

Innovative Technologies for Exploration, Extinction and Monitoring of Coal Fires in North China

Understanding self-ignition of coal

A literature study elaborated in co-operation of
Deutsche Montan Technologie GmbH (DMT)
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COAL FIRE RESEARCH - A SINO-GERMAN INITIATIVE



Abstract

The aim of this paper is to describe the process of self-ignition of coal to the experts and scientists from various disciplines who are participating in this project.

In a common sense, the phenomenon of self-ignition of coal means the onset of exothermic chemical reactions and a subsequent temperature rise within the combustible material, without the action of an additional ignition source.

For better comprehension of this process, origin and chemical components of the combustion material “coal” as well as the coalification process are briefly discussed.

A review of the most important factors contributing to the oxidation and the possible self-ignition of coal is given.

Finally, the products emitted during the oxidation, self-heating and in-situ combustion of coal are summarised and, some prediction methods of spontaneous combustion are mentioned.

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1 Introduction

In the late 19th, century Great Britain was in the height of the Industrial Revolution and the Empire, and coal played a significant role in both. In 1878, the British coal production amounted to 133 million tons, 50% of the world production, with some 15 million tons per year being exported. The bulk of this coal was transported by sailing ships, much of it being stockpiled at strategic points around the globe as bunker for the new steam ships of the Royal Navy and the American Navy. The loss of coal cargoes, ships and lives through known cases of spontaneous combustion had become a serious problem. From 1871 – 1880, the number of known cases of spontaneous combustion occurring in British registered vessels totaled 152, with 68 total and 84 partial losses. Lloyd reported that in the year 1874 the incident of spontaneous combustion of coal at sea were greater for ships engaged in long voyages and also proportional to the tonnages of the cargo. Official inquiries into lost or damaged ships were required by the Board of Trade under the Merchant Shipping Act and were held at the British port nearest to the disaster. These inquiries reported several scientific theories that may cause spontaneous combustion and precautions for coal shipping were given /1/

Since then numerous investigations were made to achieve a complete understanding of spontaneous combustion of coal, the processes of oxidation and self-heating of coal in order to develop prevention strategies /2/ due to the problem not being limited to transportation but also occurring whilst mining and storing. Uncontrolled coal fires are widespread and coincide with the occurrence of coal deposits /3/.

Between 1983 and 1992, approximately 13 percent of underground coal mine fires were caused by spontaneous combustion of coal in the United States /4/.

During the combustion process large quantities of greenhouse gases are produced and emitted and these emissions cause environmental pollution to air, water and soil endangering the local population with serious health hazards. Uncontrolled coal seam fires may cause surface subsidence which might damage buildings, infrastructure etc., as well as generating enormous financial losses /3/.

2 Coal

Coal is a sediment, organoclastic in nature, composed of lithified plant remains, which has the important distinction of being a combustible material. Alongside the intended combustion, coal possesses the capability of spontaneous ignition. For a better comprehension of this phenomenon a short roundup of the origin of coal is presented.

2.1 Precursors of coal

The organic portion of plants is built by lignin, carbohydrates and proteins as well as other polymers. These higher polymers are considered as contributors to the organic matter of coal. But the relative amount of each of these integrants varies in great margins with the particular species of a plant as well as the relative stage of growth of the plant.

The fact that each of the compound types in themselves underlie a very broad classification indicates the general complexity of the original plant debris. It is this fact, probably more than any other single contribution, that determines the complexity of the final “coal molecule” /5/.

2.1.1 Lignin

Lignin has been considered one of the most important substances involved in the transformation of plant constituents into coal /5/. It is this material which is responsible for the lignification of vegetable tissue. It may be considered as a cement binding the cellulose fibres into the firm structure.

Although the structure of lignin has not been fully clarified, it is very probable that lignin is a polymer built up by structural elements with the same skeleton as phenyl propane (Figure 2.1).

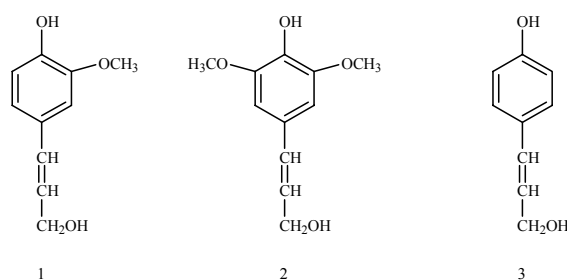


Figure 2.1: Monomeric units of and in lignin: (1) coniferyl alcohol; (2) sinapyl alcohol; (3) *p*-coumaryl alcohol /5/

One of the first substantial steps of the coalification of woody tissue is the transformation of lignin in wood into xylite in lignite /6/.

2.1.2 Carbohydrates

The simple sugars, or monosaccharides (i.e. glucose and xylose), are the building blocks of the more complex carbohydrates /6/. Carbohydrates sustained their name due to the fact that they conform approximately to the general formula $C_n(H_2O)_m$ /5/.

Although the monosaccharides do occur as such in nature, it is more common to find them occurring as high molecular weight polysaccharides. It is the polysaccharides that most probably contribute to the source material, especially the two well-known polysaccharides cellulose and starch. The fibrous tissue in the cell wall of plants and trees contains cellulose, and starch also occurs throughout the plant kingdom in various forms but usually as a food reserve.

In contrast to the somewhat speculative chemistry of lignin, the structural chemistry of many polysaccharides, especially cellulose and starch, is fairly well defined. For example, the molecular formula of cellulose is

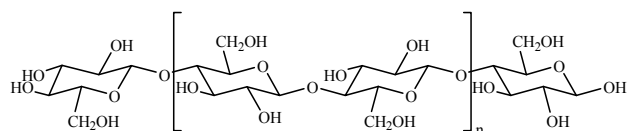


Figure 2.2: Structural model of cellulose (slightly modified) /4/

Speaking in general terms, two high molecular weight polysaccharides, having the general formula $(C_6H_{10}O_5)_n$, wherein n may represent several hundreds or even several thousands units, may have been incorporated into the plant debris and were eventually built-in, albeit in an altered form, into the coal structure.

2.2 Coalification

Coalification is the alteration of vegetation to form peat, succeeded by the transformation of peat through lignite, subbituminous, bituminous, semi-anthracite to anthracite coal. The degree of transformation or coalification is termed the coal rank (Figure 2.3).

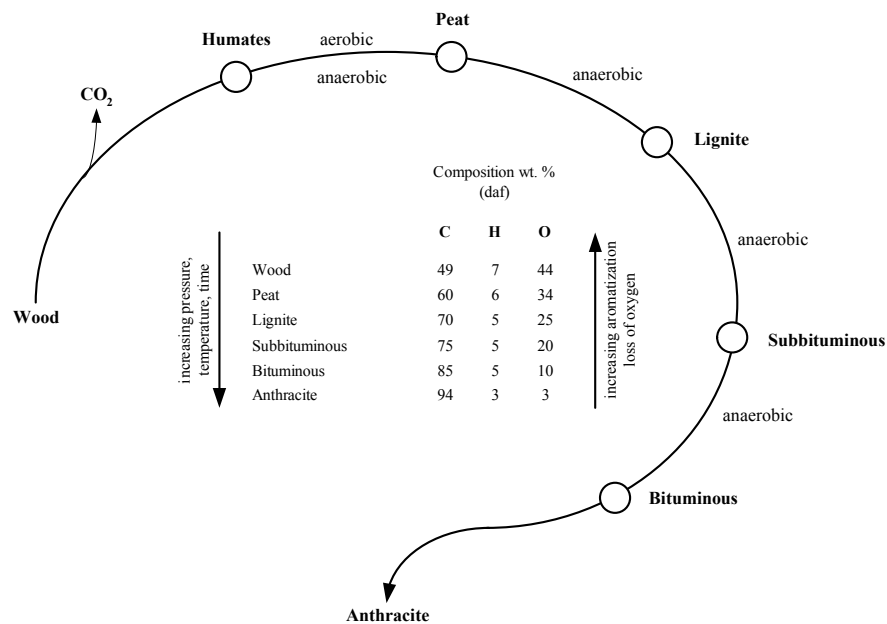


Figure 2.3: Schematic representation of the coalification process /5/

The coalification process is essentially an initial biochemical phase followed by a geochemical or metamorphic phase. The biochemical phase includes those processes that occur in the peat swamp following deposition and burial. This process is considered to be in operation until the hard brown coal stage is reached. The most intense biochemical changes occur at very shallow depths in the peat swamp. This is chiefly in the form of bacteriological activity which degrades the peat. With increased burial, bacteriological activity ceases, and is considered absent at depth greater than 10 meters. Carbon-rich components and the volatile content of the peat are little affected during the biochemical stage of the coalification. However, with increased compaction the moisture content decreases and the calorific value increases.

From the brown coal stage, the alteration of the organic material is severe and can be regarded as metamorphism. During the geochemical or metamorphic stage, the progressive changes that occur within the coal are an increase in the carbon content and a decrease in the hydrogen and oxygen content (Table 2.1) resulting in a loss of

volatiles. This, together with the continued water loss and compaction, results in the reduction of the coal volume. Products of such coalification are methane, carbon dioxide and water. The water is quickly lost and the methane to carbon dioxide ratio increases with rank. /7/

Rank	Percent C	Percent H	Percent O	Percent N	Percent S
Peat	55.0	6.0	30.0	1.0	1.3 ^a
Lignite	72.7	4.2	21.3	1.2	0.6
Subbituminous High-volatile B	77.7	5.2	15.0	1.6	0.5
High-volatile B Bituminous	80.3	5.5	11.1	1.9	1.2
High-volatile A Bituminous	84.5	5.6	7.0	1.6	1.3
Medium-volatile Bituminous	88.4	5.0	4.1	1.7	0.8
Low-volatile Bituminous	91.4	4.6	2.1	1.2	0.7
Anthracite	93.7	2.4	2.4	0.9	0.6

^a Ash and moisture content remaining weight percent

Table 2.1: Representative analyses for peat and various coals /5/

2.3 Mineral matter

The mineral content of coal is the non-combustible inorganic fraction. This is made up of minerals which are either detrital or authigenic in origin, and which are introduced in the first or second phase of coalification.

Detrital minerals are those transported into a swamp or bog by air or water. A large variety of coals can be found in coal. These are usually dominated by quartz, carbonate, iron and clay minerals, with a diverse suite of accessory minerals which may be peculiar to the local rock.

Authigenic minerals are those introduced into a peat during or after deposition, or into a coal during coalification. Common products of the mineralization are the calcium-iron minerals such as calcite, ankerite, siderite and pyrite, with silica in the form of quartz /7/.

3 Self-heating and spontaneous combustion

Self-ignition of materials means the onset of exothermic chemical reaction and a subsequent temperature rise within the a combustible material, without the action of an additional ignition source.

Generally, self-ignition is supposed to occur when the thermal equilibrium between the two counter-acting effects of heat release due to the oxidation reaction and heat loss due to the heat transfer to the ambient is disturbed. When the rate of heat production exceeds the heat loss, a temperature rise within the material will consequently take place including a further acceleration of the reaction (Figure 3.1) /8/.

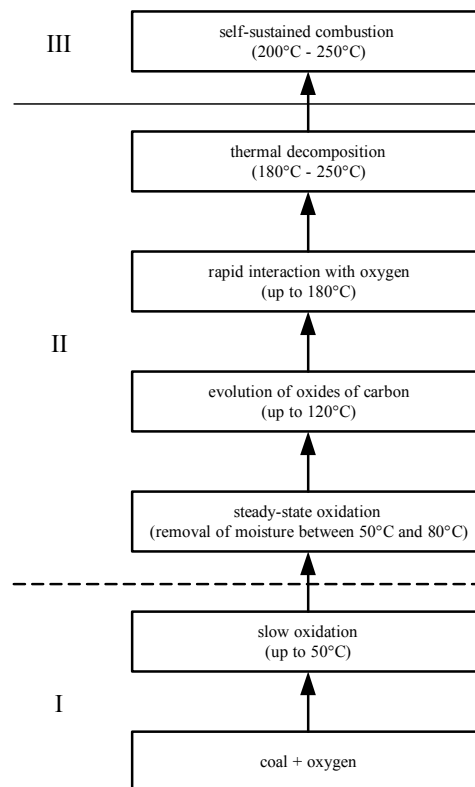


Figure 3.1: Sequential stages in the spontaneous combustion of coal /10/

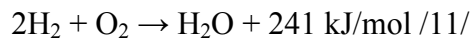
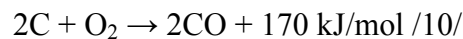
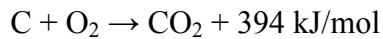
3.1 Natural oxidation of coal

In contact with the atmosphere nearly all types of coals show signs of weathering, which results in the decrease of the calorific value. A fact of great significance is that many coals are prone to spontaneous combustion. This hazard arises when the amount

of heat evolved by oxidation exceeds the amount of heat dissipated by conduction, convection or radiation /9/.

The complete oxidation of carbon to carbon dioxide is an exothermic reaction that leads to heat emissions between 21 and 42 kJ per gram of coal, taking into account that coal is not composed of pure carbon /10/.

The chemical and heat balances involved in the low-temperature oxidation of coal, spontaneous combustion and final combustion are illustrated by a few fundamental process equations, demonstrating the exothermic nature of the reactions /10/.



3.1.1 Reaction chemistry

In this section the reactions, their kinetics and activation energies will be considered. It is obvious that oxidation is not a single reaction, but rather a group of reactions sometimes competing with one another /12/.

Research has concluded that natural coal oxidation obeys an Arrhenius-type rate law /10/. The relationship between reaction chemistry, rate of reaction constant and temperature determines the Arrhenius equation /13/.

$$\ln(k) = \ln(A) - \frac{E_A}{R \cdot T}$$

k : Rate of reaction constant

A : frequency factor

E_A : activation energy

R : Gas constant

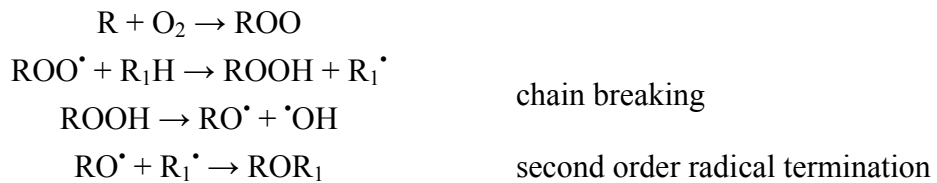
T : absolute temperature

The dependence of the activation energy on the temperature was observed. In the range between 23 and 70°C it was estimated to be 47.1 kJ/mol with first order kinetics. Higher activation energy of 82.0 kJ/mol was obtained in the range between 70 and 90°C. Below 70°C the chemisorption is dominant while above this temperature it is characteristic for

peroxide decomposition and formation of oxygen functional groups on the coal surface. Chemisorption of oxygen on the coal surface follows the reaction:



Observation of free radical concentration shows that the free radical decay obeys second order kinetics with these potential reactions:



The rate of oxidation increases by a factor of 2.2 for each 10°C rise in temperature and in proportion both to the oxygen concentration in the air and to the specific surface area of the coal. A more specific analysis yields that the rate of oxidation is slow below 40°C but accelerates by a factor of 1.8 thereafter, adding that the critical temperature above which the oxidation and self-heating process becomes self-sustaining is about 50°C for lignites and 70 – 80°C for bituminous coals. In general, the oxidation increases at ten times its usual rate as the temperature rises from 30 – 100°C.

In contrast the rate of oxidation slows as oxidation progresses. The reason for this is that the initial surface oxidation of the coal particles results in the formation of oxidised material (“oxycoal” compounds) producing in a barrier. Oxygen must penetrate through this formed barrier if further reaction proceeds. It becomes physically more difficult for oxygen to penetrate deeper into the coal, thus slowing the reaction to a minimum rate. At a constant temperature the rate of oxidation slows by one-tenth of its value per hour up to 100 hours from onset.

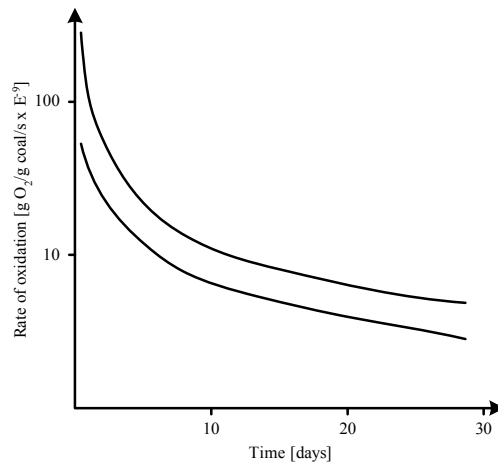


Figure 3.2: Rate of oxidation at two temperatures /10/

Three types of processes are involved in the coal oxidation. Oxygen is physically and chemically adsorbed by the coal /10/. Physically sorbed oxygen ($E_A=42$ kJ/mol /12/) diminishes in quantity with gradual temperature rise and becomes inappreciable beyond 50°C /15/. Chemical adsorption ($E_A=139$ kJ/mol /12/) leads to the formation of coal-oxygen complexes and oxygenated carbon compounds (i.e. carboxyl and carbonyl groups /16/). The formation of the carbon compounds proceeds selectively. The benzylic positions, being the most reactive, are first oxidised to produce carbonyl groups and carboxylic acids. The aliphatic groups of coal are first oxidised to form aldehydes, and then are oxidised further to form carboxylic groups and esters /17/. Finally, this results in the formation ($E_A=105$ kJ/mol /12/) and release of gaseous products, typically carbon monoxide, carbon dioxide and water vapour (Figure 3.3) /10/.

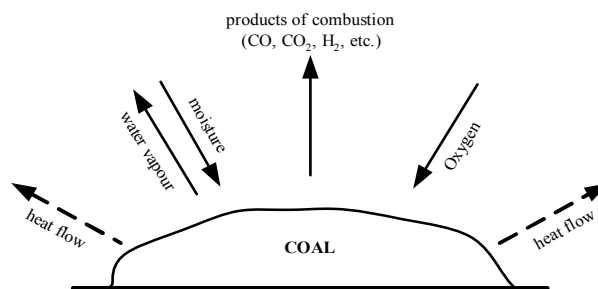
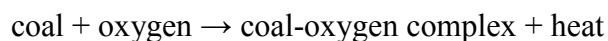


Figure 3.3: Principal chemical and physical flows during self-heating of coal /10/

At temperature below 80°C stable coal-oxygen complexes are formed /1/ as a consequence of the decomposition of peroxygen /18/ with the reaction being:



From the low temperature formation of coal-oxygen complexes and the outbreak of spontaneous combustion, the coal heats at an increasing rate. This heat is dependent on enough air being available and enough heat being retained to sustain the reaction. At temperatures above 80°C the coal-oxygen complex breaks down and releases greater amounts of heat with the reaction:



The decomposition of the chemisorption intermediates mainly generates CO while the liberation of CO₂ comes from the decomposition of the carboxyl groups. Three simultaneous reactions proceed in the thermal decomposition process. The carbonyl groups are supposed to be generated via a dehydroxylation process. The decarboxylation results in gaseous products, including CO₂, CO and H₂O, and the decarbonylation process liberates the same species. At higher temperatures of about 150°C decomposition of carboxyl and carbonyl groups occurs, leading to the production of carbon dioxide and carbon monoxide along two independent reaction pathways (Figure 3.4) /18/.

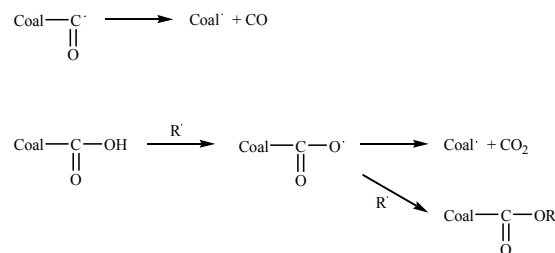


Figure 3.4: Decomposition of carboxyl and carbonyl groups /18/

In addition to the coal carbon processes, the reaction involved in the oxidation of coal hydrogen to form water are also exothermic. This is of particular importance for low-rank coals that have a high inherent hydrogen content /10/.

3.2 Factors contributing to spontaneous combustion

Aside from the natural affinity of coal to self-heating, a number of factors are significant when determining the risk of spontaneous combustion and are presented in this section.

3.2.1 Coal rank

Spontaneous combustion is a rank related phenomenon. The tendency of coal to self-heating decreases as the rank increases /1/, with lignite and sub-bituminous coals being more susceptible to self-heating than bituminous coals and anthracite /19/. As rank decreases, inherent moisture, volatile matter and oxygen and hydrogen contents increase. Medium to high volatile coals with a volatile matter content higher than 18% wt%_{daf} perform a faster oxidation rate than low volatile coals and are therefore more prone to spontaneous combustion than low volatile ones /20/. Furthermore lower rank coals often have a greater porosity than higher rank coal and therefore more surface area is available for oxidation. Low rank coals also contain long chain hydrocarbons making them less stable than for example anthracite which has a lower hydrocarbon component /1/. However, the oxidation rate for coals of the same rank may show variety within a wide range /20/.

3.2.2 Petrographic composition

The petrographic composition of a coal is determined by the nature of the original plant material from which it was formed and the environment in which it was deposited rather than the degree of coalification (i.e. rank). The homogenous microscopic constituents are called macerals on the analogy of minerals in inorganic rocks and can be distinguished in three groups. Vitrinite consists of woody material while exinite consists of spores, resins and cuticles and inertinite, the third maceral group, consists of oxidised plant material /7/.

At constant ranks, as the inertinite content of a coal increases, its self-heating propensity decreases. The general trend also indicates an increase in self-heating propensity with either increasing vitrinite or liptinite content.

Liptinite > Vitrinite >> Inertinite

Coal ranks seem to play a more significant role in self-heating than the petrographic composition of coal /1/.

3.2.3 Methane content

The emanation of methane from coal forms a sluggish atmosphere and may inhibit low temperature oxidation, exceptionally in coals with high contents of gas /20/ but methane also possesses the potential as a source of energy^o/8/. Furthermore, as the methane desorption decreases sharply with time, more of the coal surface will be exposed to oxidation. Coals with methane gas emission index less than 5 m³/t show high oxidation rates while coals containing above 8 m³ of methane/t are not oxidised /20/.

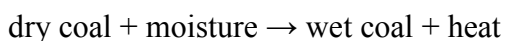
3.2.4 Minerals

Many chemicals in mineral form affect the oxidation rate to some extent, either accelerating or inhibiting it. Alkalies may have an influence of acceleration while borates and calcium chloride can act as retardants. The oxidation process is also promoted if ankerite is a constituent of the coal mineral matter. In contrast to this silica and alumina retard the oxidation /20/.

Pyrite (FeS₂) evolves heat from aerial oxidation and was believed to be the cause of the spontaneous heating of coal. The heat generation locally promotes the self-heating process of coal but the reaction products have a greater volume than the original pyrite, with the result of breaking open any coal in which they are embedded and thus exposing a greater surface of coal to the air /5/.

3.2.5 Moisture

The effect of moisture on the self-ignition is twofold: on the one hand the vaporisation of moisture consumes energy and hence the ignition process is impeded /8/. On the other hand a promotion of self-ignition by the wetting of materials prone to this has been observed /21/.



In addition to the heat of wetting moisture simply blocks the access of oxygen through the coal pores. The water vapour diffusing outwards through the pores reduces the oxygen partial pressure and hence lowers the rate of the reaction or the polar water molecules attach to the reactive sites in coal /12/.

3.2.6 Particle size and surface area

Oxidation increases with increasing fineness of coal and the rate of oxidation of coal with oxygen of air is proportional to the specific internal surface. The proportional coefficient at low temperatures is the cube root but analysis also show that both rate and extent of oxidation increase with the decrease in particle size, until a critical particle diameter is reached, below which the rate remains fairly constant /15/.

A comparison with coal particles of various sizes displayed that the smaller particles seem to have a lower alkane content than the larger particles. This can be explained as a decarboxylation reaction¹, in terms of “micropore” and “macropore” oxidation (REFFigure 3.5).

When the rate determining step of coal particle oxidation results from the diffusion of oxygen from the macropores through the bulk of the total lattice, one talks of macropore oxidation. Macropore oxidation is particle size dependent. In micropore oxidation the coal particle is “open” and oxidation is not limited but it is rather controlled by the diffusion of the surface area /12/. For very fine particles the reaction becomes Knudsen diffusion controlled for active coal or kinetically controlled for less active coal /22/.

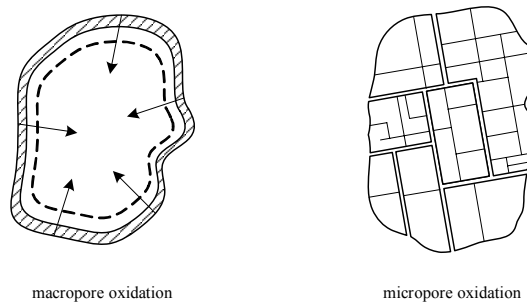


Figure 3.5: Schematic presentation of macropore and micropore oxidation models /12/

Oxidation alters the cumulative pore volume of the coal (Figure 3.6). The cumulative volume of pores greater than 10 nm (100 Ångstroms) radius is larger in the oxidised coal than in the original sample. This is reversed for smaller pores. The surface area of the oxidised coal decreases by about 40% compared to the original coal. A possible cause for this may be that the smaller pores are blocked by adsorbed oxygen and/or reaction products at the early stages of oxidation /12/.

¹ separation of carbon dioxide from free carboxylic acids

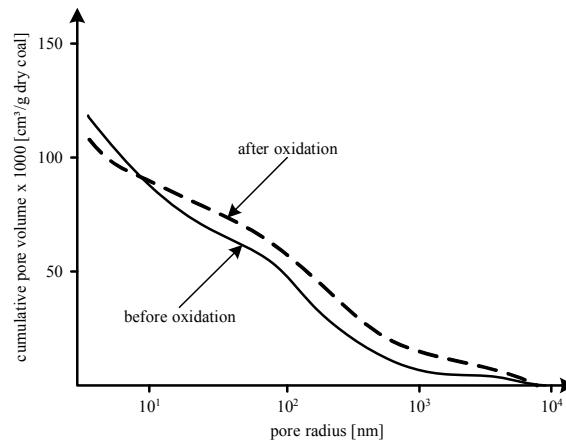


Figure 3.6: Changes in cumulative pore volume with low temperature oxidation /12/

3.2.7 Physical properties

A number of physical properties such as porosity, permeability, hardness, thermal conductivity and specific heat (c.f. energy equation) can influence the rate of oxidation and thus result in spontaneous combustion. Hardness affects the friability hence the surface area. The rate of heat transportation from the coal depends on its thermal conductivity. Coals with low thermal conductivity tend more frequently to spontaneous combustion /20/.

3.2.8 Friability

The rate of coal oxidation has a linear relation with the surface area of coal. Therefore, the more friable the coal, the greater the surface area exposed to oxidation, thus giving off more heat per unit volume of coal /20/.

3.2.9 Oxygen fraction

It need not be explained that the volume fraction of oxygen in the gas plays a key role for the rate of the reaction of oxidation. Limiting the supply of oxygen to the active surfaces of the solid matter abates the reaction considerably /8/.

3.2.10 Heat and mass transfer

The transport of the reactants and transport of heat play an important role in the spontaneous heating of coal. Heat is transported away from the sites where it is generated due to temperature gradients. The responsible mechanisms are conduction and convection. Oxygen and water participate in the heat generation processes and are transported by diffusion and convection. Convection may be caused by differences pressure at the surface (forced convection) and differences in temperature between the surface and the surrounding air (free convection) /23/.

3.2.11 Volume to surface ratio

The volume to surface ratio (V/A) is the principal geometrical quantity to assess the self-ignition behaviour of accumulations of solid bulk material, e.g. stockpiled coal. Samples of different shapes, but the same V/A ratio have the same self-ignition temperature

3.3 Emissions from self-heating and spontaneous combustion

Emissions from accelerating coal oxidation, self heating and spontaneous combustion follow clearly defined patterns. Research has suggested that patterns of gas emissions are not the same for coals from different geographical locations, in particular in relation to higher organic gases such as ethane and ethylene, acetylene, acetone and acetaldehyde. An example of the so-called “fire ladder” of gas emissions that can be used to indicate the progress in self-heating is shown in Figure 3.7. A graphical representation of the rate of emissions of various gases with increasing temperature is illustrated in Figure 3.8.

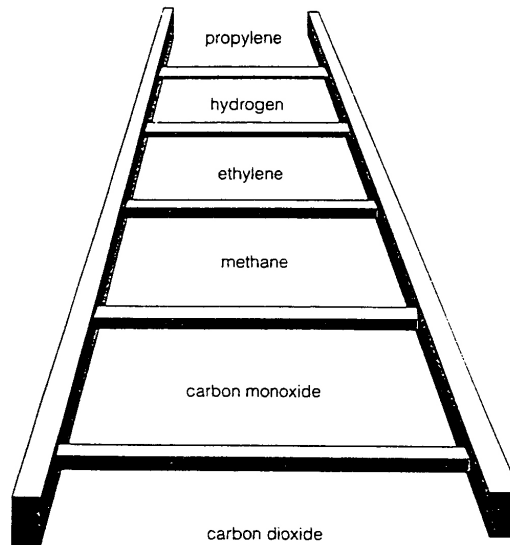


Figure 3.7: "fire ladder", showing the different products of combustion emitted with increase in fire temperature /10/

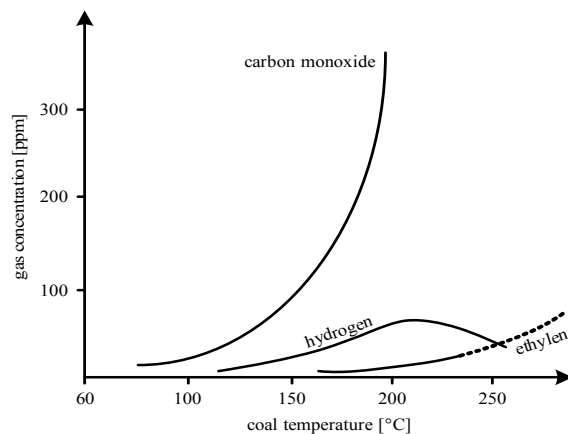


Figure 3.8: Emission concentration of typical combustion products with increasing fire temperature /10/

Accompanying, and in some cases preceding gas emissions, the various stages of self-heating can produce emissions of smells that also typify the progress of reaction.

3.4 Prediction of spontaneous combustion

It is important for coal producers and those involved at any stage in coal handling, storage and transport to know how likely a particular coal is to undergo self-heating leading to spontaneous combustion. A number of systems have been devised for

indicating the spontaneous combustion and a short survey of some methods is introduced here.

The theory of thermal explosions applies Fourier's differential equation of heat conduction to systems in which heat is produced due to one or more exothermic reactions. This theory is an analysis of the evolution with time of the temperature within the reactive system /8/.

Olpinski proposed a method to calculate the degree of spontaneous fire risk using seven different types of mining factors /20/.

The BHP Australia prediction method measures the temperature rise produced by a coal sample under controlled oxidising conditions. This increase is an indication of the susceptibility of self-heating /1/.

The relationship between oxygen/carbon ratio in coal and dry ash-free oxygen content provides reliable guidance to the potential of self-heating /10/.

The B-M Liability Index is based on the coal's proximate analysis giving a predictive equation for spontaneous combustion /1/.

The US Bureau of Mines uses an empirical relationship based on the coal dry ash-free oxygen content, the Self-Heating Temperature (SHT), as a prediction of the self-heating tendency of coal /1/.

Because the SHT prediction method does not take other factors into account (e.g. coal properties, geological and mining conditions) the US Bureau of Mines developed an expert system computer program called SPONCOM, to estimate the spontaneous combustion risk in underground coal mining /4/.

4 Conclusion

This paper “*Understanding self-ignition of coal*” serves all experts and scientists from various disciplines who are participating in this project as a brief introduction into this topic. The adequate selection of new exploration, extinction and monitoring technologies to be performed by the interdisciplinary group of experts requires a basic understanding of the causes of self-ignition. For deeper studies of this topic additional publications are compiled in chapter 8.

In a common sense, the phenomenon of self-ignition of coal means the onset of exothermic chemical reactions and a subsequent temperature rise within the combustible material, without the action of an additional ignition source. Generally, self-ignition is supposed to occur when the thermal equilibrium between the two counter-acting effects of heat release due to the oxidation reaction and heat loss due to the heat transfer to the ambient is disturbed. In case that the rate of heat production exceeds the heat loss, a temperature rise within the material will consequently take place including a further acceleration of the reaction. This positive feed-back loop ends up in self-ignition, finally.

Only the factors which are contributing to this physio-chemical process are described in this study. Beside these material-specific or internal factors, there are additional external factors which are influencing this process indirectly. These are factors such as climate, geology, geomorphology, anthropogenic influences etc. They can create an environment which might facilitate the self-ignition process.

Moreover, a further limitation of this study is that it is restricted to the combustion material coal as it is encountered on stockpiles or as a laboratory sample. In fact, the project will have to address the in-situ situation of coal seams embedded in host rocks, layered by clayey bands, fractured by cleats and joints and as a part of the hydrogeological regime.

The transfer of this widely known physio-chemical process to the in-situ coal seam situation is certainly a major task of this project. The ignition and the combustion process in-situ depends on the described internal and the - still to be studied – external factors. The combustion process under probable in-situ conditions as well as the external factors will be studied in course of the research activities in WP2400.

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7 Further publications on self-ignition

Out of a vast number of publications on self-ignition, the more recent, published since 1990, have been considered. The list of further references reflects the following structure : thermal explosion theory, experimental procedures, special problems, numerical simulations. Though this review has been prepared with care, the authors cannot exclude to possibly have missed the one or the other meaningful paper.

Among the monographs, the comprehensive book of Bowes [6] contains a summary of the fundamental works related to the thermal explosion theory published up to the appearance of this book in 1984. This includes the pioneering work done by Thomas [3] and other authors in the UK between the fifties and the seventies in applying the thermal explosion theory to cases of geometry and boundary conditions beyond the original approach of Semjonov [1] and Frank-Kamenetzki [2].

Most of the experimental and theoretical work on self-ignition has been performed for coal. Safe storage as well as safe transportation of coal was discussed e.g. in the numerous contributions of Jones and co-workers [12], [19], [22]. An essential point of interest is, that due to the different influences discussed in section 4, criticality may occur at circumstances which might have been assessed as safe using the conventional thermal explosion theory, e.g. the influence of moisture, see Gray and Wake [27].

While most of the experimental work was performed in laboratory scale, Fierro et.al. [50], [51] published data on self-ignition in stockpiled coal in large scale under the influence of wind and geometry of the stockpile, Namely, the angle of inclination of the stockpile facing the preferred wind direction is seemingly a key parameter for criticality.

Undoubtedly, advanced computer modelling of self-ignition and smouldering combustion will more and more become a common tool for assessing the risk of spontaneous fires in solid bulk materials. Numerical models for smouldering in porous media have been reviewed e.g. by Ohlemiller [52]. Again, most of the work has been dedicated to coal also in the modelling field, see Krishnaswamy et.al. [33], Hull [45], [53], Akgün and Essenhigh [54], [55] and Rosema et.al. [56]. Models differ mainly in

the number of chemical components considered which is linked to the number of transport equations treated.

Experimental work in the near future will probably be dedicated to effects influencing criticality, which cannot be treated by hot storage experiments, e.g. low-temperature oxidation or micro-biological processes promoting spontaneous heating.

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8 Appendix

Rank		Refl. Rm ₀₁	Vol. M. d. a. f. %	Carbon d. a. f. Vitrite	Bed Moisture	Cal. Value Btu/lb (kcal/kg)	Applicability of Different Rank Parameters	
German	USA							
Torf	Peat	0.2	68					
		0.3	64	ca. 60	ca. 75			
Weich-	Lignite	0.3	60			7200 (4000)		
		0.4	56		ca. 35			
Matt-	Sub-Bit.	0.4	52			9900 (5500)		
		0.5	48	ca. 71	ca. 25			
Glanz-	C	0.5	44			12600 (7000)		
		0.6	44	ca. 77	ca. 8-10			
Flamm-	B	0.7	40					
		0.8	36					
Gasflamm-	A	1.0	32					
		1.2	28	ca. 87		15500 (8650)		
Gas-	Medium Volatile Bituminous	1.4	24					
		1.6	20					
Fett-	Low Volatile Bituminous	1.8	16					
		2.0	12					
Ess-	Semi-Anthracite	2.0	12					
		3.0	8	ca. 91		15500 (8650)		
Mager-	Anthracite	3.0	8					
		4.0	4					
Meta-Anthr.	Meta-A.							

Figure 8.1: Rank parameters of coal /6/