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**LABORATORY STUDIES OF  
SPONTANEOUS COMBUSTION OF  
A VICTORIAN BROWN COAL**

by

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## SUMMARY

Spontaneous combustion of coal has been a serious problem for coal producers and users for many years, particularly during storage and transportation. Many experimental techniques and models have been developed and applied to study and describe the self-heating and spontaneous combustion phenomenon of coal. However, the inhomogeneity of coal, the complexity of the system, and the variability of experimental techniques used, caused various inconsistent, confusing, and sometimes conflicting results in the literature.

The primary objectives of the current study is to use different experimental techniques to study the spontaneous combustion behaviour of the same material. In such a way, the influence of system conditions involved in the different techniques can be understood with confidence, thus providing an improved understanding of the spontaneous combustion.

Three experimental techniques are used to examine the self-heating behaviour of a Victorian brown coal, which is well known to exhibit a high propensity towards spontaneous combustion. The experimental techniques used include isothermal reactors, adiabatic reactors, and wire-mesh reactors with both steady-state and unsteady-state methods. In every reactor, the critical ambient temperature of the coal, above which spontaneous combustion occurs, is measured and used to indicate its tendency towards spontaneous combustion. The higher the critical ambient temperature, the lower the risk of spontaneous combustion. Emphasis of the current study is given to not only the influence of coal properties and ambient conditions, but also the experimental techniques on the self-heating behaviour of the coal. Low-temperature oxidation kinetics of samples tested in each technique are estimated using an energy balance approach. The kinetic constants obtained are then used to estimate the coal reactivities and the critical layer thickness, above which a coal deposit is capable of undergoing spontaneous combustion.

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The results obtained from the isothermal reactor experiments show that there is an optimum air flow rate with which the coal self-heating could be easily induced. The effect of air flow rate on the critical ambient temperature is also investigated in an adiabatic reactor. It is revealed that within the experimental range, the air flow rate does not affect the critical ambient temperature but the time-to-ignition. The higher the air flow rate, the shorter the time-to-ignition. It is also observed that a high ambient temperature and a humid environment help accelerate the coal spontaneous combustion. Coal particle size significantly affects the coal self-heating. The smaller the particle size, the lower the critical ambient temperature, and thus the higher the risk of spontaneous combustion. The effect of coal drying methods on the coal self-heating behaviour, and the effects of reactor size, reactor specific surface area, and coal packing density on the critical ambient temperature are also investigated in this study.

Furthermore, the results obtained from different techniques show similar trends in which the critical ambient temperature varies with coal properties, ambient conditions, packing density, and reactor size. However, substantial variations are observed in the critical ambient temperature, the time-to-ignition, and the critical layer thickness predicted from different techniques. This indicates that the experimental technique used significantly affects the coal self-heating behaviour. Accordingly, precautions must be taken when comparing the experimental results obtained from different techniques.

This thesis also aims at examining the effect of inherent inorganic matter and additives on coal spontaneous combustion. Fourteen samples are prepared, namely, the raw coal, water-washed coal, acid-washed coal, and acid-washed coal doped with eleven additives. Each of the samples is then tested in an isothermal reactor to obtain its critical ambient temperature. The relative effectiveness of the additives is determined by comparing their critical ambient temperatures with that of the acid-washed coal. Potassium chloride, Montane powder, and sodium chloride are found to be the most effective inhibitors, followed by magnesium acetate, and calcium chloride. The presence of sodium nitrate and ammonium chloride in the coal

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samples does not show any significant influence on the spontaneous combustion. However, calcium carbonate, sodium acetate, potassium acetate, and pyrite promote the spontaneous combustion. The effect of additive loading is also investigated for an inhibition agent (potassium chloride) and a promotion agent (sodium acetate). It is revealed that the effectiveness of these promotion and inhibition agents is enhanced with an increase in the additive loading. Low-temperature oxidation kinetics are also estimated by an energy balance approach and compared with the self-heating potential of these samples.

The inhibition and promotion agents of coal spontaneous combustion together with the effect of anions and cations on the capability of additives to either inhibit or promote the spontaneous combustion are further studied using the wire-mesh reactor technique. Acid-washed coal, and acid-washed coal doped with fourteen additives are prepared. Each of the samples is then tested in the wire-mesh reactor to obtain its critical ambient temperature. The presence of  $\text{Na}^+$ ,  $\text{K}^+$ , and  $\text{Cu}^{2+}$  in their acetic and carbonate salts, promote spontaneous combustion, while sodium chloride and hydroxide inhibit spontaneous combustion. Magnesium acetate and carbonate inhibit spontaneous combustion.  $\text{Ca}^{2+}$  in different salts behaves differently. In its chloride and acetic salts,  $\text{Ca}^{2+}$  inhibits spontaneous combustion, while in the carbonate and hydroxide forms, it promotes spontaneous combustion. From the results, it can be concluded that the capability of the additives to affect spontaneous combustion depends on both cations and anions in the additives. The effect of additive loading is also investigated for an inhibition agent (calcium acetate) and a promotion agent (sodium acetate). The effectiveness of sodium acetate to promote spontaneous combustion is enhanced with an increase in the additive loading, while that of calcium acetate remains unchanged with varying loading. Low-temperature oxidation kinetics are also estimated by an unsteady-state energy balance approach and compared with the self-heating potential of these samples.

Whether the capability of the additives to either inhibit or promote the coal spontaneous combustion is physical or chemical in nature is also examined in this study by employing the ion exchange procedure in addition to the bulk-loading

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procedure. The bulk loading of additives mixes the additives and coal samples both physically and chemically, while the ion exchange mixes them only chemically. The self-heating behaviour of ion exchanged samples are compared with those of the bulk loading samples.  $\text{Cu}(\text{Ac})_2$  and  $\text{KAc}$  in the ion-exchanged samples show stronger promotion effects on spontaneous combustion than their respective bulk-loading samples, indicating that their effects on spontaneous combustion are mainly chemical in nature. On the other hand, the inhibition effect of  $\text{Ca}(\text{Ac})_2$  was mainly physical in nature, while  $\text{NaAc}$ ,  $\text{Mg}(\text{Ac})_2$ ,  $\text{CaCl}_2$ , and  $\text{NaCl}$  did not show a clear evidence if their effects were chemical or physical. Therefore, whether the capability of an additive to affect the spontaneous combustion is chemical or physical, is determined by the type of the additive used. SEM analysis is also employed to examine the surface structures and cation loadings of samples, bulk-loaded and ion-exchanged with  $\text{Cu}(\text{Ac})_2$ . It is observed that the pore volume of ion-exchanged sample is larger than that of bulk-loading sample. Furthermore, the amount of copper inclusion in the ion-exchanged coal particle is higher and more uniformly distributed in the coal matrix than that in the bulk-loading coal particle.

This study highlights that the coal self-heating behaviour is dependent not only on coal properties and ambient conditions, but also on the system conditions involved in the experimental techniques used. Precautions must be taken when comparing the results obtained from different techniques so that misleading information can be avoided. From the study of the effect of inherent inorganic matter and additives on coal spontaneous combustion, it is proved that the inherent inorganic matter catalyses the self-heating processes, while different additives added in the coal sample play various roles in the coal self-heating behaviour. These additives could either inhibit or promote spontaneous combustion, depending on the cations and anions in the additives, the additive loading, and the additive application procedure.

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