RELATIVE SELF-HEATING TENDENCIES OF COAL, CARBONACEOUS SHALES AND COAL REFUSE¹

by

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<u>Abstract</u>. Fires in abandoned coal mines and waste banks present serious safety and environmental hazards. Often, spontaneous combustion is assumed to be the source of these fires, although some coals have an apparently low susceptibility to self-heating. It has also been noted that in abandoned mines evidence of heating is found in the roof coal and shale. Spontaneous ignition is also a factor in coal waste bank fires. Although the spontaneous combustion of coal has been studied extensively, there is very little information about the self-heating tendency of roof coals and carbonaceous shales.

This study of self-heating tendencies utilized a modified version of a differential thermal analyzer. In this study, a combustion furnace served as the reference temperature against which the sample temperature was measured. A crossing point temperature index was used to relate self-heating tendencies of various samples. Analysis of the effluent gas for CO_2 , CO, O_2 depletion and desorbed hydrocarbons was also used to evaluate the degree of self-heating and the heat generating process. Average sample size was 3 kg; particle size was between -1/4 in and +20 mesh. Furnace temperature was between 20 and 200° C. The air flow rate was 2 scfh, and each sample was heated in moist and dry air. Ten samples, including the Pittsburgh coal, roof slate, carbonaceous shales and western coals were tested. The crossing point index was compared to known self-heating behavior, reaction rates, BTU values and compositional differences.

Additional key words: Spontaneous combustion, Abandoned mine fires, Waste bank fires.

<u>Introduction</u>

The Bureau of Mines' Environmental Technology Group at the Pittsburgh Research Center is involved in an integrated program of research on the control of fires in abandoned mines and waste banks. These fires present serious safety and environmental hazards. Extinguishing such fires by conventional methods is expensive, difficult and frequently ineffective (Chaiken, 1983). Often, spontaneous combustion is the apparent cause of a fire in an abandoned mine even though the coal in question has a low susceptibility to spontaneous combustion. It

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² Ann G. Kin is Research Chemist and Robert F. Chaiken is Supervisory Research Chemist at Pittsburgh Research Center, U.S. Bureau of Mines, Pittsburgh, PA 15236. has also been noted in many fire control projects that evidence of heating is found in the roof coal, and that the roof coal and shale may serve as continuous paths for spreading the fire. Coal refuse is also known to be susceptible to combustion, often when no apparent ignition source is determined.

The self-heating tendency of roof coals and associated carbonaceous shales is a parameter in determining how fires are spread in abandoned coal mines and has a direct bearing on the application of fire control techniques. If fires start or are spread along roof coals and shales, then any method directed toward controlling a fire in the mine void or collapsed areas will not be effective (Dalverny, 1989). For instance, methods using the heat transfer capabilities of water or other agents depend on delivering the agent to a heated zone. If the heated zone exists in the roof of the mine, any procedure which delivers the agent only to the mine void is biased toward failure.

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535 https://doi.org/10.21000/JASMR90010535 Coal is defined as "a readily combustible rock containing more than 50 % by weight and 70 % by volume of carbonaceous material..... (Schopf 1966)." The non-carbonaceous matter in coal, mineral matter, is the inorganic non-combustible compounds which form ash and the sulfur compounds. According to ASTM standards (American Society for Testing and Materials, 1983), the lowest rank coals have more than 48 % fixed carbon and a heating value of less than 8300 BTU. Carbonaceous shales are sedimentary rocks which probably developed from peats containing less than 50 % organic material (Jones and Cameron 1988). Coal refuse consists of waste coal, slate, carbonaceous shales, pyritic shales, clays and other strata associated with the coal seam. The combustible content of this material averages between 2000 and 6000 BTU.

As with any fire, fires in abandoned mines or coal waste banks require three elements: fuel, oxygen and an ignition source. In coal combustion, the fuel is the carbon in the coal. In room and pillar mining as much as 30 to 50 % of the mineable coal remains in the abandoned mine. A layer of coal near the mine roof is frequently left in place for additional support during mining. It eventually fractures and collapses after mining ceases. The material in waste banks may have a combustible content of 50 % or more.

If combustion is considered the exothermic reaction of carbon and oxygen to form carbon dioxide, written as:

 $C + O_2$ ----> CO_2 + heat,

then the amount of heat liberated is -93.7 kcal/mole. However, coal is not composed of elemental carbon. On a dry, mineral matter free basis, coal contains between 60 and 90 % carbon. The rest of the coal "molecule" is composed of hydrogen, oxygen, nitrogen, and sulfur. The combustion reaction for an average bituminous coal can more accurately be written as (Chaiken 1977):

CH1.18N0.1500.35S0.005 + 1.1202 --->

 $CO_2 + 0.58H_2O + .005SO_2$.

This reaction is exothermic, producing more energy than it consumes (from 5 to 10 kcal/g of coal) depending on the rank of the coal. The oxidation of coal occurs constantly. The temperature of the coal is a function of the rate of heat generation versus the rate of heat loss. When these processes occur at the same rate, the temperature of the coal remains constant. When the rate of heat generation is greater than the rate of heat loss, the temperature of the reacting system increases. Since the rate of heat generation is an exponential function of temperature and the rate of heat loss is a linear function of temperature, as the temperature increases, the reaction rate increases faster than the heat loss rate. Ignition, or selfsustaining combustion, will occur when the rate of heat generation exceeds the rate of heat loss (Kanury 1975). Ignition is therefore, a function of the \underline{amount} of energy released by a reaction and the \underline{rate} at which it is released, as well as the rate at which energy is transferred from the reacting mass to the surroundings. The reaction rate is a function of the concentration of reactants, carbon and oxygen, the surface area, particle size, temperature and activation energy.

In spontaneous ignition, there is no external heat source; natural reactions supply sufficient energy to sustain combustion. Spontaneous combustion in coal, carbonaceous shales or coal refuse is related to the oxidation of the coal to form CO_2 and CO. The oxidation of pyrite and the adsorption of water on the coal surface also are exothermic or heat generating reactions which increase the probability of spontaneous combustion. Thermophillic bacteria may also contribute to raising the temperature of the coal. In waste banks, most of the oxygen diffusing from the surface is consumed by bacterial activity within a few feet. However, enough oxygen is available at depth to support combustion.

The normal ignition temperature for coal is between 420° and 480° C. Under adiabatic conditions (all heat generated is retained in the sample), the minimum temperature at which a coal will self-heat is 35° to 140° C (Smith & Lazarra 1987). Normal underground temperatures are usually below 35° C. To reach temperatures at which combustion is self sustaining, heat generation must be greater than heat loss. In most abandoned mines and waste piles, conditions favor the retention of heat. As noted above, there are several exothermic reactions which release energy. Heat is lost by convection or conduction. Since abandoned mines and waste piles have an essentially stagnant atmosphere, convection accounts for very little heat loss. Most heat transfer is probably by conduction to surrounding strata. Rocks tend to be good insulators and retain heat within the mine or waste bank. Self-heating as the source of wasted coal fires is the cumulative effect of heat generation and heat removal mechanisms.

The spontaneous combustion of coal has been extensively studied in the United States, Europe and the Soviet Union, for active mines, abandoned mines and for rail and water transportation methods. Studies of self-heating of coal refuse have been primarily limited to methods of reducing the occurrence of fires in active coal waste banks. There is essentially no information dealing with the self-heating tendency of roof coals or carbonaceous shales associated with coals, yet these are the materials that comprise the fuel for spontaneous heating in abandoned mines and waste banks.

The current study is intended to determine the relative combustibility of coal, coal waste, roof coal and carbonaceous shales. Specifically, it is intended to describe some characteristic thermal behavior that can be related to the probability of self-heating. Also the study attempts to relate self-heating to the physical and/or chemical characteristics of the sample, and differentiate heat contributions (+/-) from reactions, physical processes, and heat transfer.

Materials and Methods

This experiment was designed to determine the cumulative effect of heat generation and heat loss in a relatively short period of time. It used a relatively large sample at controlled temperatures and was not oxygen limited. To determine the relative self-heating tendencies of coals, roof coals and carbonaceous shales, a version of differential thermal analysis was used. In differential thermal analysis, a temperature difference is measured between a sample and a standard. In conventional differential thermal analysis, two sample compartments within a thermal block, one filled with the sample, the other filled with an inert substance (usually sand), are heated. As the temperature of the block is raised, the difference between the two materials is measured. The point at which the temperature of the sample exceeds the temperature of the standard (the crossing point temperature) is taken as a measure of the sample's tendency to self-heat. Generally, sample size is a few grams, the atmosphere is stagnant and heat loss is minimal.

This study used an 8-in. diameter furnace as the differential thermal analyzer. The furnace temperature was the standard against which the difference in the sample temperature was measured. In this study, the sample size was approximately 3000 g, crushed and screened to -1/4 in. by +20mesh. The samples were dried overnight under nitrogen at 100° C to remove surface moisture. The samples were packed into the sample holder to create an approximately homogeneous bed. Positive displacement air flow, at a rate of 2 scfh, insured an even distribution of air in the sample. Duplicate tests with wet and dry air were run to determine the effect of water adsorption. Dry air was obtained by passing compressed breathing air through a tube of Drierite³. In the moist air tests, the air was bubbled through water immediately prior to the furnace inlet.

Each sample was heated to 100° C, level 1, and held at that temperature for approximately 24 hours. The furnace temperature was then raised to 150° C for 24 hours, level 2, and then to 200° C, level 3. The rate at which the sample heated to the furnace temperature was determined and the temperature differential between the sample and the furnace was measured.

A gas sampling line was installed in the center of the furnace. A small pump was used to purge the line through a T connection, and the gas sample collected in Vacutainers by puncturing the septum in the T. Evolved gases, CO and CO₂, were determined by gas chromatographic analysis against a standard. Oxygen consumption was determined as the difference between the concentration in the furnace gas sample and that of the influent air. All solid samples were analyzed for proximate, ultimate and BTU values (Table 1). Forms of sulfur were determined for four of the shale samples.

The F Seam sample was a lower rank Wyoming coal which was known to self-heat. The coal sample, Blue Creek, and the 4 shales labeled Zone A, B, C and D were from a mine which was experiencing selfheating, apparently from the shales in the floor. The shales were channel samples taken from undercasts, with A being closest to the mine floor. The four shale layers were separated by rock. The samples Cl, C2 and Dl, D2 were samples of the C and D layers from different areas of the mine. Albright (ALB) was a refuse sample from a waste bank which was burning; no apparent cause of the fire had been determined. The Bailey sample

 3 References to trade names are given for identification only and do not imply endorsement by the Bureau of Mines.

was a near surface sample from an active waste bank. The sample of the Pittsburgh coal (PGH) had been obtained from the Bruceton Experimental Mine.

<u>Results and Discussion</u>

In this version of the crossing point method, the extent to which the sample temperature exceeds the furnace temperature is assumed to be indicative of the tendency of the sample to selfheat. Crossing point temperatures were measured at three oven temperatures and a composite index was developed.

For each sample, the furnace temperature and the sample temperature were plotted against elapsed time (Fig. 1). At 100° C, level 1, the temperature of the sample increased until it equaled or exceeded the furnace temperature. The temperature of the sample then would return to the furnace temperature. This indicated sufficient heat generation to raise the sample temperature, followed by heat loss and cooling of the sample. The same pattern was observed at 150° C, level 2. At 200° C, level 3, some samples exhibited the previously observed heating/cooling cycle. For other samples, the temperature increased quickly and did not decrease, indicating that the sample temperature was increasing toward the point of thermal runaway. Normally the temperature of the furnace was held at the designated level for 24 hours. If the sample temperature at level 3 increased by approximately 40° C in less than eight hours, the test was terminated to prevent runaway heating and the deposition of coal tars in the furnace.

The maximum temperature differential (Table 2) between the set temperature and the sample temperature was determined at temperature levels 1, 2, and 3 (100° , 150° and 200° C). In those tests which were terminated early due to sustained self heating at level 3, the maximum temperature of the sample was often lower than that of samples which were heated for the standard 24 hours. For example, the maximum temperature of Run # 30 was 281° C at the end of the 24 hour period. Run # 24 was terminated when the sample temperature reached 264° C within 9 hours. For those runs which were ended in less than 24 hours at level 3, an arbitrary 50 ° C was added to the maximum temperature differential and designated dTmax+.

These values, in Table 2, were used to determine a self-heating rank for levels 1, 2 and 3 (Table 3).

At level 1 (100° C), the maximum temperature differential was between 0 and 9° C. At level 2 (150° C), the maximum temperature differential was between 1 and 56° C, and at level 3 (200° C) it was between 10 and 81° C. The six runs with a "+" in the level 3 column were those which apparently would have reached ignition temperature if the tests had not been terminated. The samples were ranked at levels 1, 2, and 3+ according to the maximum temperature differential at each level. A composite rank from the sum of the values at levels

1, 2 and 3+ was also determined (Table 3). All ranks are given from 1 to 22 with 1 indicating the highest temperature differential, indicative of the greatest degree of self-heating, and 22 being the least.

SAMPLE NAME	SAMPLE TYPE	MOIST	ASH	VOLATILE MATTER	FIXED CARBON	TOTAL SULFUR	BTU/LB
ALBRIGHT BAILEY F SEAM BLUE CREEK PITTSBURGH ZONE A ZONE B ZONE C	Refuse Refuse Coal Coal Coal Shale Shale Shale	4.79 2.18 9.14 0.77 1.40 0.31 0.64 0.94	57.50 79.76 4.04 36.16 4.76 79.96 59.98 75.28	12.17 12.03 40.86 19.16 37.36 17.66 15.20 10.51	25.53 6.03 45.96 43.91 56.48 5.07 27.18 13.27	7.34 1.56 0.51 0.70 1.01 0.29 0.46 2.82	4606 1573 11918 9422 14205 1573 5925 2725
ZONE D	Shale	1.04	61.58	10.51	23.12	2.82	2725 4841

Table 1: SAMPLE ANALYSIS, As-received, wt pct





Figure 1. Crossing Point Temperature, Modified

Table 2: MAXIMUM TEMPERATURE DIFFERENTIAL, Deg C

¹D: Dry air M: moist air

Table 3: SELF-HEATING RANK

RUN #	SAMPLE	AIR D/M	RANK Level 1	RANK Level 2	RANK Level 3+	RANK Total +	
23 24 18 31 32 20 30 29 27 28 14 25 16 22 12 15 17 19 26	F SEAM F SEAM ZONE C1 ZONE D2 ZONE D2 ZONE D1 BLUE CREEK PGH PGH ZONE A ALB ZONE C1 ZONE C2 BAILEY ZONE B ZONE B ZONE D1 ALB	D M M D M D M D D D D D D M D D M D M D	1 2 4 7 10 12 3 20 13 21 8 6 14 19 16 18 5 15	1 2 4 3 6 9 3 5 7 7 10 8 6 10 12 12 13 13 13 13	1+ 3+ 2+ 4+ 5+ 6+ 7 8 10 9 11 12 13 14 15 16 18 17 20	1 2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18	
11 13 21	BAILEY ZONE A ZONE C2	D D M	22 17 9	13 14 15	19 21 22	20 21 22	

The F Seam sample and the shales from Zone C and Zone D indicated the greatest overall selfheating. The coal samples also exhibited some self-heating. The Albright and Bailey refuse samples and the shales from Zones A and B exhibited the least self-heating.

The ability to self-heat is apparently not directly related to the amount of carbonaceous material in the sample, since two of the samples which exhibited the greatest degree of self-heating were shales with over 60 % ash. Similarly, heating value (BTU) is not a direct indicator of self-heating.

If self-heating of carbonaceous materials is related to heat generation and heat loss from a

single process, it could be expected that increased temperature would simply increase the reaction rate of that process. Under these conditions, the material that was most prone to self-heat at 100° C would be the material most prone to self-heat at 150° C and at 200° C. The relative ranking of the tendency to self-heat should remain the same at all three levels. In this study, the relative rankings at the three levels were different for most of the samples. This may indicate that different reactions with different activation energies are involved in the self-heating process.

The samples were dried under nitrogen at 100° C to remove normal bed moisture, since initial trials indicated that no heating of the sample would occur until the bed moisture was removed. Then, if the

SAMPLE	TOTAL Rank D	TOTAL RANK M	M-D Level 1	M-D Level 2	M-D Level 3+	M-D Total
ALB BAILEY F SEAM BLUE CREEK PGH ZONE A ZONE A ZONE B ZONE C1 ZONE C2 ZONE D1 ZONE D2	5 10 1 3 4 11 8 6 7 9 2	10 8 1 5 6 7 9 2 11 4 3	-1 ¹ 3 -1 -4 2 2 2 1 2 1 1 1	-5 1 -14 -10 3 9 0 12 -6 5 11	-26 6 -15 -11 0 38 -2 81 -22 74 6	-32 10 -30 -25 5 49 0 94 -26 80 18

Table 4: COMPARISON OF MAXIMUM TEMPERATURE DIFFERENTIAL, DEG C, IN MOIST AND DRY AIR

¹Negative values indicate higher temperature differentials in dry air.

RUN #	SAMPLE DRY	/MOIST	-02 ¹ LEVEL 1	-02 LEVEL 2	-02 LEVEL 3	-0 ₂ t = 0	
25 26 11 12 23 24 30 29 28 27 13 14 15 17 16 18 22 21 19 20 32	ALB ALB BAILEY BAILEY F SEAM F SEAM BLUE CREEK BLUE CREEK PGH PGH ZONE A ZONE A ZONE A ZONE B ZONE C1 ZONE C1 ZONE C1 ZONE C2 ZONE C2 ZONE C2 ZONE D1 ZONE D2 ZONE D2	D M D M D M D M D M D M D M D M D M D M	3.94 2.76 1.21 1.72 1.09 2.30 1.53 1.93 2.94 2.19 4.60 4.09 2.13 1.49 0.94 1.35 0.24 1.49 0.66 1.68 1.40	8.62 4.21 2.75 5.00 9.09 4.12 2.64 3.10 7.50 4.53 4.73 1.82 1.78 3.50 4.71 1.48 2.39 1.35 3.50 6.74	13.78 11.80 5.38 11.34 8.61 9.96 11.70 11.41 14.75 18.61 6.56 12.80 7.70 8.16 12.03 13.50 12.03 13.50 12.09 4.98 3.48 14.50 12.49	12.76 13.46 4.70 1.44 18.46 19.50 3.50 3.36 2.30 6.31 1.80 3.36 1.80 0.62 0.88 5.04 12.83 4.37 2.14 1.70 7.67	
•-							

Table 5: OXYGEN CONSUMPTION, Pct

¹Difference between normal air and measured concentration in furnace.

exothermic adsorption of water is a mechanism for initiating self-heating, it would be expected that the maximum temperature differential, particularly at level 1, would be greater in moist air than in dry air. In this study, the effect of moisture at level 1 was minimal. It was more pronounced at levels 2 and 3, but could not be correlated to self-heating behavior (Table 4).

When the total rank for all dry samples is compared, the F seam sample and the Zone D2 shale are the most reactive under both conditions. The Bailey refuse and the Zone B shale had approximately the same reactivity under both conditions. For all other samples the relative reactivity was substantially different under moist and dry conditions.

If self-heating is directly related to the oxidation of the carbonaceous material, the amount of oxygen consumed and the amount of CO_2 produced should be proportional to the heat gain. Oxygen consumption (Table 5) does increase with increased temperature for each sample. However, it is not proportional to the temperature increase, and the presence of moisture in the air stream has no consistent effect on the change in the concentration of oxygen.

Oxygen consumption increased at sample temperatures equal to or greater than 100 deg C (Fig. 2), indicating an increase in oxidation rate at these temperatures. For approximately half of the samples, oxygen was apparently adsorbed on the

sample when it was initially placed in the furnace(t = 0 - Table 5, Fig. 3). However, if physical adsorption is the only process involved, the consumption of oxygen should decrease at higher

temperatures, unless the removal of some carbon moieties is producing more sites for oxygen adsorption.



Figure 3. Oxygen Adsorbtion and Consumption versus Temperature

The production of CO_2 follows the same pattern as the consumption of oxygen (Table 6, Figs. 4, 5). If the initial adsorption of oxygen is low, the initial production of CO_2 is also low; both concentrations increase with temperature. If more oxygen is consumed initially, more CO_2 is produced initially. However, it should be noted that the amount of CO_2 produced is not equivalent to the amount of CO_2 produced. At temperatures above 100° C, the concentration of CO increases as the CO_2 concentration increases. At temperatures of 100° C or less, CO is apparently not a reaction product (Table 7).

RUN #	SAMPLE I	DRY/MOIST	CO2 LEVEL 1	CO2 LEVEL 2	CO2 LEVEL 3
25	ALB	0	0.04	0.44	1.77
26	ALB	м	0.06	0.16	1.42
11	BAILEY	D	0.09	0.16	0.90
12	BAILEY	М	0.03	0.19	2.22
23	F SEAM	D	0.33	1.95	2.77
24	F SEAM	М	0.28	1.38	1.81
30	BLUE CREI	EK D	0.04	0.35	4.13
29	BLUE CREI	EK M	0.03	0.26	2.47
28	PGH	D	0.49	2.26	1.44
27	PGH	М	0.08	0.35	4.34
13	ZONE A	D	0.10	0.07	0.81
14	ZONE A	М	0.06	0.48	2.25
15	ZONE 8	D	0.02	0.11	1.11
17	ZONE B	M	0.04	0.19	1.31
16	ZONE C1	D	0.02	0.26	1.96
18	ZONE C1	M	0.05	0.36	3.00
22	ZONE C2	D	0.02	0.18	1.94
21	ZONE C2	М	0.02	0.06	1.26
19	ZONE D1	D	0.02	0.13	0.72
20	ZONE D1	M	0.04	0.25	2.24
32	ZONE D2	D	0.04	0.72	2.19
31	ZONE D2	М	0.03	0.30	2.50

Table 6: CO2 PRODUCTION, Volume Pct



Figure 4. CO₂ and CO Generation, Initial Low Oxygen Adsorbtion

<u>Summary</u>

The results of this study of the self-heating characteristics of coal and other carbonaceous materials produced a large volume of data which has not yet been sufficiently analyzed to develop direct predictions of self-heating behavior. This study has, however, demonstrated that measured experimental parameters can be related to potential self-heating. It has also shown that high ash samples, such as coal wastes and carbonaceous shales are capable of generating sufficient heat to initiate fires in abandoned mines and waste banks.

The results have shown that self-heating is not a simple process, and it is apparently more complex



Oxygen Adsorbtion

than differences in heat generation and heat loss rates. In any sample, more than one heat generating reaction may occur, with one reaction providing the activation energy for subsequent reactions.

It should be noted that the system used did not allow bacterial activity and did not directly measure the possible effect of the oxidation of pyrite. Both of these factors may be significant in the generation of heat under normal conditions.

RUN	#	SAMPLE	DRY/MOIST	CO LEVEL 1	CO Level 2	CO LEVEL 3
	25	ALB	D	0.00	0.05	0.76
	26	ALB	м	0.00	0.04	0.96
	11	BAILEY	D	0.00	0.06	0.28
	12	BAILEY	М	0.00	0.05	0.40
	23	F SEAM	D	0.00	NA1	0.89
	24	F SEAM	М	0.01	0.51	0.45
	30	BLUE CRE	EK D	0.00	0.19	1.04
	29	BLUE CRE	EK M	0.00	0.00	0.94
	28	PGH	D	0.00	0.04	0.59
	27	PGH	М	0.00	0.06	1.86
	13	ZONE A	D	0.00	0.00	0.21
	14	ZONE A	М	0.00	0.05	1.46
	15	ZONE B	D	0.00	0.01	0.42
	17	ZONE B	М	0.00	0.00	0.41
	16	ZONE Cl	D	0.00	0.03	1.00
	18	ZONE C1	М	0.00	0.09	1.95
	22	ZONE C2	D	0.00	0.04	0.87
	21	ZONE C2	М	0.00	0.00	0.14
	19	ZONE D1	D	0.00	0.00	0.26
	20	ZONE D1	М	0.00	0.04	0.94
	32	ZONE D2	D	0.00	0.23	0.95
	31	ZONE D2	М	0.00	0.08	1.00

Table 7: CO CONCENTRATION, Volume Pct

¹NA: Not Available

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