method of coal samples (at the site or at laboratory), the size of coal sample, the moisture of coal samples, etc. So, this study should be carried out intensively in the future.

6.3.2 Storage period of coal samples after sampling at the site⁶⁻¹⁾

As shown in the curves of coal sample from No. 91 Enterprise Coal Mine in Figure 5-2, even same coal sample showed different incubation (retention) time before starting rapid temperature increase after 100 or 125 °C.

For the purpose of examining the reason of variations in the incubation (retention) time, several tests were conducted by changing the storage period of coal samples. As one example, a series of tests using the same coal samples of Coal Seam No. 5 of No. 91 Enterprise Coal Mine are shown in Figure 6-17. For the tests, coal samples were collected on May 19, 2016 and the samples were subjected to the tests under the same conditions except for the storage period at the laboratory. For these tests, to reduce the effects of initial oven temperature during introduction of N₂ gas, the test was started from room temperature (28 °C) and N₂ gas introduction time was kept shorter (5 min) to avoid evaporation of moisture from coal samples. In Figure 6-17, three test results of May 19 (same day, few hours after the sampling time), May 20 (one day after sampling) and May 22 (three days after sampling) are shown. From those data, the storage period might give stronger effects on the oxidation process at the tests of SIT-2.

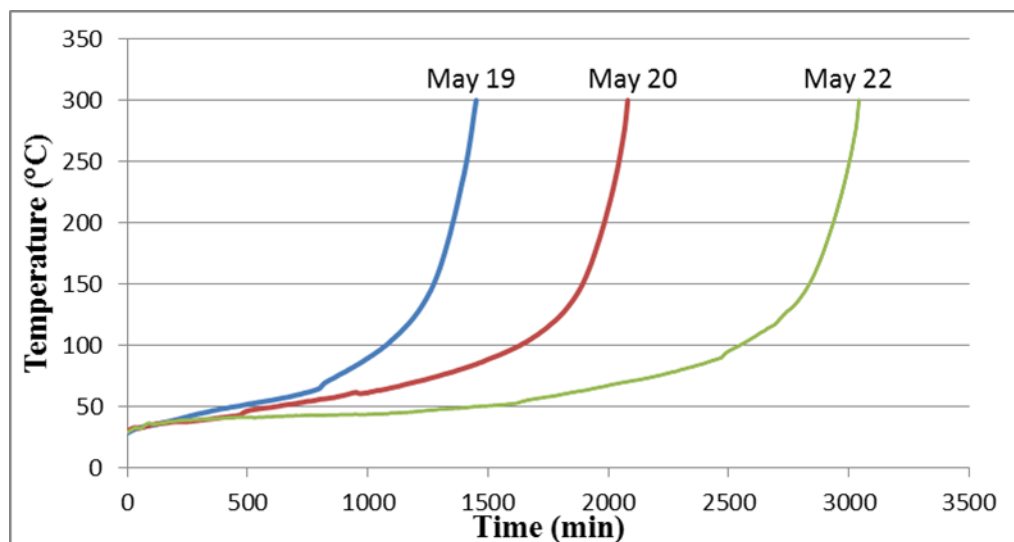


Figure 6-17: Effect of storage period of coal sample on the oxidation process by SIT-2
(Coal sample: No. 91 Enterprise Coal Mine, Coal Seam No. 5, sampled on May 19, 2016)

In case of longer storage periods up to one week, incubation (retention) time is not necessarily become longer in proportion to the storage period. For example, through the tests of a coal sample from Khanh Hoa Coal Mine, it took 329 minutes for 4 days storage, 423 minutes for 5 days storage and 343 minutes for 6 days storage to increase coal sample temperature from room temperature (28 °C) to 300 °C, in which data of 5 days storage and 6 days storage showed the opposite incubation (retention) time.

However, in general incubation (retention) time under the same condition becomes longer as the storage period of coal sample become longer. Such tendencies might come from “oxidation of sample surface” and “evaporation of moisture of coal sample” during handling in the laboratory. Dealing with the effects of such parameters on the results of tests by using SIT-2, further discussion might be necessary.

6.3.3 Discussion

As shown in Figures 6-14 and 6-16, some anthracite coals show very active oxygen absorption from just after the relevant coal face to fresh air. In addition, as shown in Figure 6-18, a very high temperature zone more than 60 °C (★ marked position of the long-wall coal face) was observed at a coal face of Quang Hanh Coal Mine, which might come from very active oxygen absorption of virgin anthracite coal faced with fresh air by coal winning activity.

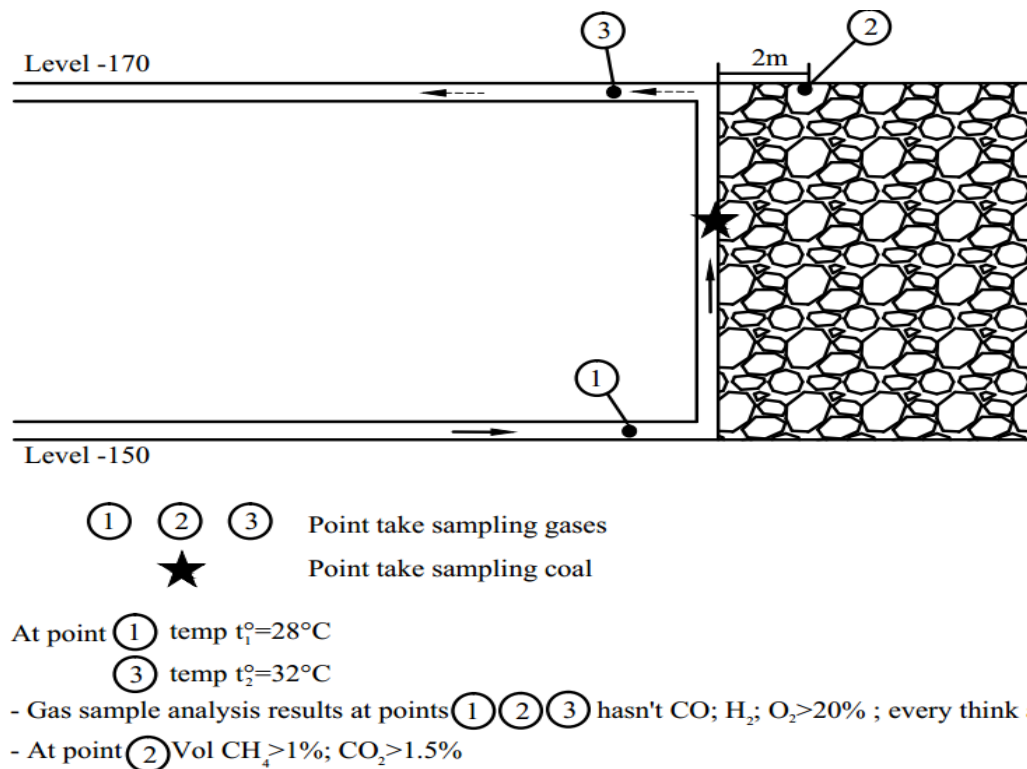


Figure 6-18: Very high temperature zone more than 60 °C at a coal face of Quang Hanh Coal Mine

From such facts, it might be essential to consider the characteristic change of the anthracite coal sample from the moment of sampling at underground site to the time of laboratory test including transportation and storage periods. In other words, how can we keep the coal sample for laboratory test under the same condition as underground virgin condition? In case of very active anthracite coal, it might change its characteristics in very short time as shown in Figures 6-14 and 6-18.

In addition, it is essential to evaluate the oxygen absorption, that is inward gas movement from ventilation air at a new exposed coal surface in relation with emission of methane gas that is outward gas movement from coal itself. Methane gas emission from newly exposed coal seam is very predominant at the first several ten days⁶⁻⁶⁾. Namely, from view point of gas dynamics, methane gas from the coal seam and oxygen from ventilation move opposite direction at the coal surface. Such discussion on dynamic of dual gases at a coal surface boundary might important research subject.

Of course we tried to keep anthracite coal samples unaffected by air (oxygen) or drying process during sampling as well as transportation and storage time by confining them in closed containers filled with inert gas (N₂). However, exposure of coal samples to air is inevitable during sampling at the site and during the handling and preparation in the laboratory before subjecting them to laboratory test.

From such consideration, it is necessary to conduct further research on handling method of coal samples to get more accurate laboratory test data for evaluating their susceptibility to spontaneous combustion.

6.4 Conclusion

In Chapter 6, effects of porosity of coal sample, moisture of coal sample and oxygen absorption on the susceptibility of anthracite to spontaneous combustion were discussed. The results can be summarized as follows:

- (1) Coal samples from the coal mines with problems of spontaneous combustion showed higher porosity value especially in pore size.
- (2) Higher oxidation rates of anthracite coal samples from west part of Quang Ninh Coal Field might results from higher porosity than those of east part of the coal field. Namely, porosity of anthracite coal is related to the oxidation process at lower temperature less than 150 °C.
- (3) Pore size might have positive correlation with the oxidation rate of anthracite coal. In addition, it might be possible to classify the susceptibility of anthracite to spontaneous combustion by using the values of pore size.
- (4) The moisture of original coal samples regulated the start and rate of oxidation of coal samples.
- (5) Replacement of the moisture in the micro-cracks of coal sample by oxygen or air might

reduce active oxidation of coal itself.

- (6) Higher moisture evaporation rate induced more active oxidation of coal sample.
- (7) Storage period of coal sample might give the effects on the oxidation process during the tests of by using SIT-2. To make clear the effects of relevant parameters affected by the storage period of coal samples, further discussion should be essential.
- (8) Because there are many factors of active oxygen absorption of anthracite just after being exposed to fresh air affecting oxidation process of coal both in the laboratory as well as coal mine site, it is suggested to conduct further studies.

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Chapter 7: Conclusion of the Study and Recommendation for Future Work

This study was carried out to find the characteristics of spontaneous combustion at anthracite coal mines in northern Vietnam coal fields as well as to find reasonable methods to evaluate the susceptibility of anthracite coal to spontaneous combustion.

For these purposes, the author conducted site surveys and laboratory tests in which gas analysis of in situ samples and samples from laboratory tests, adiabatic oxidation tests of anthracite coal from more than ten coal mines as well as some other tests at both sites and laboratory like oxygen absorption by anthracite coal at ambient temperature were carried out.

Conventional methods for evaluating the susceptibility to spontaneous combustion of coal samples were also applied to Vietnamese anthracite coal if they were applicable or not. Throughout the studies, the author also found some issues that should be solved by future studies.

The results could be summarized as follows:

7.1 Characteristics of spontaneous combustion at anthracite coal mines

Characteristics of spontaneous combustion at anthracite coal mines in Vietnam can be summarized as follows:

- (1) Even anthracite coal mine starts spontaneous combustion easily. Eighteen spontaneous combustions have occurred at seven coal seams of five coal mines up to date since May 2004.
- (2) In the shortest case, a coal seam started spontaneous combustion within two weeks after exposure to air. (Less than two weeks after a rock entry heading penetrated the coal seam.)
- (3) Spontaneous combustions started at both goaf area and freshly exposed coal wall.
- (4) Higher concentration of hydrogen gas (H_2) was detected in gas samples from inside of the sealing.
- (5) Except for methane gas (CH_4), no higher rank hydrocarbon gas, such as ethane (C_2H_6), ethylene (C_2H_4) or others, was found. This feature is completely different from the cases of bituminous or sub-bituminous.

Dealing with characteristics of gas contents inside the sealing of spontaneous combustion area, laboratory tests confirmed the same results above.

7.2 Validity of conventional methods for evaluating susceptibility to

spontaneous combustion

Dealing with the conventional methods to evaluate the susceptibility to spontaneous combustion of coal samples, it was shown that these methods were not applied to anthracite coal directly as follows:

In case of the Russian method, all anthracite coal samples obtained at coal mines with problems of spontaneous combustion showed their U_{25} values ranging from 28% (maximum) to 4% (minimum) of Russian criteria, $U_{25} = 0.025$. Namely, all these Vietnamese anthracite coals were evaluated as “Less or no susceptibility to spontaneous combustion”. However, when we compared U_{25} values of Vietnamese anthracite coal samples, the coal samples had higher U_{25} values comparing with the coal samples that had not spontaneous combustion up to date.

In case of the Japanese method, the minimum T_{180} value of No. 91 Enterprise coal showed 1.8 times higher than the criteria, $T_{180} > 115$ min for “hard to start spontaneous heating”.

In case of the Polish method, all the anthracite coal samples showed from “III: medium” to “I: too low”

From the data shown above, three these conventional methods cannot not evaluate the susceptibility of Vietnamese anthracite to spontaneous combustion directly, because their methods was developed based on the experimental studies by using mainly bituminous or sub-bituminous coal. Namely, a new method should be discussed for spontaneous combustion of Vietnamese anthracite.

7.3 Results from adiabatic oxidation tests of anthracite

In relation to spontaneous combustion properties of anthracite coal, from the analysis of oxidation reaction data of anthracite under adiabatic condition and comparison of these data with in situ data of coal mines with spontaneous combustion, the author obtained the following conclusions:

- (1) Anthracite coal samples from coal mines with problems of spontaneous combustion showed higher oxidation reaction rates at lower temperature range less than 150 °C than those of the other coal samples taken from coal mines without spontaneous combustion problem.
- (2) When these anthracite data with higher susceptibility to spontaneous combustion were compared with the data of bituminous and sub-bituminous coal with higher susceptibility to spontaneous combustion, the oxidation reaction rates were reversed at the temperature range around 150 °C. Namely, in case of anthracite, oxidation reaction rates were much higher than those of bituminous or sub-bituminous at lower temperature range; however, those data were reversed completely at higher temperature range.
- (3) At higher temperature range more than 150 °C, there was not so much difference in

oxidation reaction rates of anthracite, regardless of higher susceptibility to spontaneous combustion of each coal sample.

Based on the results shown above, the author proposed a method to evaluate the susceptibility of anthracite coal to spontaneous combustion into three groups of “high”, “medium” and “low” by using the oxidation rate at lower temperature range less than 150 °C by using an adiabatic oxidation tester.

7.4 Effects of porosity of anthracites on oxidation process

Effects of porosity of coal samples on the susceptibility of anthracite to spontaneous combustion were discussed. The results are summarized as follows:

- (1) Coal samples from the coal mines with problems of spontaneous combustion showed higher porosity value, especially pore size.
- (2) Higher oxidation rates of anthracite coal samples from west part of Quang Ninh Coal Field might result from higher porosity than those of east part of the coal field. Namely, porosity of anthracite coal was related to the oxidation process at lower temperature less than 150 °C.
- (3) Pore size might have positive correlation with the oxidation rate of anthracite coal. In addition, it might be possible to classify the susceptibility of anthracite to spontaneous combustion by using the value of pore size.

7.5 Effects of original moisture of anthracite on oxidation process

Effects of original moisture of coal samples on the susceptibility of anthracite to spontaneous combustion were discussed. The results are summarized as follows:

- (1) The moisture of original coal samples controlled the start and rate of oxidation of coal samples.
- (2) Replacement of the moisture in the micro-cracks of coal samples by oxygen or air might reduce active oxidation of coal itself.
- (3) Higher moisture evaporation rate induced more active oxidation of coal sample.

7.6 Recommendation for future work

During the study of this thesis, the author found that anthracite coal samples changed the characteristics quite rapidly due to rapid oxidation of coal sample itself due to exposure to air and due to the change in the original moisture of the coal sample during drying or absorbing the moisture from the ambient air or confining inert gas (N₂). Dealing with such rapid change of coal sample itself, the author did not clarify all the features so that the author would like to recommend

the following study:

The storage period of coal samples might give the effects on the oxidation process during the tests by using SIT-2. To make clear the effects of relevant parameters affected by the storage period of coal samples, further discussion should be essential.

Because there are many factors such as very active oxygen absorption of anthracite just after the exposure to fresh air affecting oxidation process of coal in both laboratory and coal mine site, it is suggested to conduct further studies.

Annex

Annex 1: Past Researches on Mechanism of Spontaneous Combustion of Coal

A1-1. Introduction on past researches on the mechanism of coal spontaneous combustion

Not only in Vietnamese cases of coal spontaneous combustion but also in other world cases, causes of coal fire at underground coal mines are explained by various factors. Many researches or studies or surveys on spontaneous combustion of coal have been conducted dealing with occurrence situations and outbreak mechanism or other factors of effects up to date; however, no clear mechanism of spontaneous combustion has been established as a reliable theory because many factors interrelate each other. Another unsolved reason might be that there is no rational standard that can determine the susceptibility to spontaneous combustion of coal or no enough statistical data from which we can derive a reasonable susceptibility index of spontaneous combustion of coal.

Even if it is said that this coal seam is liable to start spontaneous combustion, the degree cannot be evaluated qualitatively. Such situations might make difficult find a correlation of one factor with spontaneous combustion. Difficulties on judging the susceptibility to spontaneous combustion of coal are the same even in foreign countries. Namely, unified correlations between outbreak of spontaneous combustion and related factors cannot be found up to date. There are some factors that have been considered as a cause of the heat generation from coal as follows^{A1-1)}:

- ✓ Pyrites theory
- ✓ Bacterial action
- ✓ Heating due to earth movements
- ✓ Sorption of water to coal

However, some of above theories have been considered as insignificant effect or poor role on the start of coal spontaneous combustions. Winmill (1915-1916)^{A1-1)} investigated the influence of pyrite on the heating of coal in air. Graham (1923-1924)^{A1-1)} also examined the Winmill's results and made a conclusion on the role of pyrite on the heating process of coal. He concluded that "most of the underground heating are due to oxidation of iron pyrite by moist air", which can be represented by the following equation:



However, the influence of contained pyrite in the heating process of coal was also found inappropriate by the research of Munzer (1975)^{A1-1)}. Munzer concluded that the effect of pyrite content on heating process might be notable "if its concentration in finely dispersed exceeds more than 5 or 10 %. And if the pyrite content is less than 5 %, its effect could be neglected".

Other theory had been proposed that inorganic substances (ash) in coal accelerate the oxidation of coal^{A1-1)}, but this theory has not been proved yet. In that period, it was told that bacteria in underground might cause spontaneous combustion^{A1-1)}, but by the research of Fuchs^{A1-1)}, he concludes that “bacteria could cause only a slight heating that may not play a significant role”.

As a different approach, mechanisms of spontaneous combustion of coal were discussed from coal seam mining conditions. In old time, comparison between “advancing coal winning” and “retreating coal winning” was discussed statistically.^{A1-7)} This idea might be started from the premise that air leakage cannot be prevented even if back goaf filling is conducted, namely, if we can conduct air tight back goaf filling, this kind of comparison might be meaningless.

A1-2. Cause of coal spontaneous combustion

Spontaneous combustion of coal is an oxidation phenomenon of coal. Currently, it is commonly approved that “spontaneous combustion starts from accumulation of the heat generated from very slow oxidation of coal even from usual ambient room temperature”.

Oxidation of coal and combustion of coal are both the result of combination between carbon and oxygen, but oxidation occurs at any temperature and in combustion the combination rate between oxygen and carbon is very fast and in many cases oxidation reaction becomes a chain reaction, namely, combustion is a status of automatic oxidation process^{A1-2)}. Spontaneous combustion of coal is a result of transition from slower oxidation phenomena to combustion phenomena so that it might be possible to replace the susceptibility to spontaneous combustion of coal to the oxygen absorbing rate, namely, easiness of oxygen absorption of the coal can be an indicator of susceptibility to spontaneous combustion of coal based on the idea that ignition of the coal is a result of heat generation from slower oxidation.

However, the mechanism of oxygen absorption of coal has not been solved perfectly yet, but it may be deeply related to the outer surface of coal itself. From the idea, the coal oxidation at ambient temperature is considered as a cause of coal heating. Most researches have agreed with the idea of participation in oxidation of the surface per unit weight. At the surface of coal, it is not clear that oxygen is physically absorbed on the surface or chemically combined with molecules of coal based on some reactions. Even in case of chemical combination between oxygen and molecules of coal, the process from heat generation to ignition might be divided into 2 or 5 steps^{A1-3)}.

In recent time, a theory tells that temperature increase of coal at lower temperature range might mainly come from the latent heat that is released from the condensation of the humidity in air onto coal surface under a relative complex conditions between coal moisture and temperature and air humidity and temperature. Taking account for this idea some studies are being continued.

As overviewed above, there are many factors related to the causes of spontaneous combustion of coal so that still many researches are being underway. As concluded by the research of Morris and Atkinson (1986, 1988)^{A1-4)}, coal spontaneous combustion can be caused by some main factors classified into (A) seam factor, (B) geological factors and (C) mining factors. According to the research of Cliff and others (2014)^{A1-5)}, the factors that may affect the oxidation of coal can be listed in Table A1-1 below:

Table A1-1: Factors affecting the oxidation of coal (Cliff and others, 2014)

Intrinsic factors	Extrinsic factors
Low rank of coal	Faults
Low ash	Folds
High friability	Dykes
Weak caking properties	Weak and disturbed strata conditions
High reactivity	Seam thickness
High heat capacity	Steepness of seam
Low thermal conductivity	Shallow cover
High coefficient of oxygen absorption	Multi-seams in close proximity
High proportion of oxygen functional groups	Porous petrographic structure
High volatile matter	Increase amount of time of exposure of broken coal
Pyrites	Mine ambient temperature
Moisture content	
Particle size and surface area of coal	

The other research classified the effects of coal spontaneous combustion by dividing those factors into internal factors (coal characters) and external factors. If we divide the latter into surrounding environment, and coal reserve or mining conditions, these factors can be itemized as follows:

(1) Internal factors (Coal characteristics)

- ✓ Composition, coal rank;
- ✓ Friability, particle size and surface area of coal;
- ✓ Moisture, hydrogen contents and petrography;
- ✓ Inorganic contents of coal (Presence of iron pyrites, bacteria and other minerals)

(2) External factors (Underground environment and mining conditions)

1) Underground environment: Temperature; moisture/relative humidity; barometric pressure; O₂ concentration.

2) Geological factors

- ✓ Coal seam and surrounding strata condition;
- ✓ Seam thickness;
- ✓ Seam gradient;
- ✓ Caving characteristics;
- ✓ Faults and other geological disturbances;
- ✓ Coal outbursts;
- ✓ Friability;
- ✓ Depth of cover.

3) Mining factors

- ✓ Mining methods;
- ✓ Rate of advance;
- ✓ Pillar conditions;
- ✓ Roof condition;
- ✓ Crushing;
- ✓ Packing;
- ✓ Presence of timber or other organic waste material in abandoned areas or dumps;
- ✓ Leakage of ventilation;
- ✓ Multi-seam working;
- ✓ Coal losses;
- ✓ Worked out areas;
- ✓ Heat from machines;
- ✓ Stowing;
- ✓ Ventilation system and airflow rate;
- ✓ Ventilation pressure;
- ✓ Method of stockpiling and stockpile compaction.

In the following sections, coal characters and the conditions around coal will be discussed among many factors affecting on the oxidation of coal.

(1) Coal characteristics and spontaneous combustion

(1)-a. Rank of coal

As a definition of coal “is an organic rock (as opposed to most other rocks in the earth's crust, such as clays and sandstone, which are inorganic); it contains mostly carbon (C), but it also has hydrogen (H), oxygen (O), sulfur (S) and nitrogen (N), as well as some inorganic constituents (minerals) and water (H₂O)”^{A1-6}. The coal characteristics (carbon content, volatile matter, calorific value, moisture content) have been investigated and classified as coal type like peat, lignite, sub-bituminous, bituminous and anthracite. One of common classifications of coal by US can be shown in TableA1-2 below:

Table A1-2: Variation of selection coal properties with coal rank^{A1-6)}

Rank	Low Rank		-	High Rank	
	Lignite	Sub bituminous		Bituminous	Anthracite
Age	Increase				
Carbon (%)	62-72	72-76		76-90	90-95
Hydrogen (%)	~5		Decreases		
Nitrogen (%)	<		~1-2		>
Oxygen (%)	~30		Decreases		~1
Sulfur (%)	~0	Increases		~4 Decreases	~0
Water (%)	70-30	30-10		10-5	~5
Heating Value (BTU/lb)	~7000	~10,000		12,000-15,000	~15,000

Depending on the type of coal, the capacity of oxygen absorption is different. Coal characteristics make different effects on oxidation process of coal. As shown by a Japanese coal hand book^{A1-7)}, the relationship between oxygen absorption (%) and volatile matter (%) shows a proportional relation up to some extent. As shown in Figure A1-1, oxygen absorption increases to some extent with the increase of volatile matter or oxygen content in coal. However, the oxygen absorption becomes greatest at 36 % of volatile matter and at 9 % of oxygen contents. More than that range the correlation between oxygen absorption and volatile matter or oxygen content become irregular.

In general, there is a common understanding that lower coal rank coal starts spontaneous combustion easily than higher coal rank. However, the correlation between coal characteristics (volatile matter or oxygen content) and spontaneous combustion is still not clear. The relationship between coal rank and coal spontaneous combustion can be explained by the difference in coal rank that makes difference in the effects (reaction) on spontaneous combustion.

(1)- b. Friability, particle size of coal

Vulnerable coal can easily be fractured into small fragments or powder by blasting or mechanical power during coal winning or by rock pressure. Therefore, the relative surface area of coal increases remarkably and oxidation rate might be accelerated. In general, smaller particle size of coal gives much bigger relative surface area per unit weight of coal and gives more chances of oxidation of coal itself.

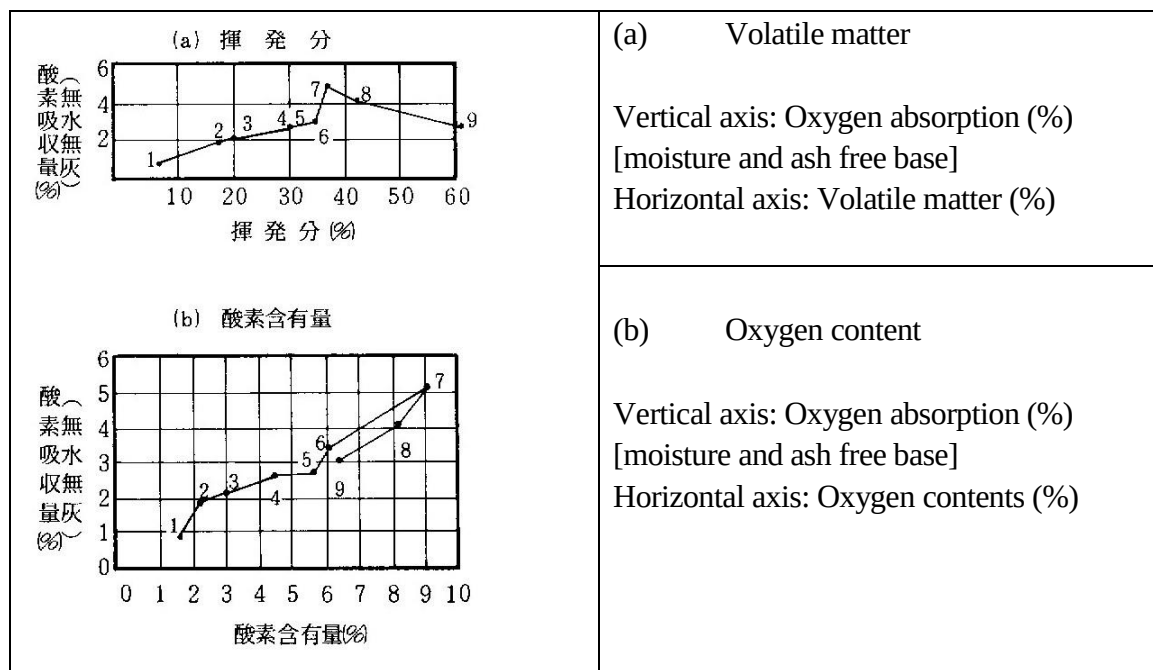


Figure A1-1: Volatile matter or oxygen content and oxygen absorption
(Oxidation temperature: 105 °C, Oxidation period: 5 days)

From a long-term observation of oxidation status of coal, it is said that the depth of coal oxidation is around 10 cm at the most^{A1-8)}.

When the relative surface of coal increases, early stage oxidation is promoted and the temperature increases due to oxidation heat, therefore, the particle size of coal has a close relation with spontaneous combustion, namely, friable coal tends to start spontaneous combustion easily. Actually, spontaneous combustions are observed at the coal mines that develop friable coal or at the places of fractured coal zone.

(1)-c. Moisture, hydrogen and petrographic contents of coal, and spontaneous combustion

According to researches of the effect of moisture of coal on the susceptibility to spontaneous combustion, there are contradictory ideas; a theory said that in some coals the moisture promotes oxygen absorption and in other coals the moisture delays oxygen absorption^{A1-9)}, and another theory said that moisture of coal works as a catalysis for promoting oxidation of coal^{A1-10)}. By the results of Nandy et al. (1967)^{A1-1)}, an optimum moisture level might be around 5% in coal that shows the maximum spontaneous combustion tendency. Vance et al.^{A1-11)} showed the effect of moisture content on New Zealand sub-bituminous coal under adiabatic conditions, namely, the rate of self-heating increased as moisture content decreased.

In general, it is said that low coal rank lignite higher content of moisture shows higher susceptibility to spontaneous combustion. This idea can be explained by the fact that at the time of evaporation of moisture from the coal into air, many small cracks are created and such newly created crack surfaces can easily be combined with the oxygen in air at a higher rate. Namely, in such lignite with higher moisture content, when the moisture works for creating new surfaces

(pores), such coal can easily start spontaneous combustion. Therefore the cause of spontaneous combustion might be different depending on the kind of coal.

Other researches on the effects of content of hydrogen or petrographic contents of coal have been conducted; however, correlation between those factors and spontaneous combustion has not been found clearly yet.

(1)-d. Inorganic contents of coal including pyrite and spontaneous combustion

In an old period, it is believed that the pyrite in coal is a cause of spontaneous combustion; however, the correlation between the contents of pyrite and spontaneous combustion is not confirmed. However, it is said that the coal with 2 % or more of sulfur shows higher susceptibility to spontaneous combustion and that spontaneous combustion tends to start from pyrite in coal ^{A1-7)}.

As shown in the above part, there are few coals in Japan that contain sulfur of 2 % or more ^{A1-8)}, therefore, the relation between the content of sulfur and susceptibility to spontaneous combustion cannot be identified; however, it is confirmed that when sulfur exist as pyrite in a coal, it generates heat by the reaction with oxygen under presence of moisture. From such facts, it is necessary to conduct the study on the coals that have sulfur of 2 % or more in relation with start of spontaneous combustion.

Dealing with the contents of other inorganic substances, it is said that some contents work as a catalysis to promote spontaneous or some contents works as an inhibitor, but details have not been clarified yet.

(2) External factors (Underground environment and mining conditions)

1) Underground environment

Underground temperature of coal mine is almost constant whole year around, in summer or winter, or daytime or night time, when surface air with different humidity is introduced, water is condensed on the surface of coal or moisture of coal is evaporated. Namely, delivery and acceptance of water or heat are conducted on the underground coal surfaces always. When the moisture of air condenses onto coal surfaces, due to the latent heat generated, the coal temperature increases. But further condensation of water onto the coal surfaces makes the coal wet and the water cools the coal on the contrary.

In case of lower humidity of air flow, the moisture of coal evaporates and as a result many micro-cracks are generated and oxidation of the coal is promoted, namely, this trend tends to promote spontaneous combustion.

As shown in 1-(c) dealing with the moisture of coal itself, in relation to the moisture of the air flow, coal shows heat generation phenomenon. And wet water on the coal surface might trigger the oxidation of pyrite inside the coal and increase the coal temperature itself.

Actions of the moisture in air flow can be summarized as follows:

- (1) By the condensation of the moisture of air onto coal surfaces, the latent heat is provided to increase the coal temperature.
- (2) The condensation of the moisture of air onto coal surfaces makes coal surfaces wet and the coal is cooled as a result.
- (3) Wet moisture promotes the oxidation of pyrite in coal and it increases the temperature of coal.
- (4) The moisture of the coal evaporates into air flow and new cracks are created in the coal, which promote oxidation of coal and increase the temperature of coal.

Ambient temperature and spontaneous combustion^{A1-8)}

As shown in Figure A1-2, if coal temperature increased more than 60 °C, oxygen absorption increased quite rapidly, namely, coal tends to start spontaneous combustion easily. Exactly, the coal mines with higher underground temperature such as Joban Coal Field in Japan often had spontaneous combustion. If the coal temperature increases more than 100 °C, oxygen absorption become even much higher and oxidation proceeds like a chain reaction, but in case of lower temperature than this range, the situation does not necessarily connect to start of spontaneous combustion even at higher temperature range.

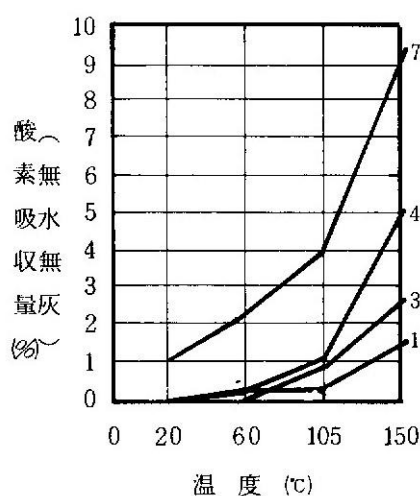


Figure A1-2: Coal temperature and oxygen absorption (Oxidation period: one day)^{A1-8)}

Volatile matter ---- 1: 6.3 %, 2: 20.1 %, 3: 29.9 %, 4: 36.6 % (moisture and ash free)

Vertical axis: Oxygen absorption (%) [Moisture and ash free base],

Horizontal axis: Temperature (°C)

2) Geological factors^{A1-15), A1-16)}

Coal seam and its surrounding strata condition also have effects on the causes of coal spontaneous combustion. In case of a thick coal seam, the technology for coal exploitation cannot recover all coal from its thickness. Some part of coal at the roof and floor part are left, which

results in residual coal in the goaf area. In case of faults and other geological disturbances, the risk of coal spontaneous combustion might increase. Fault or geological disturbance often results in change in coal seam structure such as seam thickness, dip of coal seam or even cut of coal seam. Because of exploitation technology, the coal is left at the roof and floor when long-wall or roadway pass through those areas. For a long-wall panel, faults and geological conditions also slow down the rate of coal face advance compared with the incubation time of coal to spontaneous combustion. The depth of coal seam also has effect on the causes of coal spontaneous combustion. At a shallow coal mine, the thickness of overburden is smaller since the effect of ground subsidence appears on the surface. Thus, when coal seam is mined, the goaf area might be possible to connect to surface air through the crack systems. At the shallow part area, the surface water can be penetrated to the coal seam or to the goaf and make pathways for air come into underground by negative ventilation pressure. For a deeper coal seam, the effect might come from the ground pressure on the coal pillar, especially for protective coal pillars. This effect often results in cracks and crushes of coal pillar itself and therefore, it increases the exposed area to air and induces the oxidation process.

3) Mining factors ^{A1-15), A1-16)}

3)-1 Mining method

Effects of the mining method on coal spontaneous combustion vary by the method itself. Based on the view point of combustion risk, the advance mining method is accompanied with higher risk than the retreating mining method. For a long-wall panel with advancing method, three side of the goaf always face to the tail gate roadway, transportation gate roadway and long-wall face. By the ventilation pressure and the crack systems of sealing walls, air can be penetrated into the goaf area.

3)-2 Rate of advance

The rate of coal face advance affects the time of coal expose to air at the coal face as well as at the parts of coal exposed to air through crack systems. As explained before, the coal starts its oxidation process just after facing to the air. The period from starts of oxidation to spontaneous combustion is usually called as an “incubation time”. When the coal face advance rate is smaller than the incubation time, a fire may occur due to of oxidation.

3)-3 Coal pillar conditions

Depending on the location of coal pillar, it has roles as a protection pillar for underground space or barriers between working area and goaf area. Generally, coal pillars are designed based on the ground pressure in order to prevent cracks or breakage. However, for a coal pillar existing for a long time, the possibility of spontaneous combustion might be higher due to its exposure to air. In case of barrier coal pillar, the coal pillar is usually destroyed by ground pressure, and mining pressure, which makes pathways to air to contact with old working area.

3)-4 Leakage of ventilation

The air leakage into or through a coal seam or a goaf in underground coal mine has a high possibility. As mentioned in the part of mining method, the air can be penetrated across the goaf area by the ventilation pressure difference between two gate roadways. Even in case of the sealing walls at an isolated area, the air can be penetrated into the isolated area by the pressure difference between outside and inside the walls. Air also leaks through the goaf and sealed area depending on the permeability of the materials.

3)-5 Multi-seam working

At the area of multi-seam working, the distance from a coal seam to the other coal seam affects the fall of the roof of working coal seam. The range of roof fall goes up at least 5 times of the long-wall height. For example, when a lower coal seam is worked and the upper coal seam is unworked, the ground pressure makes the fall and crushes the coal of unworked coal seam. The coal in unworked coal seam can be considered the same as coal left in the goaf area of the working coal seam. The leakage paths can be created from working coal seam and permit air to go through this crack system and bring a risk of coal oxidation in unworked coal seam.

3)-6 Coal losses and worked out areas

Generally, it is impossible to exploit entire coal seam thickness in practical mining methods theoretically; however, the residual coal in the goaf area evidently exists. The amount of residual coal depends on the mining method and no mining method can ensure that the residual coal amount can be neglected. Hence, the risk of coal spontaneous combustion is one of the important factors, which should be taken into account.

3)-7 Ventilation system and airflow rate

Ventilation in underground coal mine is essential for the requirements for supplying fresh air for workers and for diluting the concentration of dangerous gasses in coal mine atmosphere. The main ventilation system at underground coal mine is exhaust ventilation. For a local remote coal face, a booster fan may be used for forced air ventilation through a duct system to the coal face.

It is well known that spontaneous combustion starts at coal pillars or coal walls that generate cracks or fracture due to rock pressure and air leakage. In addition, at a goaf where the back-filling is not enough so that air leakage easily occurs even by ventilation pressure increase by booster local fan, spontaneous combustion tends to easily occur. Namely, ventilation pressure changes might be related to heat generation of the coal of the relevant area.

Ventilation air volume and air speed affect occurrence of spontaneous combustion. Anyhow, a ventilation condition provides (1) enough oxygen supply to oxidize the coal surfaces and (2) heat accumulation condition without release of generated heat is essential for starting spontaneous combustion; however, it is difficult to show concrete ventilation conditions that lead to a spontaneous combustion.

The changes in ventilation resistance or ventilation direction might occasionally create a condition that tends to spontaneous combustion.

A1-3. Factors of outbreak of spontaneous combustion

A1-3.1 Development process of spontaneous combustion

Coal is extracted from a coal seam by a mining method and starts to be exposed to the air at underground coal mine. Hence, coal is stored for a while after producing from underground coal mine at each step of coal processing, transportation and utilization. In every above stage, coal starts oxidation at ambient temperature following the self-oxidation process. If coal reaches to the ignition temperature, oxidation reaction and heat generation become very rapid, namely, automatic chain reaction of oxidation starts. For understanding the ignition stage in the spontaneous combustion of coal, many researches have been done to distinguish the feature of each stage. According to the research of Banerjee et al (1985)^{A1-1}, there are three stages that manage the spontaneous combustion of coal fire as shown in Figure A1-3.

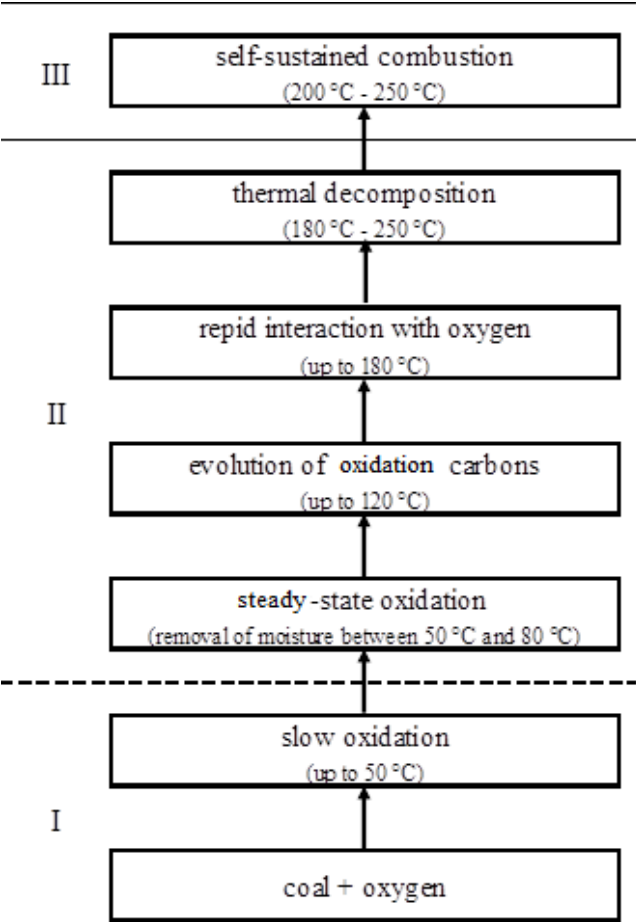


Figure A1-3: Three stages of spontaneous combustion of coal^{A1-1}

In accordance with the Newcastle Coal (2014), the stages of coal spontaneous combustion are divided into 5 stages as shown in Table A1-3:

Table A1-3: Stages of severity and risk of spontaneous combustion (Newcastle Coal, 2014)

Risk level	Stage of severity	Condition
Low	Stage 1	Coal gives off steam
	Stage 2	Coal gives off localized white smoke
High	Stage 3	Coal gives off plumes of white smoke
	Stage 4	Coal burns with yellow sulfur smoke
	Stage 5	Coal burns with flames

According to the research of GIG (Polish Central Mining Institute), theory of coal heating is accomplished with the gases generation during heating process. Coal heating steps can be divided into the following six steps:

- (1) Incubation stage: This stage starts from initial temperature of coal and rock surrounding coal seam. The maximum temperature of this stage is 60 °C where the carbon gas concentration is around 10 ppm. According to the report, at 40 °C of the coal oxidation process, if an accumulation phenomenon does not occur around hot pot, the incubation stage is interrupted.
- (2) Water vapor evaporating stage 1: If the heat from the hot pot is accumulated at the first stage, the oxidation process continues. The temperature of this stage is around 60-80 °C. The products of this stage are water vapor and CO gas of around 20ppm. The point at 60 °C is called a critical point. The end point of this stage (80 °C) is called a turning point.
- (3) Water vapor evaporating stage 2 (pyrolysis reaction): The temperature of this stage is around 80-180 °C. The water vapor continuously evaporates. The CO gas concentration is increased up to 50 ppm. The end point of this stage is called a pyrolysis point.
- (4) Up to 280 °C, emission of CO₂ of around 200ppm continues. At the end of this stage, coal ignition starts automatically.
- (5) At 300 °C, emission of CO₂ of around 300ppm continues. The process of coal self-heating is completed, coal starts a fire. The end point of this stage is called a flash point or the minimum temperature of fire.

A1-3.2 Factors that affect development process of spontaneous combustion

There are many factors that affect development process of spontaneous combustion. Partly duplicate with the former explanations; however, if we summarize those factors from viewpoints of both acceleration and inhibition, we can describe them as follows:

- (1) Action of moisture and spontaneous combustion

The moisture of coal can be classified into as follows:

Moisture or retained moisture	Attached moisture on the surface of coal surface mechanically	
	Intrinsic moisture	Absorbed moisture or intrinsic moisture (absorbed moisture in balance with the moisture of the ambient air)
		Compound water or combined water (moisture contained in the coal and its inorganic substances as crystal water)

The retained moisture of coal is kept a reversible equilibrium status depending on the ambient environment where the coal is located. Therefore, if we try to discuss the relation between moisture of coal and spontaneous combustion, we should study the behavior of “retained moisture”.

From a statistical view, if intrinsic moisture increases up to a certain value, the coal shows higher susceptibility to spontaneous combustion^{A1-12}. This trend is similar to those of volatile matter or oxygen content of coal and spontaneous combustion. However, even some coal with intrinsic moisture of 1 or 3 % shows higher susceptibility to spontaneous combustion.

In case of higher retained moisture coal such as lignite, new surfaces of coal are created due to evaporation of moisture and such new faces absorb oxygen faster. Therefore, low rank coal (low degree carbonization coal) with much moisture tends to start spontaneous combustion easily. From the same reason, it can be explained that retained water on coal tends to start spontaneous combustion after a rain fall or submerged coal seam tends to start spontaneous combustion after drainage of water from underground.

On the other hand, in case of lower moisture coal, it is not certain that spontaneous combustion starts due to faster oxygen absorption rate at lower temperature range or due to temperature increase by condensation of ambient air moisture onto the coal; however, it is confirmed that the temperature of coal increases by the latent heat that is generated at the occasion of condensation or absorption onto coal surfaces in relation with the balance between the retained moisture of coal and the moisture of air. However, the mechanism of temperature increase in the temperature range more than the 60 °C is unknown. Such mechanism is under investigation from the viewpoints that automatic oxidation process to spontaneous ignition is achieved (1) by only condensation of moisture of air onto coal surface, or (2) by combination of condensation and evaporation of moistures of coal and air each other.

At moisture absorbing experiments, coal temperature can be easily increased up to 60 °C, but if coal absorbs more moisture at that temperature range, the coal temperature starts to go down due to becoming wet on the surface. Namely, too much moisture cools down the coal temperature.

Furthermore, moisture reacts with pyrite in the coal and the reaction increases the coal temperature as a process to spontaneous ignition.

As shown above, the moisture of coal acts for four kind roles in relation to spontaneous combustion and so we should be very careful for the behavior of moisture of coal and moisture of air surrounding coal in underground.

(2) Factors that accelerate spontaneous combustion

1) Characteristics of coal (particle size of coal, friability)

Oxygen absorbing amount of coal increases in proportion with the relative specific surface area of coal itself, which increases in an inverse proportion with the particle size of coal. Friable coal easily produces smaller particles and tends to start spontaneous combustion.

Smaller particles stay close together tightly so that fresh air cannot get into inside the coal particle layer and as a result oxygen absorption might become stagnant. From this result, the relation between particle size of coal and spontaneous combustion cannot be discussed. However, the research by Akgun and Adrioy (1994) showed the positive effect of smaller particles size on the self-heating process of coal. The other researches on particle size issue also showed the important role of smaller particles size on self-heating process of coal, which might turn to spontaneous combustion^{A1-13}).

In case of low rank coal with higher moisture, during process of evaporation of the moisture many cracks are created in the coal by which the coal is fractured by itself. Remained coal pillar is fractured by rock pressure and become easier for more oxidization.

Fractured coal starts oxidation not only at the place of fractured but also at the places of accumulated or adhered places of machinery or storage bin.

2) Environmental factors

(a) Temperature of environment

Oxidation of coal receives the effect from the temperature of ambient environment. One report tells that when the temperature increases linearly, the oxygen absorption rate increases exponentially^{A1-14}).

After coal increases the temperature up to a certain level, it starts automatic oxidation process and increases the temperature very rapidly and finally reaches to self-ignition. But as shown before, even if coal keeps the temperature around 60 °C, in many cases it becomes calm not going toward self-ignition. The reason of the behavior is not clear but it might result from balance between heat generation and heat dissipation.

In general coal seams of the coal mines with higher underground temperature are easy to start spontaneous combustion.

(b) Air flow^{A1-17})

Oxygen necessary for oxidation of coal is derived from ambient air that the coal contacts. Thus, continuous supply of fresh air is essential for continuous oxidation of coal. But, too much ventilation takes away the generated heat and cools down the coal. On the other hand, at a condition of fewer ventilation in which heat generation is greater than heat dissipations, heat accumulation might result in self-ignition of coal. At an environment in which coal exists most appropriate ventilation pressure and air volume might bring the coal spontaneous ignition. In a ventilation air flow, we have the following relation:

$$\begin{aligned}\text{Ventilation pressure} &= \text{Ventilation resistance} \times (\text{Ventilation volume})^2 \\ &= \text{Ventilation resistance} \times (\text{Cross section of gallery} \times \text{Ventilation rate})^2\end{aligned}$$

From this equation, when ventilation pressure increases ventilation volume, ventilation rate increases, which might promote oxidation of coal. But too much ventilation pressure results in cooling effect on coal oxidation to calm down the heat generation. Namely, ventilation has reciprocal actions, one toward escalation of spontaneous combustion and another toward calming down.

Actual ventilation flows change depending on the pressure of fan, ventilation resistance of gallery, natural ventilation pressure, ventilation doors, underground sealing, cracks generated inside coal seam, etc.

In coal seam, not only just after coal winning, but also after a long time, cracks are created by rock pressure changes due to the effects of nearby coal winning. Air flow to such cracks might be introduced by the pressure differences due to the surrounding conditions, which might result in escalation of coal oxidation.

Even in a boring hole for gas drainage or a pilot boring hole, similar crack creation and oxidation of coal starts. Especially in case of a gas drainage boring hole, negative pressure is introduced to promote the gas drainage, so that fresh air might be introduced along through newly created cracks during boring operation. It is difficult to identify the position of heat generation along the boring hole.

Pressure difference between both sides of a ventilation door is created inevitably so that through the cracks inside the coal seam very slow air flow is created, which might result in oxidation of coal and start of spontaneous combustion.

When the negative pressure of the fan is changed, pressure differences might be created at many places in underground. For example, pressure difference increase between inside and outside a sealing might result in newly introduction of fresh air into a goaf, which might result in another spontaneous combustion even inside the sealed off area.

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- A1-16) Same as A1-3, pp. 3 -15
- A1-17) Same as A1-3, p. 11

Annex 2: Site Situations of Spontaneous Combustion of Anthracite Coal in Vietnam

A2-1. Site situation of coal mines with spontaneous combustion

As shown in Chapter 1, there are five (5) coal mines that have been reported of occurrence of coal spontaneous combustion up to date as follows: Hong Thai Coal Mine, No. 91 Enterprise Coal Mine, Khanh Hoa Coal Mine, Ha Lam Coal Mine and Mao Khe Coal Mine. In addition, Quang Hanh Coal Mine has also been reported of coal spontaneous combustion; however, the detail information is not clear, up to date. For better understanding of these spontaneous combustions, the site conditions and situations of the relevant areas of those spontaneous combustions are summarized as follows:

A2-1-1 Hong Thai Coal Mine

Hong Thai Coal Mine belongs to Mao Khe Coal Basin (Late Triassic Period in Mesozoic Era) in the west of Quang Ninh Coal Field. The mine is located south side hilly terrain that runs from west to east and the coal seams has strike toward west to east and dip toward north. Such geographical and geological conditions make the mine develop the coal seams based on the concept shown in Figure 1-3.

Hong Thai Coal Mine announced four coal spontaneous combustions at the coal seam No. 24 as shown in Figure 2-4 in Chapter 2. The places of hot spots has all been found in the goaf of three coal winning panels and two of which located in the first panel and other two located in the second and third panels, respectively. Generally, this coal seam has been prepared and opened from the surface by a couple of cross-cut roadways at upper and lower levels. When these cross roadways intercept the coal seam, the in-seam roadways have been driven toward the strike of coal seam to establish a coal winning panel. A long-wall coal face has been created by a rise that connects between intake and return roadways at two levels. Mining system is retreating long-wall system.

The long-wall panel has its basic parameters as follows:

- Long-wall length: 120m; Long-wall dip angle: 30°; The height of coal cutting layer in the long-wall: 2.2m; Supporting frame: single hydraulic supports; Coal cutting method: blasting by boreholes.
- Control of rock roof: caving method
- The distance from long-wall panel to surface: from 20m (first panel) to 100m or more.
- Rate of coal recovery: 70-80% of the original coal resources
- Safety coal pillar (distance between upper and lower panels): 10m.

Coal mining method is shown in Figure 1.3.2.1 in Chapter 1.

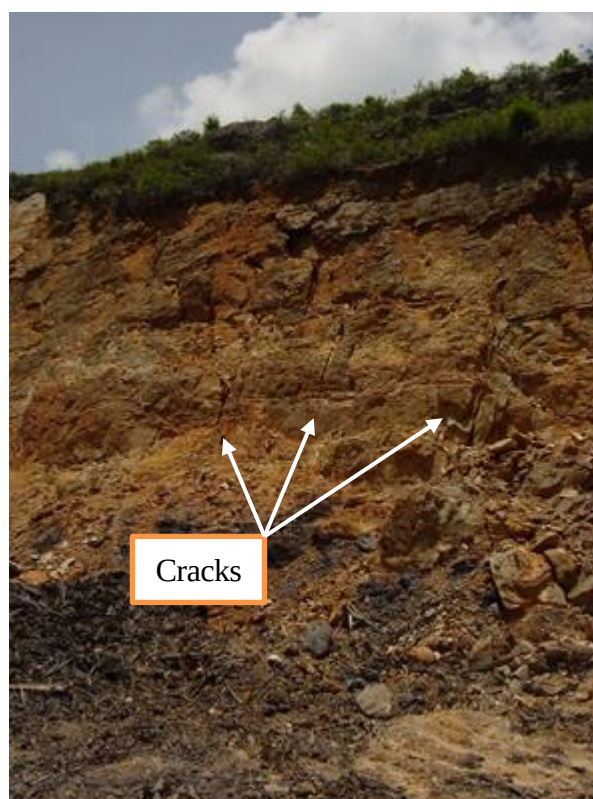


Figure A2-1: Surface cracks of rock seam above a spontaneous combustion area of Hong Thai Coal Mine^{A2-1)}

Another spontaneous combustion has been reported at a place where the coal seam No. 12 were penetrated by a rock entry as shown in Figure A2-2.

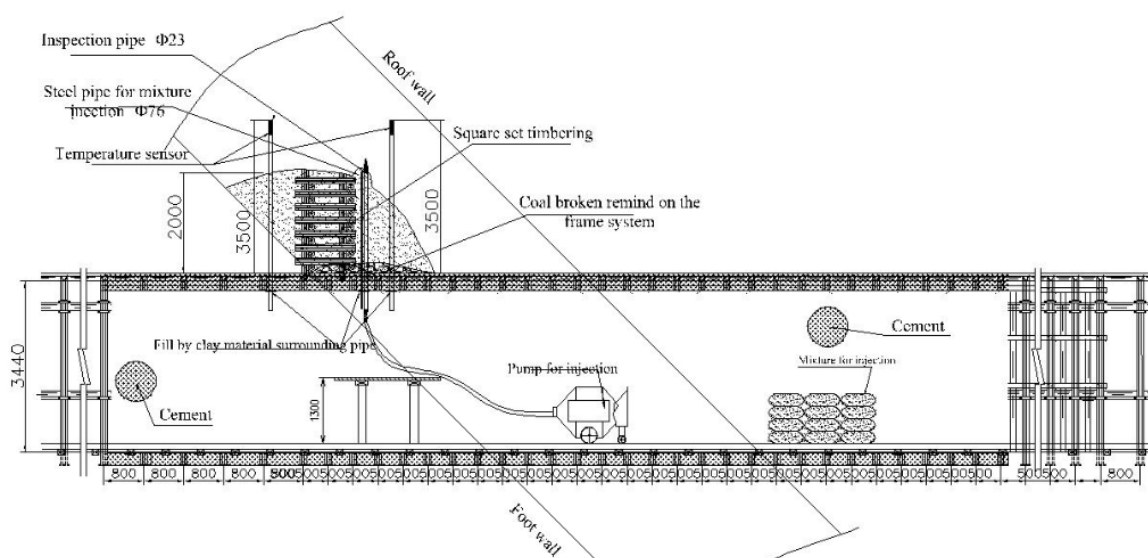


Figure A2-2: Spontaneous combustion at an intersection of rock entry and coal seam No. 12
Spontaneous combustion occurred at the roof side of coal seam No. 12.

A main cross-cut rock roadway was driven through several coal seams at the level of -150 m. The coal seams in this area were coal seam No. 10, No. 11, No. 12, No. 18 and No. 24. In accordance with the requirements of the regulation, the roadway that penetrates through a coal seam should be lined (sheathed) by cement for the whole length of the coal seam and plus each side by 5m. However, this requirement of surface lining has not been observed completely and nothing happen in cases of coal seam No. 10 and No. 11, which had been penetrated by the rock entry. In case of coal seam No. 12, the cross roadway advanced 30 m ahead toward the coal seam No. 18 after penetrating coal seam No. 12, when the heading operation was halted due to the holidays for two weeks. After the holiday, CO gas concentration was detected at a level more than the threshold. For firefighting, a single sealing wall was built and inert gas (N₂ gas) was injected into the fire area. However, the situation was not improved.

One month later, an investigation group opened the isolated area and made detailed investigation at the site. They concluded that the fire might still sustain in the roof area where the rock entry penetrated the coal seam. Finally, the exposure part of the coal seam No. 12 that penetrated by the rock entry was covered by a cement sheath layer. The floor coal seam exposure part of the rock entry was filled by the injection of the mixture slurry of cement and fly-ash.

According to the results of the investigation committee, there are some factors that might cause the coal spontaneous combustion as follows:

- Coal part on the roof side was collapsed during the driven operation of the rock entry encountered with coal seam No. 12.
- The space created by coal seam collapse was not treated by filling material.
- By the driven operation of the rock entry, the hazardous part was covered with cement lagging plates that enabled air penetration through gapes of them toward coal seam.

A2-1-2 No. 91 Enterprise Coal Mine

No. 91 Enterprise Coal Mine is located inside the Bao Dai Coal Graben, including two minable coal seams of No. 4 and No. 5. The coal mine is located in a high mountainous area of Yen Tu Mountain (1,068 m), the highest mountain in Quang Ninh area. Coal seams in this area are divided by several faults system and folded by the past geological formations. The coal seams are covered with rocks and soil with the depth varied from dozens to hundred meters. Coal seam dip angles and coal seam thickness are not constant due to the past geological activities.

*** Conditions of coal seams with spontaneous combustion**

As shown in the previous chapter, there were several coal spontaneous combustions at No. 91 Enterprise coal mine. The coal seam No. 5 is covered with the soil and rock layers of +530/+565m with the crack system created by mining activities. The coal seam thickness varies

from 0.27m to 15.93m; the coal seam slope varies from 5 to 80°; the coal seam is classified into a lower gassy level.

*** Preparation for the working place of coal seam No. 5 (Figure A2-3)**

Coal seam developing panel is prepared by two cross-cut roadways in rock at the levels of +365 (ventilation) and +300 (transportation). The panel is divided into two part: lower part from +300/+330 and upper part from +330/+365. The long-wall panel with spontaneous combustions belongs to the upper part. Coal seam thickness in this area is around 3.5 – 5 m. Coal seam dip angle is around 10-14°. The mining method is based on a long wall panel that slices the bottom part of the coal seam by drilling and blasting methods with caving upper part of the coal seam.

*** Outbreak of coal spontaneous combustions at coal seam No. 5**

During the period from 2008 to 2012, the coal seam was exploited and had several time records of coal spontaneous combustions. The latest one was occurred in 2011, when higher level of CO gas was detected from the goaf area. Even the hazard did not threatened the safety situation, the firefighting and researching the solutions was conducted based on the approval by upper authorities.

Inert gas injection into the goaf area was applied by using N₂ gas injection and in parallel with inert gas injection the crack systems from the surface and leakage way along the goaf were treated to make inert gas injection effective. The overall target threshold of the O₂ concentration in the goaf area was set at less then 13 %. After two months, the target could be achieved.

According to the results of the investigation committee, there were some factors of the causes of coal spontaneous combustion as follows:

- The design of mine structure for this area did not comply with the prevention of the hazards of coal spontaneous combustion. There are many of penetrated ways for air leakage from the wooden chock systems at upper and lower systems of the long-wall panel.
- The depth of cover soil and rock layers is not thick enough for strata control. By mining activities, the ground subsidence occurred at the area beneath the long-wall goaf. Hence, the crack system was created from the surface to the long-wall's goaf area. (Figure A2-4.)

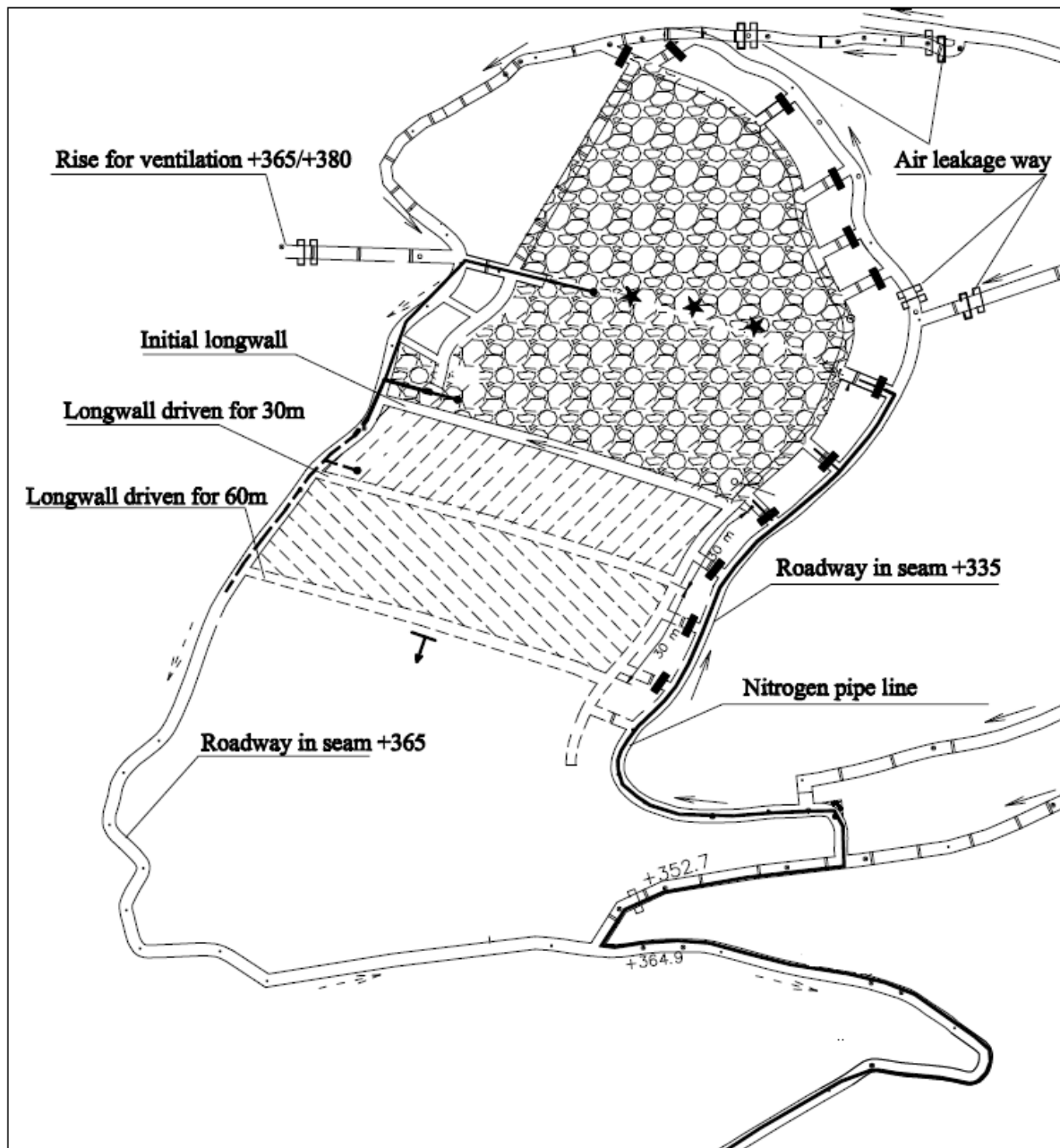


Figure A2-3: Plan view of coal seam No. 5 of No. 91 Enterprise Coal Mine

- The ventilation system was not well controlled from the view point of air leakage into the long-wall goaf area. Because the air flow volume was bigger than the design value, some air went into the goaf area up to the distance around 40 m from the long-wall face (the oxygen concentration in this area was the same as the oxygen concentration at the long-wall face). (Figure A2-5.)
- Even when the air flow going to long-wall was under control, there was some air leakage through crack systems come from the roadway located behind the goaf area.

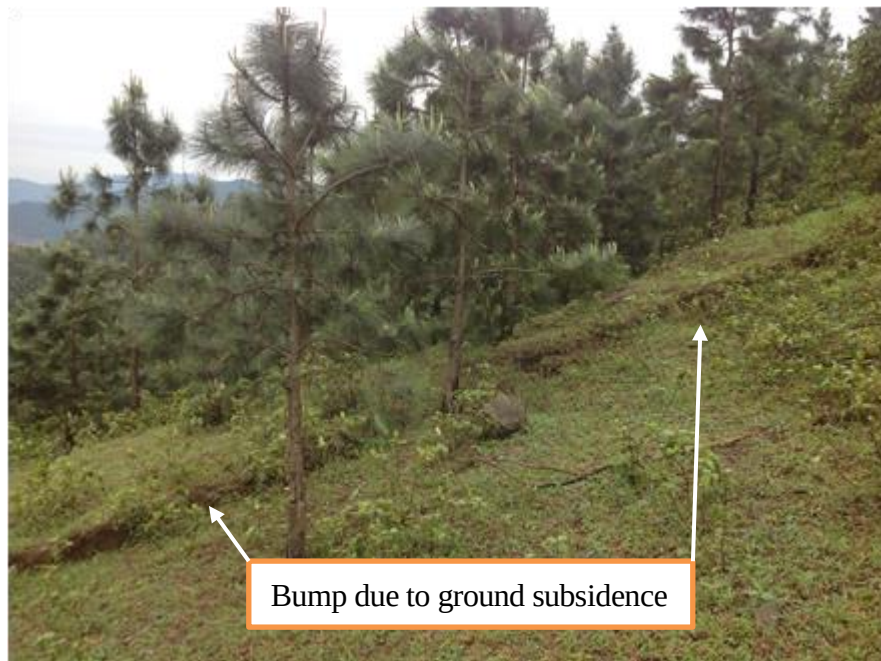


Figure A2-4: Ground subsidence above goaf area of No. 91 Enterprise Coal Mine^{A2-1)}

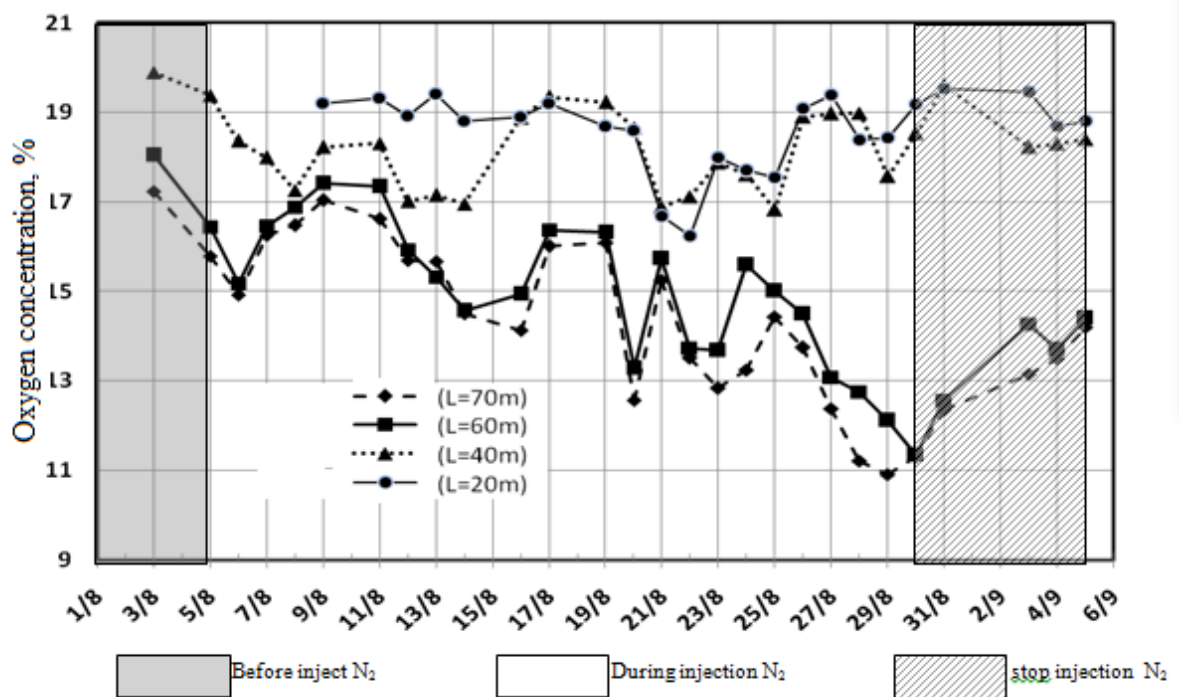


Figure A2-5: Oxygen concentration at each distance in the goaf from the long-wall face

Khanh Hoa Coal Mine in Thai Nguyen Province started as an open pit mine to develop a very thick coal seam more than 20 m, which belongs to Late Triassic Period in Mesozoic Era the same as the Quang Ninh Coal Field that includes Hong Thai Coal Mine, No. 91 Enterprise Coal Mine, Mao Khe Coal Mine and Ha Lam Coal Mine.

After exceeding the economic stripping ratio, the mine started underground developing by installing most of the skeleton main galleries in coal seams. The pit mouths of -87 mL horizontal gallery and -87 m to -183 mL incline shaft are illustrated in Figure A2-6.

A vertical cross section of the underground skeleton structure of Khanh Hoa Coal Mine is shown in Figure A2-2, in which the places of six spontaneous combustions are shown by “●” marks. Three of them occurred at the sub-level coal winning area at around “●” mark positions of in-seam -53 East Gallery in May 2011 (twice) and in March 2012. Position and status of spontaneous combustion is illustrated as shown in Figure A2-7, namely all three spontaneous combustions at the ceiling part of the gallery where air and water penetrations through cracks are alternated between sunny days and rainy days.

Other three spontaneous combustions occurred at newly excavated -183 m level water drainage horizontal gallery in August (twice) and September (one time) both in 2017. The situations of the spontaneous combustion are similar to the case shown in Figure A2-8.



Figure A2-6: Pit mouths Khanh Hoa Coal Mine driven from the outcrop wall of coal seam No. 16 after finishing the open pit mining.

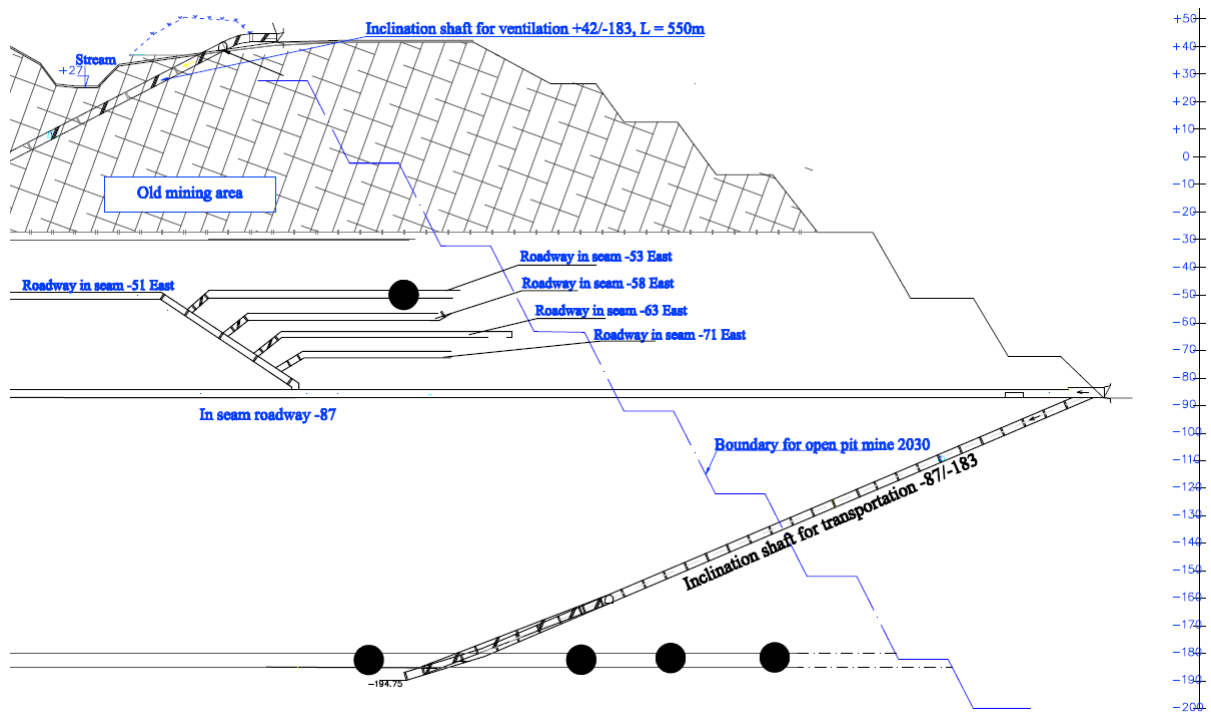


Figure A2-7: Underground skeleton structure of Khanh Hoa Coal Mine

(All the galleries except for the ventilation incline shown in left top located inside the coal seam No. 16. Right top step like boundary is the wall after open pit mining)

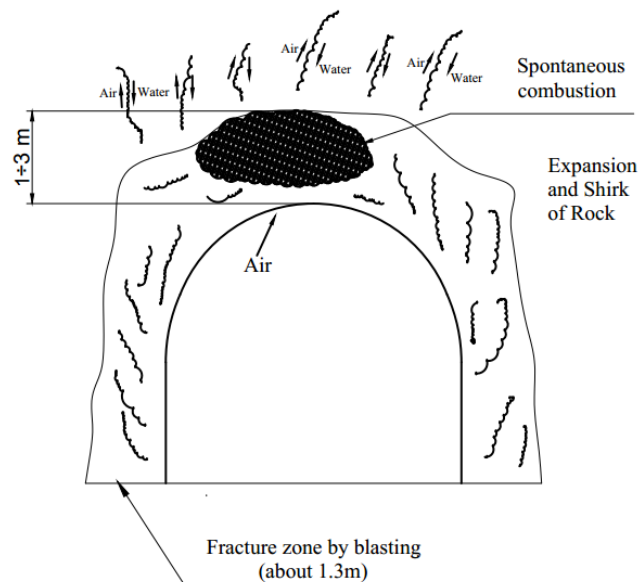


Figure A2-8: Spontaneous combustion at -53 m Level horizontal in-seam gallery to the coal winning faces

A2-1-4 Mao Khe Coal Mine

As introduced in Chapter 1, Mao Khe Coal Mine belongs to the west part of the Quang Ninh Coal Graben (Late Triassic Period in Mesozoic Era). Most coal seams in Mao Khe Coal Mine have the same dip direction from south to north. Coal mine area is located in a hilly terrain area with the heights around 400 m or less. Except for partial open pit mining, most of coal seams are mined by underground mining methods (Figure A 2-9).

The geological conditions are difficult for arranging roadways in seam since old time. The coal seam floor is weak, leading to swelling or floor heave.

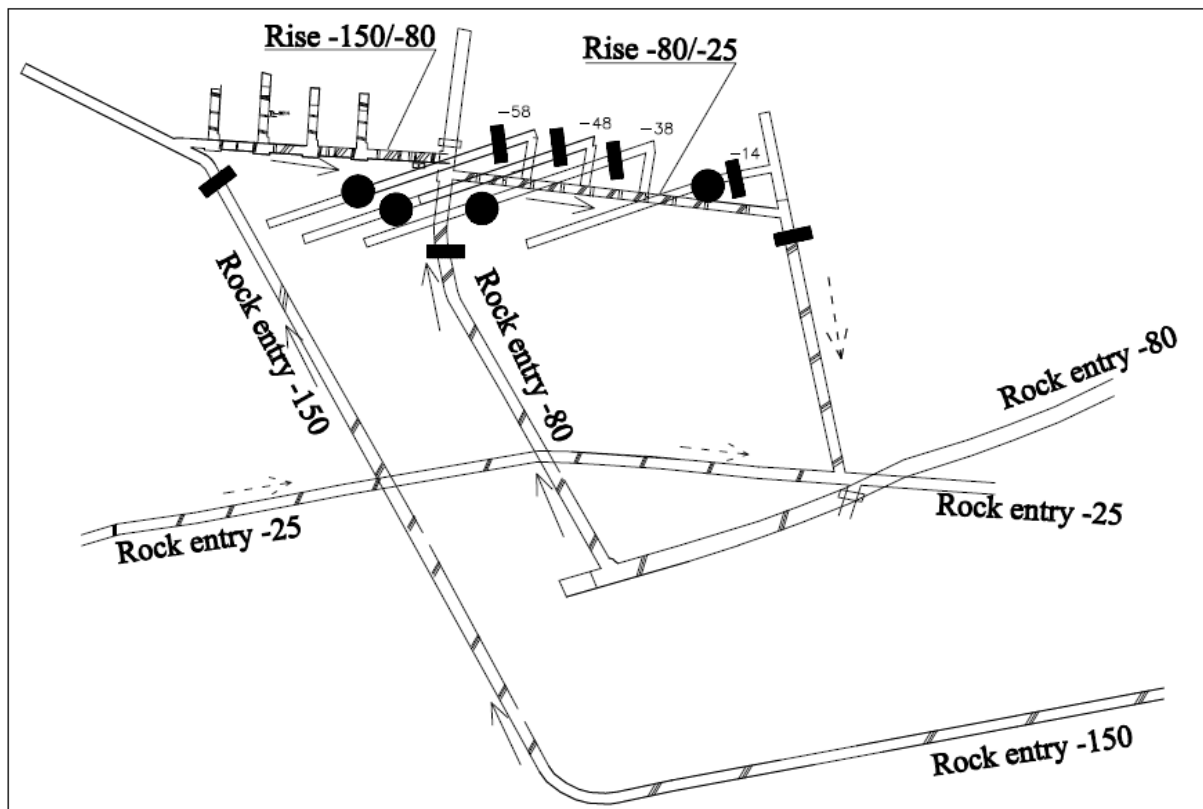


Figure A2-9: Layout of sub-level coal stopping at coal seam No. 10 of Mao Khe Coal Mine

*** Conditions of the coal seam with spontaneous combustions**

The coal spontaneous combustion at Mao Khe coal mine was reported on Jan 13th 2017 at coal seam No. 10, which is a steep slope coal seam with average angle of 35°; the coal seam thickness is 4 or 5 m. the coal seam is classified into a gassy coal seam.

*** Preparation for working place for the coal seam No. 10**

The coal winning area is prepared by three rock entries at the levels of -25 m (ventilation), -80 m and -150 m (transportation). Depending on the coal seam inclination, coal seam No. 10 is divided into several blocks (panel) along the seam strike. For the block with a coal seam inclined less than 40°, a long-wall panel system that exploits the full coal seam thickness by drilling and

blasting method is applied. For the area with the coal seam inclined greater than 40°, the sub-level stopping mining method is applied. The combustion area was found in the latter case. From two level roadways in rock, a rise in rock is driven to connect two levels. From the rise, the short cross-cut roadways are driven to approach the coal seam. Thereafter, in-seam roadways are driven toward to the boundary of block. Coal is mined from the end point of this roadway retreating to the rise. The fresh air is supplied by local forced fans and air pipe system to the coal seam faces. Usually, the interval between two sub-levels is around 10 m.

*** The occurrence of coal spontaneous combustion at coal seam No. 10**

On January 13th, 2017, high concentrations of CO were found at four coal faces of -14, -38, -48 and -58 m levels at the maximum concentration of 344 ppm. The workers at these areas were evacuated and safety measures were applied. Four roadways at each level were isolated by two sealing walls. After that, the inert gas was injected into the isolated area. For several weeks, the gas concentrations did not reduce because of air leakage at these sealing walls. In order to prevent air leakage, three sealing walls were constructed additionally at the levels of cross-cut roadways of -25, -80 and -150 m levels. Up to now, the most gases were reduced except for H₂ concentration still remained at higher than the threshold.

According to the results of the investigation committee, there are some factors affecting the coal spontaneous combustion as follows:

- The design of the coal winning structure for this area does not comply with the prevention of the hazard of coal spontaneous combustion.
- Although those sub-level coal face driving design in this area is arranged for keeping a distance between each other enough for preventing air leakage, some cracks might be created between these levels. The air could penetrate between them and contact with the coal left in the goaf area.
- The area is located not far from the main fan station, hence, the air pressure in this area is the lowest in comparison with other areas. The more fan increases its capacity the more air even from surface could penetrate into their goaf area.

A2-1-5 Ha Lam Coal Mine

(1) General situation of Ha Lam Coal Mine

*** Area and topography**

Ha Lam Coal Mine covers an area extending 5 km from north to south and 5 km from east to west. Ha Lam Coal Mine belongs to Hon Gai Coal Basin (Late Triassic Period in Mesozoic Era) in the center of Quang Ninh Coal Field. Long time ago, Ha Lam coal mine has been exploited since the French colony time at the shallow depth with a single roadway. Coal mine is located near the high way of No. 18. The coal mine has several coal seams in which five coal seams are capable for extraction.

The Ha Lam Coal Mine is located at a valley of lower mountains of around 400 m high. Some area of the surface near coal seam outcrops is exploited by open pit coal mines. The topography of Ha Lam coal mine is mainly shaped by dumping wastes from open pit coal mines of Nui Beo, Ha Tu, etc.

*** Conditions of coal seam with spontaneous combustion**

As shown in Chapter 2, Ha Lam Coal Mine has two coal seams of No. 7 and No. 10 that had coal spontaneous combustions up to date. They are both very thick coal seams with the thickness of 17 m or 20 m, practically. The coal winning strike length is 154 m and the coal seam angle varies from gentle slope to steep slope. Both coal seams are classified into gassy coal seam.

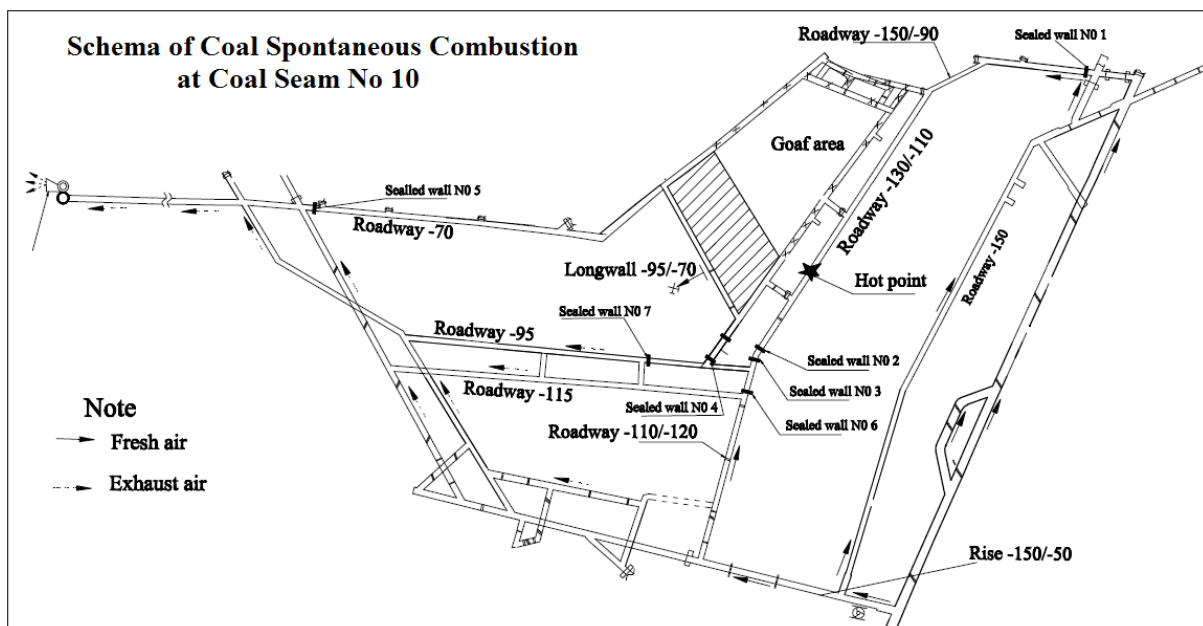


Figure A2-10: Schema of coal spontaneous combustion at coal seam No. 10

(2) Spontaneous combustion at coal seam No. 10

*** Preparation for working place for the coal seam No. 10**

A long-wall panel is prepared by two seam roadways of level -70 and -95 m. Actually, this long-wall face was not operated continuously, because it was prepared only for the requirement of increase of coal production. The long-wall face was operated with an interval of every two weeks just for moving the frame system. During its temporary halt of operation, the ventilation was still kept as the normal condition. Coal seam is mined by a controversial system of retreating long-wall system, and the long-wall direction is perpendicular to the strike of the coal seam. Underneath of this panel, there is another lower level coal panel of -130 m/-150 m. Two panels are isolated by a safety coal pillar with the width of 25 m (Figure A2-10).

*** Occurrence of coal spontaneous combustion at coal seam No. 10**

On June 3rd, 2017, smoke and high concentration of CO gas was found in the roadway -130 m/-150 m at a place far from a T-junction of 210 m. After evacuating all the workers, firefighting solutions were applied to this area.

- To build a sealing wall in the intake air roadway -150 m/-190 m (Sealing No. 1).
- To build two sealing walls in the exhaust air roadway -130 m/-110 m (Sealing No. 2 and No. 3).

After finishing the construction of three sealing walls, the inert gas (nitrogen) was injected into the isolated area through a pipe from sealing walls of No. 2 and No. 3. However, CO gas was also detected at the main fan station. From this fact, it was supposed that the scale of coal spontaneous combustion might increase and the fire products were penetrated from the heating point to goaf area through the coal galleries along the roadway -130 m/-150 m. Hence, additional sealing wall was built at the roadway -70 m used for ventilation from long-wall face (No. 5).

Even after three months of inert gas injection, the CO concentration inside the isolated area was still kept at higher levels. Leaked air with O₂ might be supplied to the isolated area from the crack system around the mining panel. Then, another two sealing walls of No. 6 and No. 7 were built at the positions at the extension area from the sealing wall No. 4 and No. 3 on October 4th, 2017.

Up to now, the gas concentrations inside the isolated area are still remain at abnormal levels.

(3) Spontaneous combustion at coal seam No. 7

*** Preparation for the working place of coal seam No. 7**

Two main horizontal cross-cut roadways were driven to the strike direction of the coal seam at two levels. Mining area is separated by two in-seam roadways both of which are perpendicular to the strike of coal seam. The long-wall panel 7-2-1 was surrounded by the incline transportation roadway and incline ventilation roadway from 170m level to -300m level. The panel is divided into two parts based on the depth of the coal seam: upper part from -170 m to -165 m level and lower part from -165 m level to -300m level. An active long-wall face was located at lower part of the mining panel. Long-wall face length is 120 m, and the advancing speed of the long-wall is 20 m per month. Long-wall face direction was set toward the dip direction of the coal seam. The safety coal pillar between two panels has the width of 20 m.

*** Mining method**

Coal is mined by a long-wall panel system with the top-coal caving method. Coal cutting is carried out by a cutting machine. The coal cutting height is 4.5 m, which is roughly one quarter of the total coal seam height (20 m). When the self-advancing support is advanced toward the long-wall direction, the caving coal behind the supports is recovered through the opening doors

behind the frame. Thereafter, coal is transported to the transportation roadway by a conveyor system (Figure A2-11).

In order to protect safety coal pillar, the tail gate roadway and intake ventilation roadway were not forced to collapse even by withdraw of the steel frame. They are kept by strengthening supports of square chock set timbering.

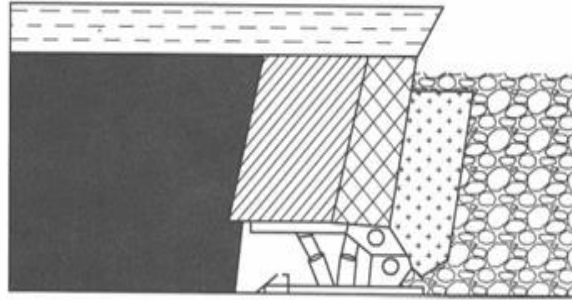


Figure A2-11: Diagram of mining method of full mechanized long-wall with top-coal caving method at coal seam No. 7

*** Occurrence of coal spontaneous combustion at coal seam No. 7**

According to the rule, Ha Lam people should check the gas situation at every sealing wall with a frequency of twice a week. On September 14th 2017, a safety staff found strong smell, and then he checked the gas situation in sealing wall No. 1 (T-junction of rise, goaf area and roadway in seam -165 m level) and detected CO gas of 4,000 ppm. Actually, the sealing walls in this area were built for the purpose of entry people only, then the leakage of air could still pass through. The initial solution was taken by building a new sealing wall next to the old sealing wall. The measurements of gas concentrations are shown in Table 2.5 in Chapter 2. The long-wall No. 7-2-1 was still working at that time. After evacuating the people in the whole area related to the ventilation system, the solution for the coal spontaneous combustion was carried out as follows:

- Strengthening the new sealing wall with the purpose of air leakage prevention as well as installing of gas monitoring tool.
- Injection of inert gas into the isolated area through the sealing wall No. 1.

After several days, some boreholes were drilled through the safety coal pillar from the ventilation roadway of 7-3-1. Boreholes of No. 1, 2 and 3 (8 m long) were used for measuring coal temperature. Borehole No. 4 (29 m long) was used for gas monitoring.

- Periodical measurements of gasses at 4 points along the long-wall.
- Fourth days after, the second injection point was installed and inert gas was injected through the borehole No. 4.

After 14 days of inert gas injection (September 29th 2017), the gas analysis results showed the safety situation for the long-wall face area (Figure A2-12).

After the occurrence of coal spontaneous combustion, the investigation of this incident was carried out in order to find the causes and examine a proposal of prevention methods for this coal

seam. As shown in Chapter 4, Vietnamese coal industry does not have its own methods for research on coal spontaneous combustion. Hence, Ha Lam Coal Mine does not have any prevention measures for coal spontaneous combustion in this long-wall face. There are some factors affecting coal spontaneous combustion as follows:

- The sealing walls at goaf area side do not have enough function for air leakage prevention because of its structure. The roadways were driven by the blasting method that created many cracks around the boundary of the roadway. The sealing walls were built by wood and clay that did not prevent the air leakage sufficiently through its self as well as surrounding sealed wall.
- During the long-wall face advance, the roadway in seam for transportation and ventilation was not completely fall down but remained by the square chock sets of timbering system. The air from long-wall, hence, penetrated into the goaf area by ventilation pressure and promoted the oxidation of coal left in the goaf area.
- The inert gas system was not installed in the long-wall coal face area for blowing out the oxygen in the goaf area.
- The advancing speed of long-wall was lower than the time of coal incubation period toward spontaneous combustion.

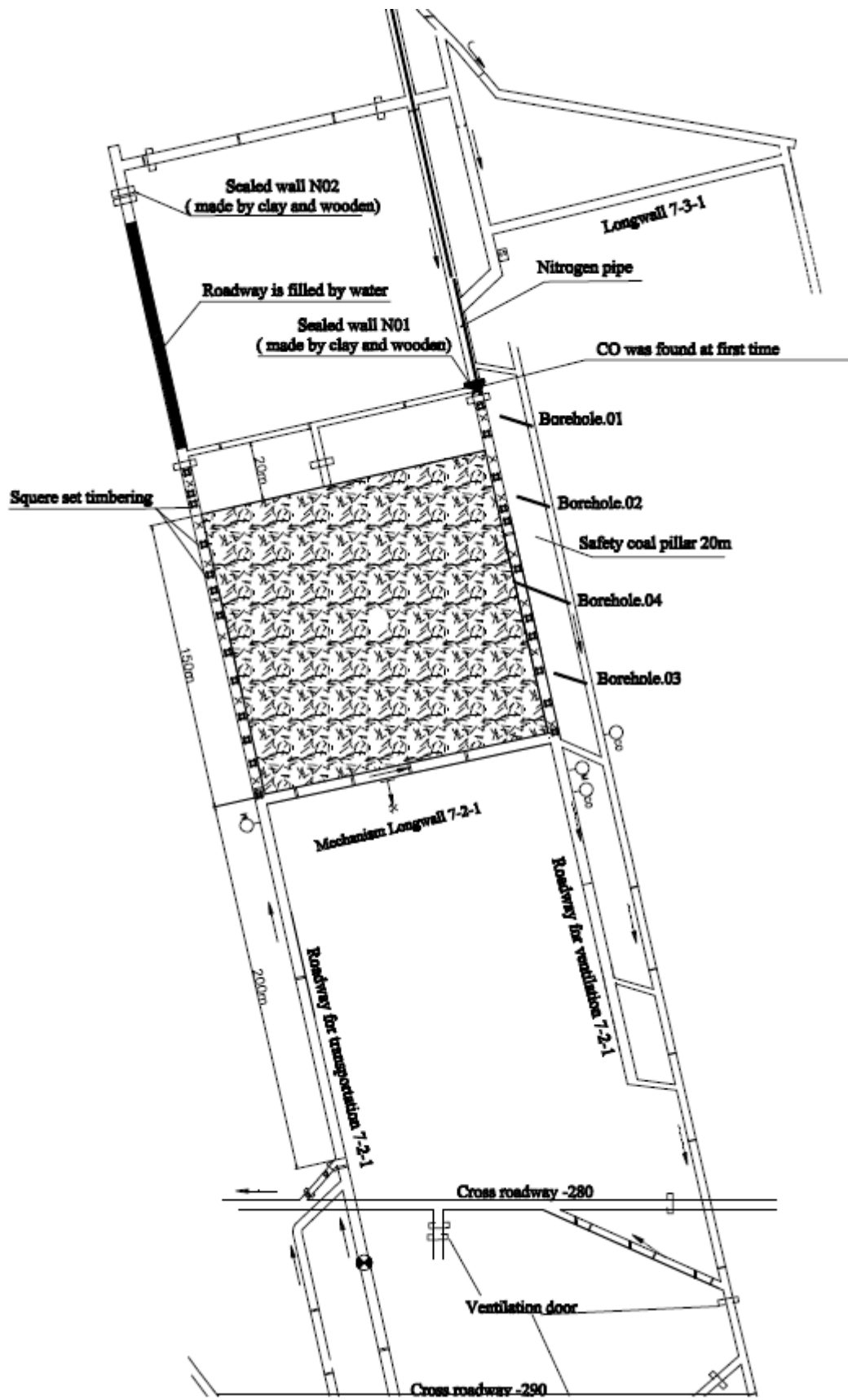


Figure A2-12: Layout of the long-wall coal face 7-2-1 of the coal seam No. 7

A2-2. Site countermeasures against spontaneous combustion

Coal spontaneous combustion is one of the serious accidents or incidents at underground coal mines. It not only makes miner poisoned but also brings other disasters. If the spontaneous combustion lasts an enough time, other disaster could occur such as gas explosion or open fire to enlarge the damages. Besides that, the explosion caused by coal spontaneous combustion might repeat itself until its energy sources end. As shown in the Table 2-2 to Table 2-6 of the analysis results of gas samples from inside the sealing of coal spontaneous combustions at Hong Thai, Mao Khe, Ha Lam coal mines, CO concentrations were much higher than the regulation threshold. The current regulation of CO gas concentration for Vietnamese underground coal mines is 17 ppm. The existence of higher gas concentrations also make firefighting difficult at underground coal mine. When coal spontaneous combustion occurs at underground coal mine, high methane gas content or methane gas emission also enlarges the damages for miner by the risk of gas explosion. According to the annual classification of underground coal mine by its dangerous methane gas, some underground coal mines were classified as supper level of methane gas emission such as Mao Khe Coal Mine or high level of gas content such as Quang Hanh, Duong Huy, Ha Rang coal mines, etc. among these coal mines, the coal spontaneous combustion occurred at coal seam No. 10 of Mao Khe Coal Mine on January 13th 2016. The coal spontaneous combustion at underground coal mine also makes impact of fire on some underground coal mine facilities such as belt conveyor, ventilation duct, and electricity cable. As shown in the investigation report, the spontaneous combustion at Mao Khe, Hong Thai, No. 91 Enterprise and Ha Lam coal mines showed strong impacts on these facilities.

From the view point of economy, the spontaneous combustion at Vietnamese underground coal mine have shown their consumption of much money for counter measures against spontaneous combustion. According to the report, the time to cope with spontaneous combustion takes at least three months even for the case of spontaneous combustion at cross-cut roadway at Hong Thai Coal Mine. As usual, the area of spontaneous combustion was completely closed by sealing walls to prevent O₂ supply to spontaneous combustion process. Then, the sealed area is made inert by introducing N₂ gas and gas samples are taken from inside this area for inspection. The sealed area is considered as normal situation when the gas analysis results show the same level of normal condition.

In case of coal spontaneous combustion in the period of coal mine preparation, the work is stopped completely. In case of Vietnamese long-wall technology, the gate roadways are driven by blasting that makes many crack system around the roadway face. The new mining panel is prepared by using a couple of roadways, in which the tail gate roadway is separated from a previous long-wall panel by the small protection coal pillar. This coal pillar could not prevent the air leakage through the crack system. As a result, the air leakage from ventilation pressure goes more inside the coal pillar and induces its oxidation process. In case of weak ventilation, the

heat from this oxidation process is accumulated and results in coal spontaneous combustion, lately. The coal spontaneous combustions at Hong Thai (2006 and 2008), Khanh Hoa (2010) and No. 91 Enterprise (2012) correspond to this cause.

Another scenario of coal spontaneous combustion in Vietnam is related to the coal left in the goaf area. The coal left in goaf area also comes from the mining method at a coal winning face. The coal seam thickness is usually higher than the height of the frame support system. The main part of coal seam is cut by using explosive or by a cutting machine, the other coal at roof part area is automatically fell down behind the frame support system. Coal from this part cannot be recovered 100 %. Hence, some coal is left behind the long-wall frame system. That remained coal also has some advantageous factors for encouraging the oxidation process because of completely fresh surfaces created by caving process by such leaked air with O₂ through crack systems.

Coal spontaneous combustion at the goaf area makes the solutions more difficult for extinguishing measures. The necessary countermeasures would increase with tremendous scale and time. It also makes impact on underground ventilation network, transportation system, and finally, the coal production at that coal mine. In case of coal seam No. 24 of Hong Thai coal mine, the long-wall had spontaneous combustion during its installing process for hydraulic frame. Just after finish installing process, the hot spot of spontaneous combustion occurred behind the frame system with the signs of high temperature and CO gas concentration. At the first step, this long-wall panel was closed for treatment of spontaneous combustion. All of long-wall's facilities were permanently remained inside the sealed area. Then, the whole mine ventilation system should be changed due to the safety reason.

For the long term of firefighting, most equipment left inside the sealed area gives the coal mine very strong negative damages. Some coal resources in the relevant spontaneous combustion area should often be abandoned permanently. As shown in the coal mine report at Hong Thai, Khanh Hoa, No. 91 Enterprise and Mao Khe, at least, 30 % of coal sources still remains in these sealed area at three coal mines. The incident of coal spontaneous combustion also made more difficult to keep coal production stable for energy security policy.

A2-3. Basic skeleton structures of anthracite coal mines in relation with spontaneous combustion

(1) Less awareness of spontaneous combustion

Because it is said that anthracite hardly starts spontaneous combustion, most of anthracite coal mines in Vietnam have been designed putting the weight to “economy and efficient advantages of coal development”. From such considerations common countermeasures for precaution against spontaneous combustion shown below are not taken into account in the coal mine skeleton design as well as coal seam developing plan:

- ✓ Sectional coal winning panel

- ✓ Under seam rock entry gate roads (except for Mao Khe Coal Mine)
- ✓ Lining of coal wall facing with air (in seam gallery, coal pillar, coal seam penetrated part, etc.)
- ✓ Side wall sealing of goaf
- ✓ Sealing of abandoned (finished) coal winning panel
- ✓ Full goaf filling coal winning (Kakkuchi mining method, etc.)

One of the typical underground skeleton structures can be seen in case of Khanh Hoa Coal Mine that started underground development by in-seam permanent galleries for both intake and exhaust galleries as shown in Figure A2-6. These galleries were developed from the coal seam wall of old open pit mine. As shown in Table 2-1, this Khanh Hoa Coal Mine have had six spontaneous combustions up to date.

(2) Geological limitations

1) Variety of coal seam thickness up to 20 m or more

In both Bao Dai Coal Graben and Quang Ninh Coal Graben, there are more than 60 anthracite coal seams that widely varied thickness from 0.1 m to 92.2 m.^{A2-2)} Even in one coal seam, the thickness varies widely. In addition, one coal mine has multiple minable coal seams with thickness from 2 or 3 m up to 20 m such as Mao Khe or Ha Lam coal mines shown above.

2) Steeper inclination of coal seams

A variety of inclinations of coal seams in Quang Ninh Coal Field also range from several degree up to nearly vertical as shown in Figure A2-6 (Khanh Hoa Coal Mine). General coal seam conditions in Quang Ninh Coal Fields can be summarized in Table A2-1:

Table A2-1: Coal seam conditions of Quang Ninh Coal Field

Coal Basin	Bao Dai	Mao Khe	Hong Gai	Cam Pha
Geological Time	Late Triassic Period in Mesozoic Era			
Geological Structure, Inclination	Syncline	Monocline	Monocline + Fold	Syncline (E-W)
	30 – 50°	25 – 45°	-	20 – 80°
Maximum thickness	26.8 m	29.4 m	56.6 m	92 m

3) Mining method taking account for the geological condition

As shown above, anthracite coal seams in Quang Ninh Coal Field vary in both thickness and inclination so that Vietnamese coal industry should adopt many mining method taking account for the geological conditions of owned mining area. Those mining methods are summarized in

Chapter 1 from Figure 1-3 to 1-12. Even in one mine, they adopt different mining methods due to the variations of underground coal seam conditions.

In such varied underground condition, it is impossible to recover 100 % of coal resources by the practical mining methods especially in cases of thick coal seams or steeply inclined coal seams so that it is noted that sources of combustion always remain inside goaf. In addition, many of safety coal pillars should be arranged to keep the safety of mining area and these coal pillars could be the source of spontaneous combustion if these naked walls are kept in touch with ventilation directly. Furthermore, geological structures of syncline, monocline and fold often force a rock entry to penetrate coal seams at many places and such naked coal seam parts could also be the sources of spontaneous combustion.

From the discussion above, it is concluded that potential sources of spontaneous combustion exist inevitably in many places of underground coal mines so that secondary countermeasures against spontaneous combustion are essential.

4) Secondary countermeasures against spontaneous combustion^{A2-3)}

There are many method as the secondary countermeasures against spontaneous combustion, all of which aim to prevent human disasters and secondary disasters, and to minimize the loss of natural resources and the delay of coal production. These methods can be summarized as follows:

- (a) Underground skeleton structures
 - i) Layout of permanent galleries
 - ii) Partition coal winning (sealing-off after coal winning, to minimize the abandon of coal resources even in case of spontaneous combustion)
 - iii) Appropriate coal pillar arrangement (protection of multiple use in-seam galleries, prevention of air leakage, etc.)
- (b) Mining methods
 - i) Decrease of the remaining coal as less as possible
 - ii) Coal winning speed (not let remaining coal to keep enough incubation period to start spontaneous combustion)
 - iii) Treatment of goaf (side wall sealing and other prevention measures of air leakage into the goaf)
- (c) Ventilation
 - i) Appropriate ventilation pressure (avoid higher ventilation pressure)
 - ii) Prevention of short-circuit of ventilation
 - iii) Prevention of air leakage into goaf
- (d) Prevention measures
 - i) Lining of coal faces (No naked coal faces at coal pillars and coal seam penetration part)
 - ii) Repair of caving parts of in-seam gallery

- iii) Back-filling
- (e) Early detection of spontaneous combustion
 - i) Strengthen central monitoring (increase of number of sensors, higher sensitivity sensors, etc.)
 - ii) Strengthen site patrol and observation (gas and smell monitoring, temperature measurement, ventilation check [including air leakage], etc.)
 - iii) Precise gas analysis
 - iv) Training of workers
- (f) Back-up system in case of outbreak of spontaneous combustion
 - i) Higher quality sealing wall construction standard
 - ii) Fly ash slurry delivering station
 - iii) Water supply system to underground
 - iv) Storage of essential goods (gas sampling bags, smoke tubes, materials for sealing walls, etc.)

Reference

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- A2-2) 財団法人 石炭エネルギーセンター、「世界の炭田の概要」、1999 年 3 月、pp. 411 – 435 (Japan Coal Energy Center, “Outline of Coal Fields in World”, March 1999, pp. 411 - 435)
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