

JANUSZ CYGANKIEWICZ *

ABOUT DETERMINATION OF SUSCEPTIBILITY OF COALS TO SPONTANEOUS COMBUSTION USING AN ADIABATIC TEST METHOD

O OZNACZANIU SKŁONNOŚCI WĘGLI DO SAMOZAPALENIA METODĄ TESTU ADIABATYCZNEGO

The research work on spontaneous combustion carried out in the Central Mining Institute's Ventilation Division, Katowice, has been discussed in the article. During the tests, the airflow was forced through a crushed coal sample of 520 g placed in a reaction chamber and heated up to 400°C, which caused oxidation of coal and changes of its humidity. Changes of coal temperature, humidity and oxygen consumption with time characterise the dynamics of the process of spontaneous combustion. In order to explain the nature of those changes and to determine the thermal balance of spontaneous combustion, a mathematical model of the phenomenon was elaborated, based on simplifying physical and chemical assumptions. Also, the results of tests on coal samples taken from "Borynia", "Marcel", "Andaluzja" and „Julian” mines were presented.

Key words: occupational safety, mine ventilation, coal, spontaneous combustion, testing method.

Skłonność do samozapalenia zależy od szeregu czynników, przede wszystkim od następujących jego własności: stopnia metamorfizmu (typu węgla), zawartości siarki pirytowej, wilgotności, zawartości części lotnych, porowatości, kruszalności, własności petrograficznych, przewodności cieplnej.

W literaturze naukowej opisanych jest wiele metod oceny skłonności węgla do samozapalenia. Znane są zwłaszcza metody kalorymetryczne, metoda pomiaru absorpcji tlenu, metoda temperatury krytycznej (punkt przecięcia), czy też metoda regresji wielowymiarowej. Z polskich metod wymienić należy metodę Olpińskiego z modyfikacją wprowadzoną w 1993, opisaną w normie PN-93/G-04558 oraz metodę perhydrołową Maciejasa i Lasonia.

W pracy zaproponowano metodę badania skłonności węgla do samozapalenia polegającą na obserwacji procesu samozagrzewania danej próby węgla w warunkach zbliżonych

* GŁÓWNY INSTYTUT GÓRNICTWA, 40-166 KATOWICE, PL. GWARKÓW 1

do rzeczywistych. W tym celu zaprojektowano i zbudowano prototypowe stanowisko badawcze, którego zasadniczym elementem jest kalorymetr adiabatyczny. Pomiarzy wykonano od temperatury 40°C do około 185°C. W pracy przedstawiono wyniki badań przeprowadzonych na próbkach węgla pobranych z pokładów: 415/1-2 KWK Borynia, 419 KWK Andaluzja, 707 KWK Marcel i 503/2 KWK Julian. Reakcje zachodzące w komorze reakcyjnej kalorymetru w oparciu o metodę podaną przez Moore'a (1962).

Szybkość zachodzących reakcji została określona na podstawie konsumpcji tlenu, według wzoru (5). Po podstawieniu wartości wydatku powietrza V_a oraz objętości komory reakcyjnej V_0 otrzymano wzór (10) umożliwiający obliczenie współczynnika k i jego logarytmu. Następnie wyznaczono równania dwóch prostych odpowiadających rozmieszczeniu punktów pomiarowych, przy czym jedna prosta odpowiada wynikom uzyskanym dla niższych temperatur, a druga dla wyższych. Współrzędne przecięcia tych prostych pozwalają wyznaczyć temperaturę krytyczną węgla.

Doświadczenia, które objęły węgle o różnych skłonnościach do samozapalenia pozwalają podzielić te ostatnie na dwie zasadnicze grupy:

a) węgle, które podczas samozagrzewania osiągnęły temperaturę 185°C,

b) węgle, które tej temperatury nie osiągnęły, gdyż po początkowym okresie wzrostu temperatura zatrzymała się na pewnym poziomie, a następnie zaczęła maleć.

Dla węgla, które w kalorymetrze adiabatycznym ulegały samozagrzewaniu i zostały zaliczone do grupy (a), istotny jest czas osiągnięcia temperatury 185°C. Wymienione wyżej własności węgla, tj. wzrost temperatury w kalorymetrze adiabatycznym i czas osiągnięcia temperatury 185°C lub zatrzymanie wzrostu mogą stać się podstawą wstępnej kwalifikacji węgla.

W procesie samozagrzewania węgla w zakresie od temperatury początkowej do temperatury 185°C obserwuje się trzy zasadnicze fazy:

Faza 1 — szybkiego wzrostu temperatury,

Faza 2 — spowolnienia tempa wzrostu temperatury, a czasem nawet chwilowego jej spadku, spowodowanego ciepłem parowania,

Faza 3 — ponownego szybkiego wzrostu temperatury.

Na podstawie wyniku testów w kalorymetrze adiabatycznym i długości okresu t , w którym węgiel osiąga temperaturę 185°C, badane węgle sklasyfikowano do czterech grup.

Słowa kluczowe: bezpieczeństwo, aerologia, węgiel, samozapalność, metoda badań.

1. Bibliographic remarks

Some coals reveal susceptibility to spontaneous combustion. The mechanism of this process is very complicated and has not been sufficiently explained so far. In the presence of air, coal becomes oxidised even at low temperature, which is accompanied by heat emission. Coal is characterised by low specific heat, being approximately four times lower than that of water. Also, the thermal conductivity is low and equal to ca. 0.3 W/(m²K). This means that the heat released causes a relatively high increase of temperature and undergoes slow dissipation. In certain conditions, when accumulation of heat of oxidation takes place, a self — heating centre can originate which sometimes becomes transformed into an endogenous fire.

Two characteristic periods can be distinguished in the process of spontaneous combustion of coal: initial period, also called the incubative one, in which the

increase of chemical activity of reagents takes place, and the period of self — heating of coal which sometimes leads to spontaneous ignition, in the case when proper preventive procedures can not be maintained (B a n e r j e e, 1985; B u d r y k, 1956; B y s t r o Ń et al., 1974; M a c i e j a s z & K r u k, 1977).

During the initial period of contact of the coal susceptible to spontaneous combustion with air, no temperature rise is found, both of coal and surroundings, as the process of physical absorption of oxygen proceeds very slowly.

The contact of coal with atmospheric oxygen is also a cause of weathering, being very harmful for its calorific value and coking properties.

Self — heating and spontaneous combustion of coal and other materials has been the subject of many research works, which, however, did not succeed in explaining all complex phenomena and processes forming them. Many hypotheses and theories were formulated from among which the hypotheses of pyrite, bacteria, phenol and carbon-oxygen complex are worth mentioning. The names of the first hypotheses give the supposed main cause of self — ignition. They are only of historical value. Following the carbon — oxygen complex theory the main role in the self — warming process is played by the ability of coal to adsorb oxygen (adsorption and chemisorption) and emit large quantities of heat. When investigating the first stage of coal oxidation, R. E. Jones and D.T.A. Townsend (K r e v e l e n, 1959) found that the reactivity of adsorbed oxygen was very high in low temperatures. Coal adsorbs 0.1—0.5 cm³ of oxygen per 1 gram of coal substance during 24 hours. It has been proved that low-temperature oxidation of coal begins with the origination of surface hydroperoxides.

The stages of coal oxidation were studied by W. F. Oreshko (1949) who was using a thermal laboratory balance. Most of his experiments were performed at a constant rate in time of the temperature change of the sample. Apart from measuring the temperature and variations of the mass of coal sample, a quantitative chemical analysis of the gases emitted was performed, in particular of carbon monoxide and dioxide, as well as of water vapour.

On the basis of these experiments, W. F. Oreshko distinguished 4 stages of the oxidation process. These were characterised by different activation energies *E*. In the first stage, proceeding up to the temperature of ca. 70°C, the mass of the sample increases, probably as a result of chemisorption of oxygen. When the peroxide is formed, the activation energy equals to 12.5—16.8 kJ/mole. In the second stage, between the temperatures 70—150°C, the mass of the sample decreases, which is a result of decomposition of the adsorption complex accompanied by origination of carbon monoxide and release of hygroscopic moisture. The activation energy is equal to ca. 25 kJ/mole. When the critical temperature of ca. 60—80°C is reached, the adsorption complex begins to decompose, the rate of the process of its oxidation increases which also accelerates the chemical reaction the main product of which being CO. The process of decomposition of peroxides takes place, which entails accumulation of additional amount of oxygen on the porous surface of the particle. During this stage, following Oreshko, in the temperature up to 120°C the process

takes place in which the main products are carbon monoxide and dioxide, water vapour, oxalic acid, unsaturated hydrocarbons of a specific smell. After reaching the temperature about 150°C, the oxidation process undergoes further acceleration. In the third stage, in the range of temperatures between 150—230°C a considerable amount of heat is emitted and the mass of the sample is increased as a result of origination of stable oxygen complex. The activation energy increases up to 67 kJ/mole. With further increase of temperature, up to ca. 300°C, coal combustion begins, the process being counted in the fourth stage. Emission of large amount of heat takes place together with a rapid decrease of mass of the sample and origination of gaseous combustion products and soot. The activation energy determined for the fourth stage is in the range from 104 kJ/mole to 151 kJ/mole.

Coal is an organic polymer with a variable amount of aromatic and aliphatic coal, the composition of which changes following the progress of the carbonisation process. Apart from hydrocarbons, it contains mineral compounds and water. The rock coal is composed, first of all, of the elements: hydrogen, oxygen, nitrogen and sulphur. In the self-heating processes, the physical and chemical reactions take place on the surface of pores of the coal substance. In parallel with the low-temperature oxidation, the sorption phenomena take place together with chemical decomposition of certain oxygen compounds. Lignites and later hard coals are more often subject to spontaneous combustion than anthracites.

The occurrence of an endogenic fire is conditioned by airflow supplying oxygen necessary for reaction. At the same time the air stream takes over and carries away the heat generated in the reaction. The excessive yield provides unlimited, in relation to consumption, amount of oxygen but effectively carries away the availability of oxygen but the heat remains in the place of generation. The flow supplying oxygen for reaction and not dissipating the generated heat is called the critical flow.

The inflow of air and oxygen into the coal substance is facilitated in the case when coal is crushed. In the disintegrated state of coal, the surface of flow and permeability are greater, which increases the access of gas. Small particles have a large specific surface and generally show a tendency to spontaneous heating and combustion. In coal mines, the crushed coal is most endangered by spontaneous heating and combustion. In general, in the laboratory testing of the processes of low-temperature oxidation of coal, the samples of highly disintegrated coal are used, which is the cause of temperature changes appearing in a shorter period of time. The moisture content also affects the course of the self-heating process; the loss of moisture is connected with the consumption of heat, and, causes slowing down of the temperature rise during the self-heating process. The gases saturated with water vapour, filtering through their surroundings of a lower temperature, convey heat (both sensible and latent), and partially condense causing heating and wetting of coal. A. K i m (1977) suggests that at the temperature below 100°C, the heat of wetting can be higher than that of self-heating.

The susceptibility of coals to spontaneous combustion depends on many factors, and, in this connection, there are many methods to assess the reactivity of coal in

relation to oxygen contained in the air. Below, two different classifications of these methods are discussed, as given in the works (B a n e r j e e, 1985; K i m, 1977).

Taking into account the chemical and petrographic composition of coal and the results of investigation on chemical interaction of coal and oxygen and their thermal effects S. Ch. B a n e r j e e (1985) divided the methods of determining the susceptibility of coals to spontaneous combustion into two groups. Among the specific measuring methods to be used in testing the samples, the following were discussed: perhydrol tests, determination of the Olpiński's index and tests conducted using the methods of adiabatic calorimetry.

Another classification of the methods to investigate the susceptibility of coal to spontaneous combustion was elaborated by A. G. K i m (1977) in which four principal groups are distinguished, based on: adiabatic calorimetry, isothermal calorimetry, sorption of oxygen, rate of rise of sample temperature in relation to the temperature of a certain reference system. A similar division was proposed on the basis of the work carried out by M. G a n e y & D. J. H o d g e s (1969).

Calorimetric methods

A d i a b a t i c c a l o r i m e t r y

In the issue of low-temperature oxidation of coals, calorimetry enables to measure the amount of heat in specified, constant, external conditions. Usually, the amount of heat is determined on the basis of temperature change of the sample and the amount of the evaporated moisture. In the calorimetric measurements, either elimination of the heat losses or taking them into account plays an important role. In the case when the heat released from the sample is measured, it is necessary to supply oxygen needed for reaction. The discussed thermodynamic system is an open one, as air and oxygen flow into the reaction chamber, while the gaseous reaction products flow out of it.

In the course of an experiment with an adiabatic calorimeter, the external casing is heated up at the same rate as the temperature of the calorimeter increases. Therefore the difference between the temperature of calorimeter and the casing is close to zero. This state is achieved with the use of many methods, which usually utilise automatic control. The methods of adiabatic calorimetry are particularly suitable in the case of experiments conducted over long periods of time (K i m, 1977; K u c h t a et al., 1979; R e n & R i c h a r d s, 1994; S h o n h a r d t, 1984). These methods were used many times in investigating low-temperature oxidation of coals (M. G a n e y & D. J. H o d g e s, 1969).

The reaction took place in adiabatic reaction chambers built in a metal thermostated tube. Inside the tube there is a sample of crushed coal; the air flows among the particles. In order to limit heat exchange between the reaction chamber and the surroundings, the Dewar flask is used, placed in an oil, water or air bath. When considering the question of bath in adiabatic calorimeters used in testing coals, J. A. S h o n h a r d t (1984) paid attention to advantages and drawbacks of the

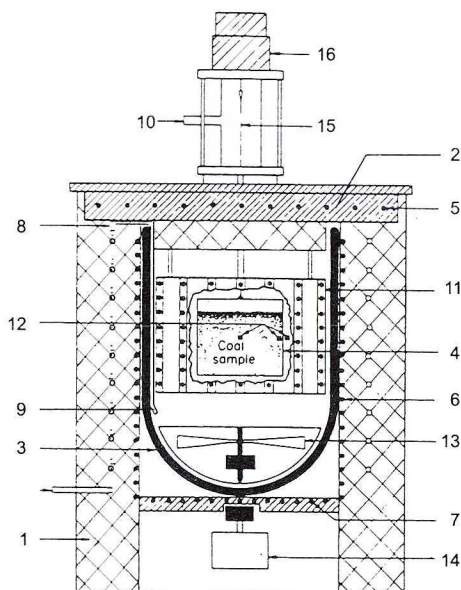
individual solutions. The water bath can be used at low-temperature of the reaction of oxidation, lower than the boiling point, while oil is flammable and produces vapours. On the other hand it is difficult to maintain a required temperature in the air, the metal block as a casing has defined advantages. The heat loss in metals can be determined fairly accurately on the basis of the dimensions and measured values of temperature. It is easier, as compared with other cases, to determine the parameters that are necessary to balance heat exchange. By applying a sufficiently high heat capacity, one obtains effective damping of temperature fluctuations caused by switching over the heating elements. Testing is possible in any range of temperatures (e.g. above 100°C), which is necessary in investigating coals oxidising with more difficulty. In order to equal the temperature of the column surroundings with that of its inside the heaters are used, connected to the control circuits and levelling the differences of temperature on both sides of the wall. Usually, the rise of the temperature of coal is slow and the experiments last for many days. The heat flux produced is so small, that inadequate thermal insulation can be the cause of stopping the rise of temperature and interruption of the self-heating process.

In the J. B. S t a t t and C h e n X i a o D o n g ' s work (1992) the results are discussed of the measurements conducted in the tube of 2 m in length and 0.3 m in diameter. In the tube, 110 kilograms of coal were to be contained, causing some difficulties with preparation of the sample and even crushing the raw coal.

Heat insulation was assisted by 16 heaters placed along the column and reacting to the changes of temperature; the outlet gases were analysed continuously and gas samples were taken each day in 8 locations along the column. The results were presented in the form of changes of temperature, moisture content in the coal, partial pressure of oxygen in the flowing gas and oxygen adsorbed by the coal.

At the Bureau of Mines' Pittsburgh Research Centre a research was conducted (A. K i m, 1977; J. M. K u c h t a et al., 1979) on the process of self-heating of coal, using adiabatic calorimetry methods. The measuring apparatus, presented in Fig. 1, had been designed in such a way so that there would be no heat exchange between the coal sample and its surroundings. Therefore, the temperature of the coal sample and temperature of the air and walls of the reaction chamber were equal to one another. At the same time, under the effect of the system of heaters, the temperature of the air and the reaction chamber followed the measured temperature of the coal sample.

Under these conditions the processes in the coal sample proceed in adiabatic conditions. The calorimeter is composed of a shield vessel 1 made of an insulating material and of an upper cover 2. Inside the shield vessel there is a Dewar flask 3 of the diameter of 150 mm with a steel-wire cage to contain the coal sample. The diameter and the height of the cage are the same and equal to 76 mm. In the shield vessel 1 and cover 2 there are three heaters: upper 5, lateral 6 and lower 7. The heated air flowing into the Dewar flask, through the conduit 8, is fed by means of the metal pipe 9 under the coal sample. The fan 10, magnetically controlled and located in the bottom part of the Dewar flask provides the circulation of the air in the oxidation area.

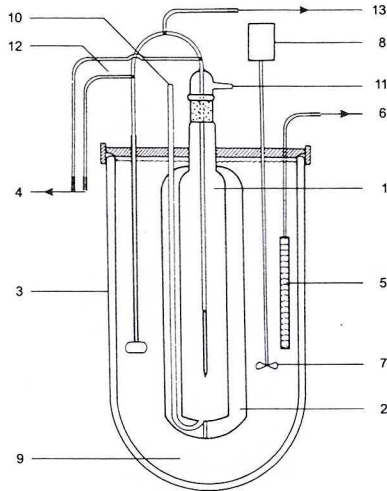


- 1 insulating casing,
- 2 cover,
- 3 Dewar flask,
- 4 cage with coal sample,
- 5 upper heater in the cover,
- 6 lateral heater,
- 7 lower heater,
- 8 air inlet to Dewar flask,
- 9 feed of air below the sample,
- 10 reaction gas outlet,
- 11 internal heater,
- 12 thermocouples,
- 13 magnet-operating air mixing fan,
- 14 small motor with external magnet,
- 15 suspension of sample,
- 16 balance holder.

Fig. 1. Schematic of apparatus for testing spontaneous combustion of coal in quasi-adiabatic conditions, constructed at Bureau of Mines Pittsburgh Research Center

In order to secure temperature measurement, the thermocouples are used, placed in different points of the apparatus and serving for detection of the sample temperature variations and for setting the temperature of the heater.

The electronic control unit provides programmable heating of the measuring system as well as the measurement of the sample and the furnace. The automatic control unit detects the temperature differences of the sample and the furnace equal to ca. 0.020 C. Variations of the mass of a sample during the process of measurement were determined by means of a suspended balance. However, at low changes of the mass, the measurements were charged with a large error. The subject of testing were samples of newly collected coal of 100 g in mass. The grain size of the samples



- 1 reaction chamber,
- 2 vacuum casing,
- 3 Dewar flask,
- 4 connection to potentiometer,
- 5 electric heater,
- 6 connection to autotransformer,
- 7 propeller mixer,
- 8 mixer drive,
- 9 oil bath,
- 10 gas inlet,
- 11 reaction gas outlet,
- 12 fragment of measuring set-up,
- 13 connection to galvanometer.

Fig. 2. Schematic of apparatus for testing spontaneous combustion of coal in quasi-adiabatic conditions, constructed at the University of Nottingham

prepared was in the range from ca. 0.2 to 0.3 mm, and from 0.075 to 0.15 mm. Before the measurement, coal samples contained in the calorimeter were heated with flowing nitrogen to equalise its temperature (with the accuracy of 0.1°C) with the internal temperature of the reaction chamber. Either dry or humid air was passed thorough the chamber.

Earlier, it was set that for the described apparatus, the air outflow had to be 50 cm³/min. The temperature rise of the coal sample takes place exclusively under the influence of the process of oxidation and can not be a result of any effects connected with the presence of heaters. It is important for the temperature of the sample surroundings not to exceed its temperature in the course of the self-heating process.

Next, in the Bureau of Mines' Pittsburgh Research Centre an apparatus was constructed for investigation of low-temperature self-heating of large coal samples, with the mass of 3 kg. The measurements of mass and heat transport taking place in the sample became necessary. The apparatus belongs to the instruments functioning in quasi-adiabatic conditions. The coal sample is placed in an aluminium cylindrical container with the diameter of 146 mm and height 406 mm. The thickness of the cylinder walls is 0.8 mm. The apparatus is set up on the base in which a gas inlet is

mounted. The sample is placed in an insulating casing composed of two layers of an insulating material, filling the space between two aluminium cylinders. The cylinder is closed at the top by a cover.

The equipment used by M a h m e t G u n e y and D. J. H o d g e s (1969) for investigation of low-temperature self-heating of coal, conducted at the Department of Mining Engineering of the University of Nottingham, is composed of a reaction chamber, heat insulation, and monitoring and control system. It was designed in a way enabling to measure the temperature changes of a coal sample in quasi-adiabatic conditions. Its schematic is presented in Fig. 2. An 100 g coal sample is put into the reaction chamber 1, placed in a vacuum casing. The thermal insulation is composed of the 3 gallon Dewar flask 3, provided with a cover. The flask is filled with a bath 9 prepared of the Shell Tellus, type 72, oil with the fire point of 232°C. The electric heater 5, propeller-mixer 7 and temperature sensors are inserted in the bath liquid. Air from the inlet 10 is fed beneath the sample, and the reaction gases are carried away by the hole 11. The figure presents, schematically depicted, connections to power supply, monitoring and control circuit.

Isothermal calorimetry

In this method, the sample is put into a bath of high heat capacity kept at a constant temperature. The heat released in the course of reaction of oxidation or wetting of the sample is measured by thermocouples and dissipated in a relatively large heat container. The value of heat of reaction represents the coal susceptibility to spontaneous combustion.

Method based on sorption of oxygen

Air or oxygen is fed to the coal sample contained in a vessel, and then the container is closed. The gas reaction products are periodically removed, and then air (or oxygen) is added to keep the pressure constant. The amount of the consumed oxygen is determined.

In a flowing system, the amounts of the used air and released gases are used to determine the adsorption of oxygen on coal. The temperature increase per unit volume of oxygen consumed indicates the susceptibility of coal to spontaneous combustion. G. Y. Q i a n (1987) described the method for measurement of oxygen adsorption used in China. It relies on determining the amount of oxygen adsorbed by coal in the laboratory testing conditions. Depending on the amount of oxygen adsorbed at temperature 30°C, calculated per 1 gram of dry coal, a given coal is classified into one of the three groups of susceptibility to spontaneous combustion

Group	Susceptibility to spontaneous combustion	Amount of oxygen, cm ³ /g at 30°C
I	high	> 0.80
II	medium	0.40—0.80
III	low	< 0.40

Critical temperature (point of intersection) method

One of the methods to evaluate the susceptibility of coal to spontaneous combustion is the critical point method. A coal sample is placed in a bath heated with a constant heat flux. At the beginning, the temperature follows the temperature of the bath. Under the influence of the self-heating reaction, heat is generated within the sample, and, at a certain moment, its temperature exceeds the temperature of the bath. The time span from the beginning of heating of the sample to the moment called the point of intersection when the temperature of the sample exceeds that of the bath and the corresponding value of critical temperature are the measure of susceptibility of coal to spontaneous combustion. Many researchers, among them K. K. Feng et al. (1973) conducted the experiments based on the method described. The temperature of the sample in the point of intersection is the lowest temperature at which, in given laboratory conditions, measurable self-heating of coal takes place. Depending on the initial rate of temperature rise and total increase during the test, the following classification is introduced for susceptibility of coals to spontaneous combustion.

Susceptibility of coal to spontaneous combustion	Test result	
	Initial rate of self-heating °C/hour	Total temperature increase °C
Low	<0.6	<2.5
Medium	0.6—1.2	2.5—4.5
High	1.2—2.0	4.5—7.0
Very high	>2.0	>7.0

Multidimensional regression method

R. N. Singh and S. Demirebilek (1987) elaborated a statistical method that takes into account the effects of various properties of coal on its susceptibility to spontaneous combustion. The relationship is analysed between the initial rate of coal oxidation and thirteen independent variables characterising the following parameters of coal: density, calorific value, vanishing moisture, analytical moisture, content of volatile matter, moisture, carbon, total iron, ash, total sulphur, pyrite sulphur, organic and sulphide sulphur and non-pyrite iron.

On the basis of the statistical analysis of the effects of the above listed parameters on the initial oxidation rate, a system of equations was derived for initial rate of coal heating and total increase of temperature that characterise the susceptibility of coal to spontaneous combustion.

Polish methods

Olpiński's method for determination of spontaneous combustion index

This method, modified in 1993, is described in the Polish standard PN-93/G-04558. The principle of this method relies on continuous measurement of

temperature of a pellet prepared from the tested coal and introduced to the air stream of the temperature 237°C and then 190°C, and on determining the rate of temperature rise of the pellet in adiabatic points. Depending on the value of calculated on that basis index Sz^a in [°C/min] and the value of activation energy A , in [kJ/mole], the coals are divided into five groups of susceptibility to spontaneous combustion presented in the table below.

Spontaneous combustion index, Sz^a °C/min	Activation energy of coal oxidation, A kJ/mole	Group of spontaneous combustion	Assessment of susceptibility of coal to spontaneous combustion
upto 80	above 67	I	coal of very low susceptibility to spontaneous combustion
	46 to 67	II	coal of low susceptibility to spontaneous combustion
	below 46	III	coal of medium susceptibility to spontaneous combustion
above 80 to 100	above 42		
above 100 to 120	below 42 or equal to	IV	coal of high susceptibility to spontaneous combustion
	above 34		
above 120	below 34 or equal to	V	coal of very high susceptibility to spontaneous combustion
	not standardised		

Maciejasz's and Lason's methods

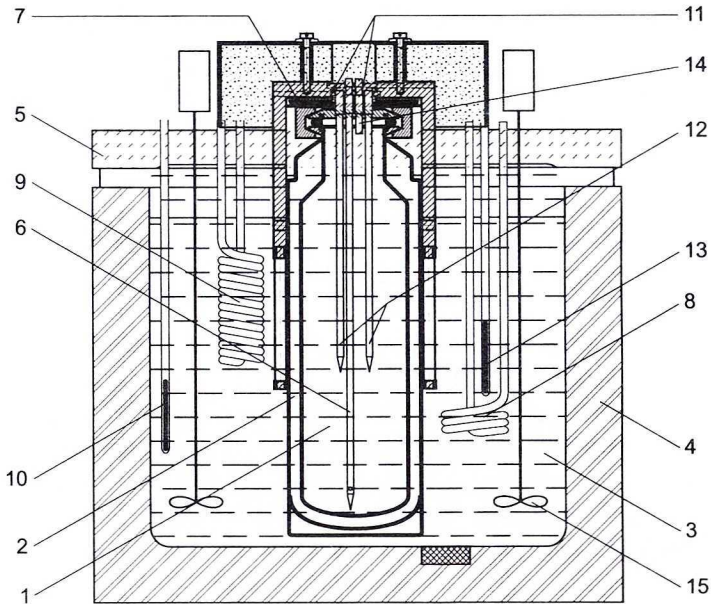
In the Maciejasz's perhydrole method (Maciejasz, 1959; Maciejasz & Kruk, 1977), the rate of temperature increase is determined for a sample adequately saturated with aqueous solution of perhydrole. The 3-gram coal sample screened by the 0.06 mm screen and wetted with 1.5 cm³ of distilled water is mixed with a porcelain mixer. Then the sample is poured into a Dewar flask and 9 cm³ of perhydrole of 29% concentration. The content of the flask is carefully mixed until the temperature of 40—50°C is reached. The temperature of the non-susceptible coals samples rises slightly after long time period and then drops slowly. In the course of reaction of a sample of coal susceptible to spontaneous combustion initially a slight temperature increase takes place. After reaching the temperature of 50°C the reaction gets accelerated and, within short time, the temperature of the sample increases to 90—97°C. Temperature changes of the sample are recorded until the maximum value is reached. The time of measuring the sample, counted down since the moment of applying perhydrole is usually about one hour in the case of coal susceptible to spontaneous combustion while for non-susceptible coal it considerably longer.

M. Lason (1959) as a measure of the susceptibility of coal to spontaneous combustion took the rate of carbon dioxide release in the reaction of adiabatic

oxidation of coal by means of perhydrole (hydrogen peroxide). The sample to be measured and its testing are the same in the Maciejasz's method. The index of susceptibility of coal to spontaneous combustion is calculated from a formula for the rate of carbon dioxide release, in $\text{cm}^3/\text{minute}$, at the temperature of 90°C .

2. Investigation of variations of temperature, humidity of coal sample and chemisorption of oxygen in the self-heating process

To determine the dynamics of heat emission during the reaction of low-temperature oxidation in the bed of crushed coal, the tests were carried out on the testing stand shown in Fig. 3. The principal element of the diameter $d = 70 \text{ mm}$ and height $h = 120 \text{ mm}$, is placed in a thermostat in order to ensure the adiabatic conditions of the process. The total volume of the column is $V_0 = 4.6181 \times 10^{-4} \text{ m}^3$.



- 1 reaction chamber,
- 2 vacuum casing,
- 3 oil bath,
- 4 mineral wool insulation,
- 5 insulating cover of expanded bakelite,
- 6 pipe feeding air to coal sample,
- 7, 8, 9 heaters,
- 10, 11, 12, 13 temperature sensors,
- 14 reaction gas outlet,
- 15 mixers

Fig. 3. Schematic of calorimeter for observation of self-heating of coal, constructed at the Central Mining Institute

A 5-kg mass coal sample taken from a newly uncovered coal mass is placed in an air — tight container, sprinkled with coal fines and delivered to the laboratory. Then, the sample is broken to pieces of 0.1 kg in mass each. 10 portions of coal with the total mass of ca. 1 kg are collected from evenly distributed places. The mass obtained this way is subjected to crushing, in a screw-disk crusher. During the process of crushing, the coal is washed with nitrogen. The coal with the grain size less than 0.5 mm, in the quantity of ca. 0.52 kg is the analytical sample for further testing. The sample is placed in a tight glass container, blown with nitrogen and heated to the required temperature (initial temperature of reaction). Simultaneously with heating the sample, the reaction chamber is also heated. Then, the sample is poured into the reaction chamber and placed on the measuring post. In the course of the experiment, the temperature of the thermostat bath and coal in the reaction column are nearly equal with accuracy $\pm 0.2^\circ\text{C}$, owing to automatic control. The air was fed to the sample having the temperature of the thermostat bath, with the volume flux was $V_a = 2 \text{ dcm}^3/\text{hour}$ or $0.5 \text{ dcm}^3/\text{hour}$. The share by volume of oxygen in the air entering the column was constant in time and equal 20.93%. Before the experiment was started, the coal was heated to 40°C .

Because of heat release during the reaction of oxygen with coal, the temperature increase took place automatically. The oxygen content in the gases flowing out from the column decreased. The test was cancelled at the temperature point of $T = 185^\circ\text{C}$ because of some technical reasons related to the properties of the oil bath.

During the test changes of sample humidity occurred. Water contained in the coal, referred to as coal humidity can be found there either in its proper form (in pores of the sample) or bound by adsorption forces on the molecular level.

The process of heat exchange between the sample and the medium taking water vapour away determines the mechanism of heat and humidity flow inside the sample. In addition, mass and heat flux depend on properties of the physical and chemical properties of coal material. The process of drying can be described using the drying kinetics curves: one describing dependence of average humidity of the drying material on duration in time of the drying process and another curve presenting the volume of water dried out as a function of temperature and humidity of the material.

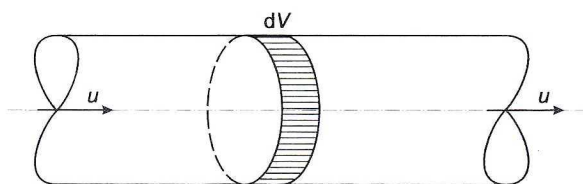


Fig. 4. Volume unit of a reactor

W. J. M o o r e (1962) elaborated a method to study reactions in a flowing system that can be used in the case of the column depicted in Fig. 4. The change of concentration of a reactant in a volume-element of the reactor is linked with mass

loss and inflow of the reactant. In a steady (or quasi-stationary) state the equation of mass balance of the reactant c has the form:

$$r_i dV - u dc_i = 0, \quad (1)$$

where:

dV — differential of volume, as in Fig. 4 (m^3),

u — velocity of air flow through the sample (s),

c_i — concentration of i reactant (oxygen) in the air,

r_i — stream of used reactant in the volume unit of the reaction chamber.

If the reaction is of first degree, the loss of mass for the reactant is:

$$r_i = -k'c_i, \quad (2)$$

where:

k' denotes the reaction rate. Therefore,

$$-k' \frac{dV}{u} = \frac{dc_i}{c_i}. \quad (3)$$

After integration and transformation one obtains:

$$k' = \frac{u}{V_0} \ln \frac{c_{i1}}{c_{i2}}. \quad (4)$$

The coefficient k' is equal to the average time of flow of a molecule of a reactant through the reactor. The ratio V_0/u is the time of contact of the reactants. In the case of the particular reactant one can use another definition of k : the ratio u/V_0 is replaced by the quotient V/V_0 (volume of the reactant versus volume of reactor).

$$k = \frac{\dot{V}}{V_0} \ln \frac{c_{i1}}{c_{i2}}. \quad (5)$$

All the parameters on the right side of the equation (5) are known, so the of reaction rate can be determined.

The Arrhenius' formula

$$k = k_0 \exp\left(-\frac{E}{RT}\right) \quad (6)$$

takes the form:

$$\ln k = -\frac{E}{RT} + \ln k_0. \quad (7)$$

The reaction rate (k) can be described as the concentration loss (or gain in concentration) of a reactant in the unit of time. It is usually assumed that the pre-exponential coefficient does not depend on time. The energy of activation E can be defined as the smallest surplus of reactant's molecules enabling collisions between them to be productive in the chemical sense (Device, 1991; Frank - K a -

mieniecki, 1987; Glasstone & Lewis, 1960; Karapetjanc, 1983; Moore, 1962).

When the energy of activation is constant the formula (7) in the $\ln(k)$, $1000/T$ co-ordinate system is of linear type. Taking the calculated values of k one obtains the points that can be approximated by a single line section or by sections of two lines (Fig. 5). For each of the lines one can determine the constants m and b :

$$\ln(k) = -\frac{E}{1000RT} \cdot \frac{1000}{T} + \ln k_0 \quad \ln(k) = m \cdot \frac{1000}{T} + b. \quad (8)$$

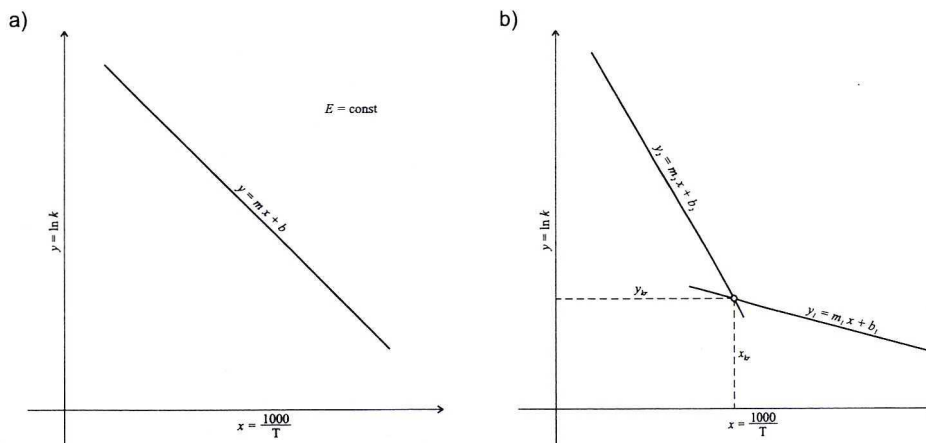


Fig. 5. Reaction rate as a function of temperature

These values are used to calculate the activation energy E and coefficient of frequency, i.e. pre-exponential k_0 . The following notation is introduced:

$$x = 1000/T, \quad y = \ln(k). \quad (9)$$

The co-ordinate of individual points x^i , y^i obtained on the basis of measuring results and calculations are distributed in such a way that they can be approximated by two straight-line sections. The course of the relationship in the $1000/T$, $\ln k$ co-ordinate system can be interpreted as a result of superposition of two straight-line sections 1 and 2. The co-ordinates of the point of intersection are called critical, x_{kr} , y_{kr} , in particular those of the critical temperature T_{kr} .

Physically, this means that the reaction proceeds following two mechanisms — at lower temperatures the process following up the line 1 prevails, while at higher temperatures in accordance with the relationship 2. At medium temperatures when, in a certain part of the coal mass the temperature is higher then critical and in another lower, both mechanisms occur simultaneously.

The values of activation energy in low-temperature process of coal oxidation given by J. M u z y c z u k (1974) and many Russian researchers (E. J. G l u z b e r g, 1986; W. G. I g i a s h e v, 1987) equal to $E = 20-80$ kJ/mole. The lower values of

this energy characterise susceptible coals, conversely, the upper ones, non-susceptible to spontaneous combustion. In accordance with the Polish standard, the coals most susceptible to spontaneous combustion have the activation energy $E < 34$ kJ/mole, while the coals with low susceptibility to self-ignition $E > 67$ kJ/mole. The experiments show that the rate of reaction of oxidation depends on temperature as well as on disintegration of coal grains, concentration of oxygen and other factors.

3. Discussion of measurements results

In the year 1998, in the Central Mining Institute's Department of Mine Ventilation, a test stand was constructed under the author's leadership for testing spontaneous combustion of coal in adiabatic conditions. The stand is composed of ten pieces of equipment that are schematically depicted in Fig. 3. Over the years 1998—1999, nearly 100 coal samples were tested on the stand, collected from currently mined seams of Upper Silesia and Lorraine (France) coal basins. Presented in this work are the measurement results of 4 coal samples collected from seams: 415/1-2 of Borynia mine, 707 of Marcel mine, 419 of Andaluzja mine and 504/2 of Julian mine.

Coal from the 415/1-2 seam of Borynia mine

In the reaction chamber (Fig. 3), a coal sample, collected at the 415/1-2 seam of Borynia mine, was subjected to testing. Its composition is given in the table below, showing the quantities expressed in percent by weight

W^{ex}	W^a	A^a	V^a	S^t	S^p	H	C^a
1.00	2.00	5.5	29.10	0.90	0.47	4.62	81.02

The following notation has been assumed in the table:

- W^{ex} — expiring humidity,
- W^a — analytical humidity,
- A^a — ash content,
- V^a — volatile matter content,
- S^t — sulphur content,
- S^p — pyrite sulphur,
- H — hydrogen content,
- C^a — carbon content.

The percentage share of grains with diameters, in mm, was

>0.3	0.3–0.2	0.2–0.1	0.1–0.06	<0.06
6.0%	19.4%	30.5%	19.0%	25.1%

Determination of the spontaneous combustion index, carried out in accordance with the standard in force, enabled to qualify the tested coal to the group II of spontaneous combustion. The activation energy was $A = 78$ kJ/mole, spontaneous combustion index $Sz^a = 45^\circ\text{C}$ to $185^\circ\text{C}/\text{min}$, that is the tested coal show low susceptibility to spontaneous combustion. The dynamics of the parameters of coal sample, showing low-temperature heating during calorimetric tests, are presented in Fig. 6—8. The temperature changes are given in Fig. 6. The time period of

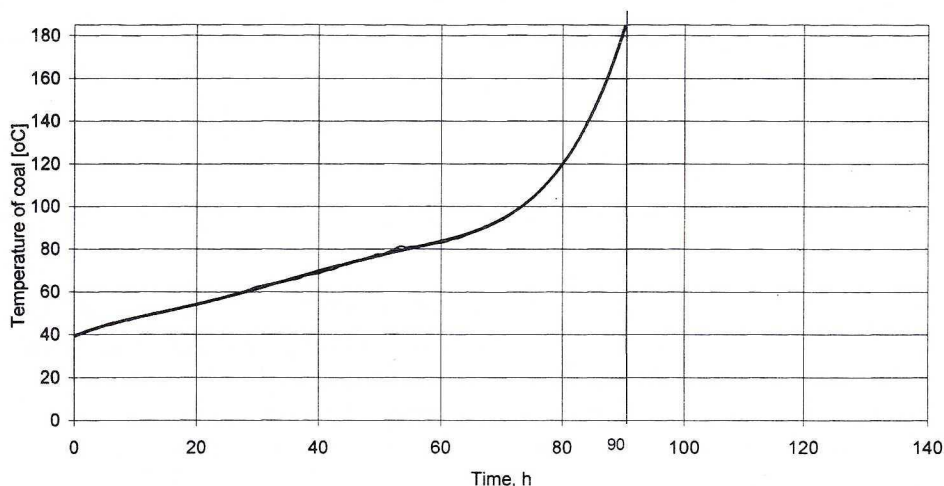


Fig. 6. Changes of temperature of coal from 415/1-2 seam, "Borynia" mine during self-heating in adiabatic conditions

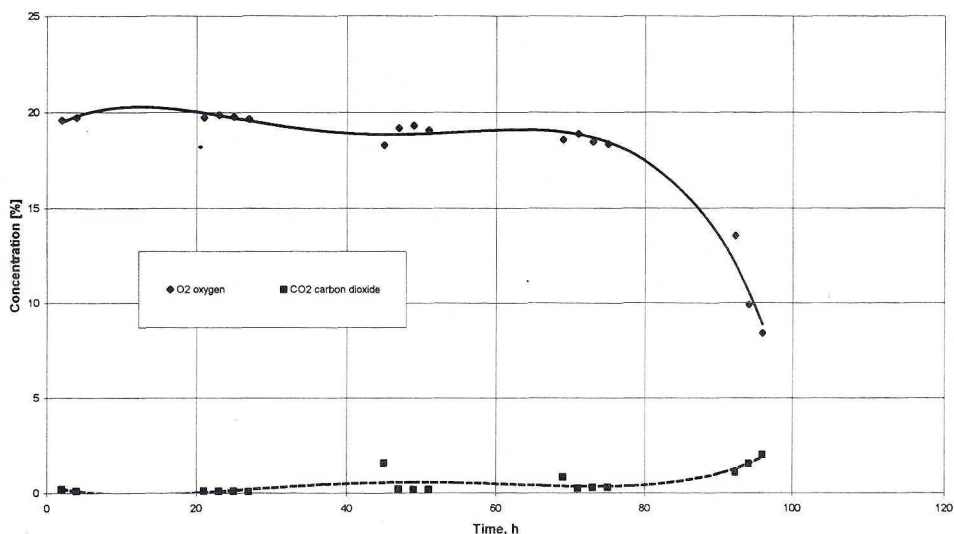


Fig. 7. Changes of oxygen and carbon dioxide concentrations at the outlet of the reaction chamber

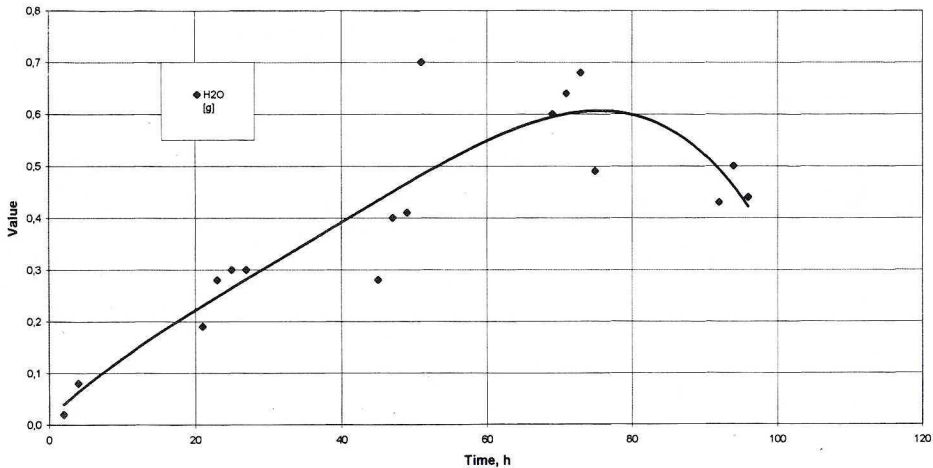


Fig. 8. Water loss in the coal sample from 415/1-2 seam, "Borynia" mine, during self-heating process

temperature increase from the initial value of ca. 40°C — to 185°C was only 90 hours. Therefore, this is coal with very high susceptibility to spontaneous combustion. The rate of temperature rise was changing in time.

During 70 hours after starting the process, the temperature changed from 40–90°C linearly in time. During the next period, the sample temperature rise per time unit increased.

Fig. 7 shows the changes of oxygen and carbon dioxide concentrations measured at the outlet of the reaction chamber. After 70-hour duration of the process, oxygen consumption increased considerably.

Fig. 8 presents the loss of water coming out of the coal sample during the process of low-temperature oxidation. The results of measurements are characterised by a relatively high scatter, therefore another method must be found to measure the mass of water removed from the coal sample. During the 70-hour duration of the process, the mass of H₂O coming out of the coal sample per unit time first increased and then decreased. The maximum intensity of H₂O removal was observed at the temperature of the sample about 90°C.

The rate of the occurring reaction was determined on the basis of oxygen consumption, following the formula (5). After substituting the air consumption $V_a = 5.55 \cdot 10^{-7}$ [m³/s], and the volume of the reaction chamber $V_0 = 4.6181 \cdot 10^{-4}$ [m³], one obtains

$$k = 1.2018 \cdot 10^{-3} \cdot \ln\left(\frac{20.93}{c_2}\right). \quad (10)$$

In table 1 and in Fig. 9 collected are the values of coefficient k of reaction rate and its logarithm $\ln(k)$, and the reciprocal of absolute temperature T multiplied by 1000: $1000/T$ for 17 measurements of coal from Borynia mine.

In the considered case, the equations of straight lines corresponding to the distribution of points defined on the basis of measurement results, in which the sections $y = f(x)$, lie, have the form

TABLE 1
Coal from "Borynia" Coal Mine, 415/1-2 seam.
Values of reaction rate constant (k) and reciprocal of absolute temperature of the coal, T multiplied by 1000

No	$k \times 10^3$	$\ln k$	T, K	$1000/T$
1	0.079516	-9.440	301.96	3.31
2	0.071567	-9.545	303.46	3.30
3	0.071567	-9.547	317.96	3.15
4	0.063671	-9.662	319.56	3.13
5	0.068524	-9.588	321.06	3.115
6	0.075841	-9.487	322.16	3.104
7	0.16138	-8.732	336.16	2.975
8	0.10494	-9.162	337.26	2.965
9	0.09744	-9.236	338.26	2.956
10	0.11185	-9.098	339.26	2.948
11	0.14378	-8.847	353.96	2.825
12	0.12452	-8.991	354.46	2.821
13	0.15092	-8.799	355.56	2.812
14	0.15810	-8.752	356.56	2.805
15	0.52254	-7.557	384.56	2.600
16	0.89973	-7.013	390.86	2.558
17	1.0572	-6.852	397.16	2.518

$$y_1 = -1.2x - 5.668 \quad (11)$$

$$y_2 = -6.667x - 9.993.$$

The section lying on the straight line y_1 corresponds to the measurement results obtained for lower temperatures, while y_2 — for higher ones. The point of intersection of these lines has the co-ordinates $x_{kr} = 2.854$, $y_{kr} = -9.09$. On the basis of the first of formulas (11), it is possible to determine the temperature corresponding to the point of intersection. The temperature value, called critical T_{kr} = is obtained:

$$T_{kr} = \frac{1000}{x} = 350.4 \text{ K} = 77.2^\circ\text{C}. \quad (12)$$

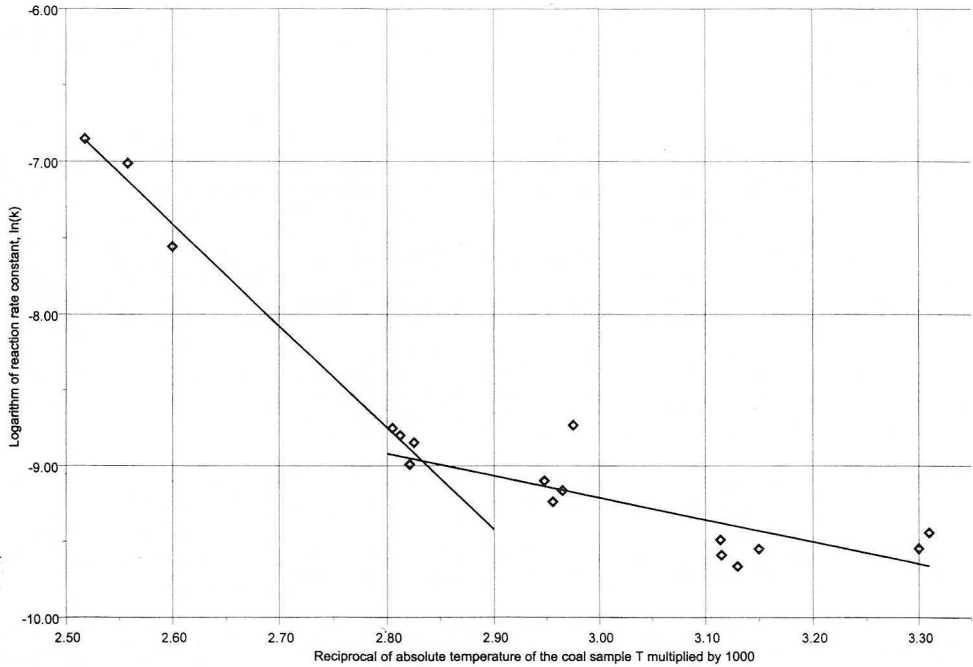


Fig. 9. Values of logarithm of reaction rate constant, $\ln(k)$ for the coal sample from 415/1-2 seam, "Borynia" mine

After the critical temperature T_{kr} is reached, the change of the dynamics of the oxidation process takes place. From the relationship (8) it also follows that, in the temperature range corresponding to linear course of the function $y = f(x)$, the activation energy E has a constant value which can be calculated on the basis of the slope coefficient of the straight line:

$$y = f(x) = mx + b \quad (13)$$

As there is (8),

$$E = -1000 \cdot R \cdot m \quad (14)$$

By substituting the universal gas constant $R = 8.3145 \text{ J/mole K}$, one obtains:

$$E_1 = 1000 \cdot 8.3145 \cdot (-1.2) = 9.98, \quad E_1 = 55.4 [\text{kJ/mole}].$$

At a temperature higher than the critical one, the energy of activation E_2 is more than twice greater than E_1 . The approximate energy balance of the process of self-heating of a coal sample was prepared on assumption that the sensible heat q_{sij} generated in this process proceeding from a state i to the state j can be determined on the basis of the mass of sample m_w and temperature rise ΔT_{ij} :

$$q_{sij} = m_w \cdot c \cdot \Delta T_{ij}. \quad (15)$$

It has been assumed in the calculations that the value of the mass of sample $m_w = 0.52 \text{ kg}$, specific heat $c_w = 1200 \text{ J/(kg} \cdot \text{K)}$, and the temperature rise ΔT_{ij} is equal to the difference $T_j - T_i$ of temperatures measured.

On the other hand, the latent heat is related to the enthalpy carried away by water vapour. Its value is the product of moisture ΔW_{ij} and specific enthalpy of water evaporation r_w :

$$q_{1ij} = \Delta W_{ij} \cdot r_w. \quad (16)$$

In the calculations, the changes of latent heat of water evaporation r_w vs. temperature have been taken into account. The value ΔW_{ij} was determined in measurements.

The amount of the water vapour released by some samples was higher than the humidity determined in the way as specified in the standard. The surplus water vapour is probably a result of chemical reactions accompanying the process of coal self-heating. In this case, the amount of heat calculated for the process of water evaporation (16) is lowered in relation to real latent heat.

The results of calculations made on the basis of measurements carried out for Borynia mine are presented in table 2. In the last but one column of the table, the

TABLE 2
Coal from "Borynia" Coal Mine, 415/1-2 seam.
Values of heat of reaction during spontaneous combustion process

Process from <i>i</i> to <i>j</i>	Time, h	<i>T</i> , °C	ΔT °C	r_w J/g	<i>Q</i> sensible, J	ΔW g	<i>Q</i> latent, J	ΣQ J	$\Sigma \dot{Q}$ W
0—1	2	28.8		2433.8		0.065	158	158	0.0220
1—2	2	30.3	1.5	2430.2	936	0.065	158	1094	0.152
2—3	16.5	44.8	14.5	2395.6	9048	1.165	2791	11839	0.199
3—4	2	46.4	1.6	2391.8	998.4	0.2675	640	1638	0.228
4—5	2	47.9	1.5	2388.2	936	0.27	645	1581	0.220
5—6	2	49.0	1.1	2385.6	686	0.27	644	1330	0.185
6—7	18	63.0	14.0	2351.6	8736	3.215	7560	16293	0.251
7—8	2	64.1	1.1	2348.8	686	0.405	951	1637	0.227
8—9	2	65.1	1.0	2346.3	624	0.43	1009	1633	0.227
9—10	2	66.1	1.0	2343.7	624	0.45	1055	1679	0.233
10—11	18	80.8	14.7	2306.0	9172.8	4.8705	11231	20404	0.315
11—12	2	81.3	0.5	2304.7	312	0.582	1341	1653	0.230
12—13	2	82.4	1.1	2301.9	686	0.6	1381	2067	0.287
13—14	2	83.4	1.0	2299.4	624	0.6	1380	2004	0.278
14—15	17	111.4	28	2227.1	17427	4.864	10833	28260	0.462
15—16	2	117.7	6.3	2210.0	3931.2	0.485	1072	5003	0.695
16—17	2	124.0	6.3	2192.4	3931.2	0.485	1963	4995	0.694
0—17	95.5		95.2		59358.6	19.089			

sum was given of the sensible and latent heat, and in the last the heat flux calculated on the basis of the values from the last but one column and duration of the process from the state i to j .

Figures 6 to 12 show the results of measurements performed in the adiabatic calorimeter. For coal from "Borynia" mine, seam 415/1-2 one obtains:

$$\begin{aligned} y_1 &= -1.2 \cdot x - 5.668, & E_1 &= 9.98 \text{ kJ/mole,} \\ y_2 &= -6.667 \cdot x - 9.933, & E_2 &= 55.4 \text{ kJ/mole,} \\ x_{kr} &= 2.654, & y_{kr} &= -9.09, & T_{kr} &= 77.2^\circ\text{C.} \end{aligned}$$

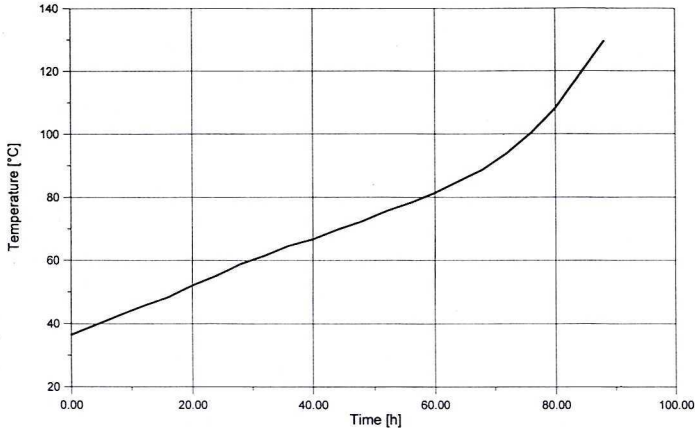


Fig. 10a. Changes of temperature of coal from 707/1-2 seam, "Marcel" mine during self-heating in adiabatic conditions

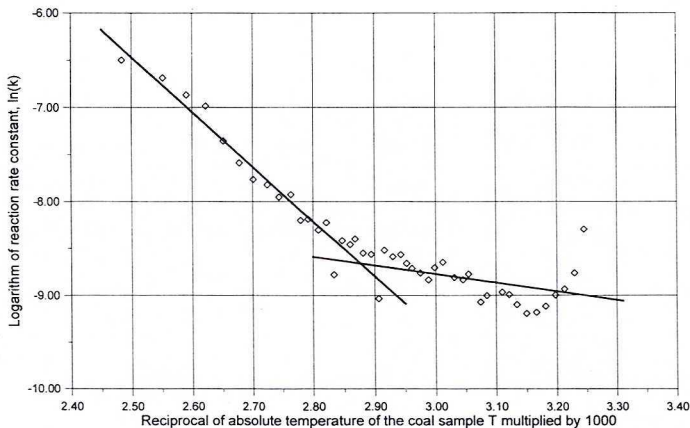


Fig. 10b. Values of logarithm of reaction rate constant, $\ln(k)$ for the coal sample from 707/1-2 seam, "Marcel" mine

Below are presented similar calculations performed for coal from the following coal mines:

“Marcel” mine, seam 707

Parameters of coal sample from 707 seam of “Marcel” mine; the values given in weight percents

W^{ex}	W^a	A^a	V^a	S^t	S^p	H	C^a
0.80	2.37	6.15	30.15	0.60	0.17	4.98	84.22

$$y_1 = -1.471 \cdot x - 4.34, \quad E_1 = 12.23 \text{ kJ/mole},$$

$$y_2 = -5.833 \cdot x - 8.167, \quad E_2 = 48.5 \text{ kJ/mole},$$

$$x_{kr} = 2.866, \quad y_{kr} = -8.55, \quad T_{kr} = 75.8^\circ\text{C}.$$

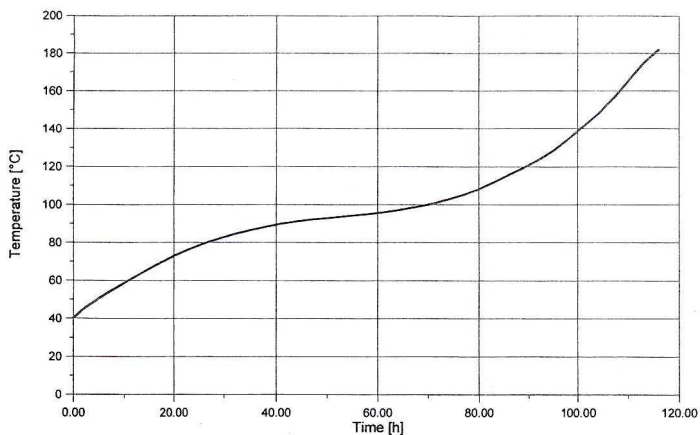


Fig. 11 a. Changes of temperature of coal from 419 seam, “Andaluzja” mine during self-heating in adiabatic conditions

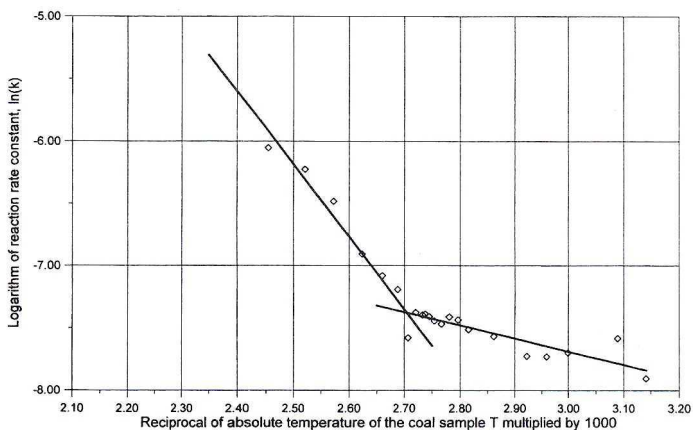


Fig. 11 b. Values of logarithm of reaction rate constant, $\ln(k)$ for the coal sample from 419 seam, “Andaluzja” mine

“Andaluzja” mine, seam 419

Parameters of coal sample from 419 seam of “Andaluzja” mine

W^{ex}	W^a	A^a	V^a	S^t	S^p	H	C^a
1.50	8.94	4.97	33.26	1.90	1.42	4.43	67.52

$$y_1 = -0.6897 \cdot x - 5.483,$$

$$y_2 = -6.5 \cdot x - 10.31,$$

$$x_{kr} = 2.718, y_{kr} = -7.358,$$

$$E_1 = 5.73 \text{ kJ/mole},$$

$$E_2 = 54.04 \text{ kJ/mole},$$

$$T_{kr} = 94.8^\circ\text{C}.$$

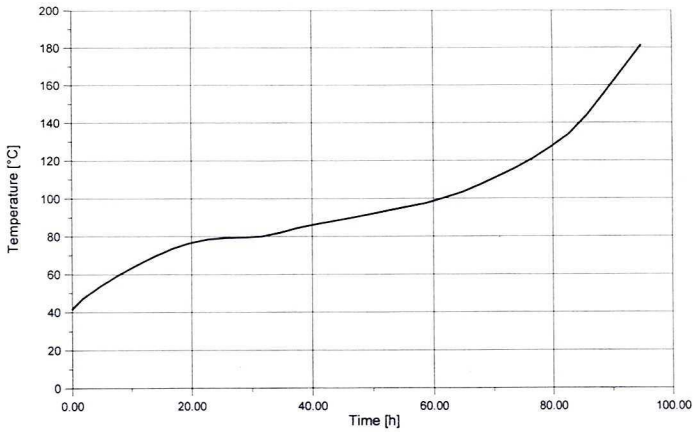


Fig. 12 a. Changes of temperature of coal from 502/4 seam, “Julian” mine during self-heating in adiabatic conditions

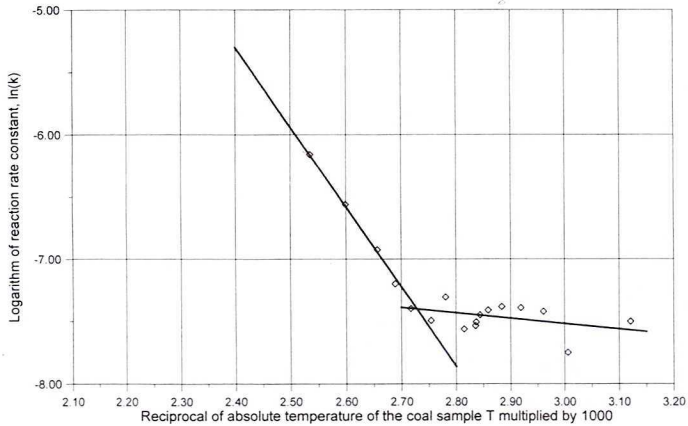


Fig. 12 b. Values of logarithm of reaction rate constant, $\ln(k)$ for the coal sample from 502/4 seam, “Julian” mine

“Julian” mine, seam 504/2

Composition of coal sample from 504/2 seam of “Julian” mine

W^{ex}	W^a	A^a	V^a	S^t	S^p	H	C^a
3.02	8.22	4.93	33.20	0.69	0.43	3.97	72.11

$$\begin{aligned}
 y_1 &= -0.8696 \cdot x - 5.05, & E_1 &= 12.23 \text{ kJ/mole,} \\
 y_2 &= -5.83 \cdot x - 8.43, & E_2 &= 48.5 \text{ kJ/mole,} \\
 x_{kr} &= 2.717, \quad y_{kr} = -7.415, & T_{kr} &= 94.9^\circ\text{C.}
 \end{aligned}$$

Note that at the lowest output of air flowing through the crushed coal sample, after the temperature of 150°C had been reached, considerable consumption of oxygen took place together with decrease of its concentration in the outlet gases, followed by the reduction of the rate of oxidation process. The points relating to these results were not taken into account when determining the activation energy E_2 .

4. Summary

The susceptibility of coal to spontaneous combustion depends on a number of factors: degree of metamorphism (coal type), pyrite sulphur content, humidity, volatile matter content, porosity, crushability, petrographic properties, heat conductivity.

A method has been proposed in the paper for investigating the susceptibility of coals to spontaneous combustion relying on observation of the process of self-heating of a given coal sample in the conditions close to real ones. To this end, a prototype test stand has been designed and assembled, the basic element of which is an adiabatic calorimeter. Then the measurement was carried out from the initial temperature of 40°C up to ca. 185°C . The experiments allow to divide coals of various susceptibility to self-heating into two main groups:

- coals which, in the course of self-heating reached the temperature 185°C ,
- coals which did not reach this temperature, as, after initial period of increase, the temperature stabilised at a certain level, and then started to decrease.

For the coals that got self-heated in the adiabatic calorimeter and were counted into group (a), important is the time of reaching the temperature of 185°C .

The above mentioned properties of coals, i.e., temperature increase in the adiabatic calorimeter, and the time of reaching 185°C or stoppage of increasing, can be a basis for initial classification of coals.

In the self-heating process of coal, in the range from the initial temperature up to 185°C , three main phases are observed:

- phase 1 — fast temperature increase,
- phase 2 — retardation of the rate of temperature increase, and sometimes its momentary drop, which is caused by heat of evaporation,

phase 3 — renewed fast increase of temperature.

On the basis of the results of tests in an adiabatic calorimeter, and duration t , in which coal reaches the temperature 185°C , the tested coals were classified into four groups.

Group of susceptibility of coal to spontaneous combustion	Result of test in the adiabatic calorimeter
Low susceptibility	$t > 500$ h
Medium susceptibility	$200 < t \leq 500$ h
High susceptibility	$100 < t \leq 200$ h
Very high susceptibility	$t < 100$ h

The proposals above should be considered as initial ones. They can change on the basis of results of further investigations.

The assessment of susceptibility of coal to spontaneous combustion carried out on the basis of petrographic tests is in accordance with the results of calorimetric tests. Another classification was obtained using the index Sz^a . This relates in particular to the coal from the 415/1-2 seam of Borynia mine. The calorimetric and petrographic tests point at high susceptibility of coal to spontaneous ignition, while the value of Sz^a makes it count this coal to those of low susceptibility to spontaneous combustion.

A certain drawback of the calorimetric method is the necessity to supply for testing only new, non-oxidised coal samples. Such samples should be delivered to the laboratory in air-tight containers.

I am convinced that the calorimetric method of determining the susceptibility of coal to spontaneous combustion, being the closest to real conditions, will find wide application in the Polish mining industry.

The presented investigations were performed within the research project No T12A00515 "Numerical modelling of the process of low-temperature self-heating and self-combustion of hard coals", financed by the National Committee for Research.

REFERENCE

- Banerjee S. C. 1985. Spontaneous combustion of coal and mine fires, A. A. Balkema, Rotterdam.
- Banerjee S. C., Banerjee B. D., Chankravorty R. N., 1970. Rate studies of aerial oxidation of coal at low temperature ($30\text{--}170^{\circ}\text{C}$). Fuel 49, No. 3, p. 324—331.
- Brötz W. 1964. Podstawy inżynierii reakcji chemicznych. WNT, Warszawa.
- Budryk W. 1956. Pożary i wybuchy w kopalniach. Cz. 1, Pożary podziemne. WGH, Katowice.
- Bystron H., Jaron S., Kolodziejczyk B., Markefka P., Strumiński A. 1974. Pożary podziemne. Poradnik Górnika, t. 3, cz. III. Śląsk, Katowice.
- Denby B., Ren T. X. 1992. A knowledge-based decision support system for spontaneous combustion control. The Mining Engineer, Vol. 151, March, p. 253—258.

- Deńca A., 1991. Niskotemperaturowe utlenianie węgla kamiennych *Wiadomości chemiczne*, t. 45, p. 101—118.
- Feng K. K., Chakravorty R. R., Cochrane T. S. 1973. Spontaneous combustion — a coal mining hazard. *C/M Bulletin*, October, p. 75—82.
- Frank-Kamieniecki D. A., 1987. *Diffuzja i ciepłopieredacza w chemiczkiej kinetyce*. Nauka, Moskwa.
- Glasstone S., Lewis D., 1960. *Elements of physical chemistry*. D. van Norstrand Company, Inc. Princeton.
- Gluzberg E. J., *Teoreticheskie osnovy prognoza i profilaktiki szachnych požarow*. Nedra, Moskwa.
- Guney M., Hodges D. J., 1986. Spontaneous heating of coal. Part 1. *Colliery Guardian*, 1969, February, p. 105—109.
- Guney M., Hodges D. J., 1969. Spontaneous heating of coal. Part 2. *Colliery Guardian*, March, p. 173—177.
- Igiszew W. G., 1987. *Borba s samowozgaraniem uгля w szachtach*. Nedra, Moskwa.
- Karapetjan M. Ch., 1983. *Wstęp do teorii procesów chemicznych*. PWN, Warszawa.
- Kim A. G. 1977. Laboratory studies on spontaneous heating of coal — a summary of information in the literature. U. S. Bureau of Mines, I. C. 8756.
- Krevelen van D. W., Schuyer J., 1959 *Węgiel. Chemia węgla i jego struktura*. PWN, Warszawa.
- Kuchta J. M., Rowe V. R., Burgess D. S., 1979. Spontaneous combustion susceptibility of U. S. coals. Bureau of Mines, RI 8474.
- Lasoń M., 1959. Szybkość wydzielania dwutlenku węgla w reakcji węgla kamiennego z perhydrolem jako metoda badania samozapalności węgla kamiennych. *Arch. Górn.*, t. 4, No. 1—2, p. 55—63.
- Maciejasz Z., 1959. Wskaźniki skłonności węgla do samozapalenia. *Arch. Górn.*, t. 4, No. 1—2, p. 125—134.
- Maciejasz Z., Kruk F., 1977. *Požary podziemne w kopalniach*. Cz. 1. Śląsk, Katowice.
- Moore W. J., 1962. *Physical chemistry*. Prentice Hall, London.
- Muzyczuk J., 1974. Samozagrzewanie węgla w procesie jego utleniania strumieniem powietrza w warunkach zbliżonych do adiatermicznych. *Prace GIG, komunikat No. 665*. Katowice.
- Olpiński W., 1952. Analiza wyników masowych oznaczeń samozapalności węgla. *Prace GIG, kom. 130*. Katowice.
- Olpiński W., 1959. Kalorymetryczne oznaczanie ciepła utleniania węgla w temperaturze około 50°C. *Arch. Górn.*, t. 4, No. 1—2, p. 111—123.
- Qian G. Y., 1987. Research on the use of spectrum technique for determining the liability of coal to spontaneous combustion Fushun Coal Research Institute of Technology, P. R. China (in Chinese).
- Oreszko W. F., 1949. Izuczenije okislenija iskopajemych uglej. *Izw. Akadem. Nauk. SSSR, Otd. Tech. Nauk.* No. 11, p. 1642—1648.
- Ren T. X., Richards M. J., 1994. A computerised system for the study of the spontaneous combustion of coal. *The Mining Engineer*, November, p. 121—127.
- Shonhardt J. A., 1984. Calorimeter design and the assessment of self heating in coal. *The Coal Journal*, November, p. 79—85.
- Singh R. N., Demirbilek S., 1987. Statistical of intrinsic factors affecting spontaneous combustion of coal. *Mining Science and Technology*. Vol. 4, April, p. 155—165.
- Smith A. C., Lazzara C. P., 1987. Spontaneous combustion studies of US Coals. US Bureau of Mines, RI 9079.
- Stott J. B., Chen X. D., 1992. Measuring the tendency of coal to fire spontaneously. *Colliery Guardian*, January, p. 9—16.

REVIEW BY: PROF. DR HAB. MIECZYŚLAW ŻYŁA, KRAKÓW

Received: 03 January 2000.