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The influence of oxygen concentration on the composition of gaseous products occurring during the self-heating of coal and wood sawdust

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A B S T R A C T

This article deals with an assessment of the influence of oxygen concentration on the composition and amount of combustion products generated in the course of heating coal particles and wood sawdust at 150 °C. This was done both with normal air and at 15% oxygen in the air in an isothermal furnace. The generated gases were analyzed by a Fourier Transform infrared spectrometer. Results show that under both conditions, the same substances are formed: water, carbon dioxide, carbon monoxide and aliphatic hydrocarbons. However, the quantities changed. At 21% oxygen, the concentrations of carbon monoxide and methane were higher than at 15% oxygen both in coal and wood. The oxygen concentration was also found to affect the rates of release of CO and CO₂. The rate of release of CO was higher at 21% oxygen, but that of CO₂ was higher at 15%, indicating two different mechanisms. In all cases, the concentrations of these gases were higher for coal than for wood. The results have implications for the specification of safe conditions of storage of coal and wood substances and the selection of safety measures.

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1. Introduction

Fires caused by self-heating of stored bulk solids are difficult to recognize and thus can pose a serious safety problem. Therefore, close attention should be paid to self-heating and spontaneous combustion. By understanding these problems, adequate safety precautions can be taken and the occurrence of spontaneous combustion of stored materials can be minimized.

Many materials tend to spontaneous combustion. Coal is one of many materials that can oxidize at normal temperature. Vegetable materials, e.g. wood, wood sawdust, pulp chips, hay, straw, and fodder, also demonstrate this tendency. In the Czech Republic, 521 cases of spontaneous combustion were recorded in the last five years (2008–2012): of these 163 were incidents involving coal and 119 farming products (Statistical Yearbooks[®] 2010).

Spontaneous combustion is defined as ignition caused by the increase of temperature without a separate ignition source (ČSN EN ISO 13943, 2011). The tendency of materials to spontaneous combustion is influenced by many factors. The most important factor is the process of oxidation, when the surface of a substance reacts exothermally with oxygen in the air even at normal temperature. The heat released by the reaction causes the temperature to rise within the bulk and when the amount of heat gained is higher than that lost to the surroundings, then spontaneous combustion occurs.

Self-ignition is not only a simple fire hazard. During self-heating, gases dangerous to lives, health, and environment can be released. The composition of these gases depends on material properties and storage conditions. The main gaseous products of self-heating of coal and wood sawdust are water, carbon dioxide, carbon monoxide and methane. Concentrations of these gases and their relative ratios are

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used in the case of mining operations and stored coal or coal spoil to determine temperature and self-heating. The ratio of carbon monoxide to carbon dioxide is most commonly used.

It is known that biomass decomposes chemically and biologically. Self-heating occurs during this process and toxic gases including CO may accumulate in storage of various biological materials including bulk foods such as grains and peas (Reuss and Pratt, 2000). Svedberg et al. (2004) deal in their study with occupational and household health risks connected with biomass storage and subsequent occurrences of high levels of hexanal and carbon monoxide. The oxidation of biomass in closed spaces is accompanied by stench and can cause irritation to the eyes and respiratory system of people working in this environment or its vicinity.

Particulate wood and particulate wood products are, like coal, stored and transported in large amounts. Moreover, there is increasing interest in partially or totally replacing coal with biomass (commonly wood derived) in electricity power stations, which may involve large quantities and long journeys. For example 10,000 t bulk carriers transport wood pellets from North America to Europe, and there are storage times of up to 10 weeks in silos of 3500 t (Guo et al., 2013).

It would thus be of interest to see if monitoring gases can provide a similar warning of self-ignition in wood product storage. A study has therefore been made to compare the amounts and rates of gases produced under similar conditions with coal and wood sawdust. As well as the effect of temperature, the influence of oxygen concentration was also tested. Where oxidation is taking place, the concentration of oxygen in the air spaces between particles may be expected to go down. In addition, partial inerting with nitrogen is a possible preventative measure (Lohrer et al., 2005; Koppejan, 2011). Hence tests were carried out with concentrations of 21 and 15% oxygen.

2. Gaseous products resulting from coal and biomass self-heating

Wang et al. (2003) studied the low-temperature oxidation of coal very thoroughly. Hydrocarbons, carbon monoxide, carbon dioxide, and water were found to be the most important gaseous products resulting from the oxidization of coal in mines. Nitrogen and hydrogen can be released in lower amounts.

Apart from the products of oxidation, other gaseous substances (in particular methane), can be released from coal by desorption or pyrolysis.

Lu et al. (2004) dealt with the quantitative and qualitative analyses of the gaseous products of low-temperature coal oxidization at various temperatures, and concluded that the presence of carbon monoxide and ethylene can be used to indicate the occurrence of a spontaneous combustion process. According to these authors, the low-temperature oxidization of coal can be divided into four phases that were identified on the basis of the changes of carbon monoxide and ethylene concentrations. Carbon monoxide and ethylene are never present in the first phase (<50 °C) products. The second phase (50–65 °C) is characterized by the presence of carbon monoxide, while ethylene is not present among the products. Both gases can be identified during the third phase (65–130 °C). The concentrations of these gases rise slowly during the second and third phases. The last phase (above 130–140 °C) is characterized by rapid growth of carbon monoxide and ethylene concentrations. If this phase continues, the concentrations of both constituents keep growing to the flammable level.

Monitoring of the changes of the concentrations of gases therefore provides one means of determining the spontaneous combustion process of coal stored on dumps and banks. These gases can be called index or indicator gases for self-ignition.

The concentration of carbon monoxide is taken as a generally recognized criterion of self-heating. Monitoring of this parameter is very important with regard to safety because of toxicity and explosivity of carbon monoxide. Apart from the indicator gases, the ratios of some gases, e.g. carbon monoxide to carbon dioxide, can be used as indicators of the extent of self-heating. The temperature of coal mass at inaccessible places in mines can be inferred by means of these criteria (Veznikova and Adamus, 2007).

Most studies deal with the problem of spontaneous combustion and the resulting products under normal atmospheric conditions (20.9 volumetric % oxygen in air). However Veznikova (1993), Davidi et al. (1995), Lohrer et al. (2005), Malow and Krause (2008) and Yuan and Smith (2013) also looked at coal with lower oxygen concentrations. Nitrogen is sometimes used in storage and processing of solids to reduce the oxygen concentration. Even where this is not done, lower oxygen concentrations can be expected inside the bulk, since oxygen is absorbed by the coal surface very rapidly, while its supply from the heap surface to the stored material can be a very slow diffusion process.

Even at reduced oxygen concentration, gases are released from the coal at increased temperatures. Their ratios can differ from that of gases occurring under normal conditions.

Veznikova (1993) looked at the possible use of nitrogen to control smouldering heaps or fires within bulk coal, and found that the content of oxygen in air influences the formation of carbon oxides, ethylene, propane, normal butane, and propylene. The amount of unsaturated hydrocarbons is much lower when heated in nitrogen rather than their amount when heated in air. Ethylene, propane, iso-butane, normal butane and propylene are released in nitrogen at temperatures higher by 20–40 °C than in air. Propylene was not detected in coal samples heated to 200 °C in the absence of oxygen. The amount of ethylene during heating in nitrogen was much lower than when the coal was heated in air. It was further found that the amounts of methane, ethane, and propane do not depend on the amount of oxygen in air.

Davidi et al. (1995) studied the release of hydrocarbons in air and in an inert atmosphere. According to these authors, the emission of saturated hydrocarbons occurs in two ways. The fast emission process occurs in the presence of oxygen, while the process is slow when oxygen is not present. There is no emission of unsaturated hydrocarbons such as ethylene in an inert atmosphere (The conversion of ethane to ethylene is an oxidation.).

Lohrer et al. (2005) found that self-ignition could occur in German lignite coal even at low oxygen concentrations and studied the influence of water on the self-heating of coal.

The change of carbon monoxide and carbon dioxide when heating coal in the temperature range from 50 to 110 °C at 3, 5, 10, 15, and 21% of oxygen in air was established by Yuan and Smith (2013). The concentration of carbon oxides rises slowly at 3–15% of oxygen in air, while it rises rapidly at 21%. The maximum concentration of carbon monoxide was measured at 21% of oxygen. The results of this work further show that the concentration of oxygen has no influence on the carbon monoxide to carbon dioxide ratio and that the emissions of carbon oxides depend on temperature. The carbon oxides emissions rise with rising temperature.

According to Oren et al. (1987), the gaseous products of thermal degradation of wood are first released at the temperatures from 150 to 200 °C, but in very small amounts only. Hydrogen, methane, carbon monoxide and dioxide, aldehydes, ketones,

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