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Self-heating behavior and ignition of shale rock

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ABSTRACT

The combustion of shale, a porous sedimentary rock, has been reported at times in outcrop deposits and mining piles. However, the initiating event of most of these fires is unknown. It could be that, under the right conditions, shale rock undergoes spontaneous exothermic reactions in the presence of oxygen. This work studies experimentally and for the first time the self-heating behavior of shale rock. Because shale has high inert content, novel diagnostics such as mass loss measurements and visual observation of charring are introduced to detect self-heating ignition in respect to other self-heating materials with lower inter content. Using field samples collected from the outcrop at Kimmeridge Bay (UK) and the Frank-Kamenetskii theory of ignition, we determine the effective kinetic parameters for two particle-size distributions of shale. These parameters are then used to upscale the results to geological deposits and mining piles of different thicknesses. We show that for fine particles, with diameter below 2 mm, spontaneous ignition is possible for deposits of thickness between 10.7 m and 607 m at ambient temperatures between −20 °C and 44 °C. For the same ambient temperature range, the critical thickness is in excess of 30 km for deposits made of coarse particles with diameter below 17 mm. Our results indicate that shale rock is reactive, with reactivity highly dependent on particle diameter, and that self-ignition is possible for small particles in outcrops, piles or geological deposits accidentally exposed to oxygen.

1. Introduction

Unconventional oil, as opposed to conventional oil extracted from reservoirs where petroleum can flow naturally, is trapped inside tight porous media that requires enhanced recovering processes like hydraulic fracturing to release oil and gas [1]. Unconventional oil may be trapped in rocks, sands or coal; the most common examples being oil sands, coalbed methane, shale gas and shale oil [1]. Shale oil and shale gas both originate from the same source rock, shale, as shown in Fig. 1. Shale is a general term used to describe a large array of clay rich sedimentary rocks. It is fine grained and is estimated to represent 50% of all the sedimentary rocks deposited on Earth [2]. The thickness of shale rock deposits varies widely with location around the world, but it ranges from 1 m to 600 m [3].

Sedimentary rocks containing significant amount of organic matter are reactive porous media. This includes coal, oil sand and shale. Reactive porous media are materials where small free spaces (pores) are embedded in the solid together with a presence of a carbon-rich component [2], as shown in the sketch on the lower right of Fig. 1. This allows the rock to be permeable to a variety of fluids such as air, water or oil, and greatly increases its surface area making the organic particles reactive because it allows oxidation to take place if O_2 is supplied [2]. Such reactive porous rocks might undergo self-heating. Self-heating is the tendency of certain materials to undergo spontaneous exothermic reactions in oxidative atmospheres at low temperatures [4]. This process starts by slow oxidation at ambient temperature, but the reaction alone is insufficient to raise the material temperature. The temperature rise is determined by the balance between the rate of heat generation and the rate of heat losses [5]. Fire initiated by self-heating ignition is a well-known problem for many types of porous reactive media [6]. Of the reactive porous sedimentary rocks shown in Fig. 1, extensive studies on self-heating ignition behavior have been conducted for coal, both experimentally and computationally [6–13]. Some work is present in the literature on the thermal degradation of shale and kerogens (in environments without oxygen) [14,15]. However, very little work has been done in understanding the behavior of shale rock exposed to an oxidizing environment which might undergo self-heating. Early work was carried out on shale rock ignition 1982, when the US Mining bureau reported initial measurements of the self-heating of shale dust [16]. The report acknowledges that self-heating of shale rock is of importance, and states that in-depth investigation is needed. No studies have been reported in literature since.

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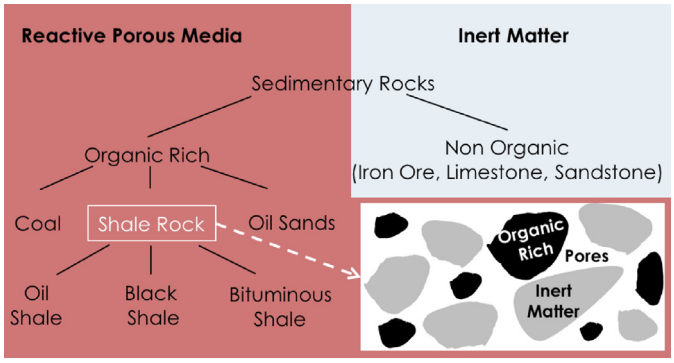

Fig. 1. Sedimentary rocks arranged according to their reactivity and geology. A sample sedimentary rock is sketched on the lower right showing the presence of inert matter, organic rich material, and pore space within the porous rock.

There are two systems of interest when studying shale self-heating, because of the presence of oxygen: The first is piles of shale accumulated on the surface during excavations, also known as heaps. These are very common from coal mining, both historically and currently. The second is geolocical formations, especially outcrops.

The ignition of heap is common. For example, the landfill in Texas in 2000 [17]. The excavated shale rock was piled loosely to one side of the landfill, and the hot summer brought the environmental critical conditions for self-heating and igntion, causing the shale to burn for more than 12 months [17]. Another common shale fire is witnessed in spoil heaps, or bings as they are known in Scotland [18]. Spoil heaps were formed in the period when coal mining boomed in the UK, from the late 18th century to the mid 20th century. These heaps are piles consisting of shales, siltstones and coal fines that were separated from usable coal as well as rocks that were removed during mining operations, and there are as many as 560 of such heaps in Scotland alone [18]. Because of their porous nature, and high carbon content, these heaps are susceptible to self-heating ignition. Heap shale fires have been witnessed for years, even recenlty, and in 2008 the Bogside fire in a 34 m tall heap was documented through an experimental campaign [18].

Combustion of shale outcrop formations has been observed in the past, with the most recent case being the Windfall Mountain in Alaska in 2012 [19]. A geological formation of shale ignited and burned for more than 24 months. The site was analyzed by the US National Park Service to determine the cause of the fire, and initial conclusions point towards self-heating as the most likely ignition event [19]. Other shale rock fires have been observed in several regions in California [20], over the course of many years, and the cause of fire was not found but self-heating was not ruled out either.

Other shale fires can be found all over the world, with outcrop or formation fires reported over the last centuries in India, Russia, UK, Australia, USA and Greenland [21,22]. However, even with so many shale fires pointing to self-heating as the possible ignition event, self-heating of shale has never been thoroughly investigated until now.

For the first time in literature, this work experimentally studies the self-heating behavior of shale rock. The technique used for the self-heating study is known as oven-basket experiments [23]. This paper contributes to understanding and predicting the initiation of shale fires and related geological combustion processes [21] by finding the effective kinetics and thermal properties of shale.

2. Self-heating ignition theory

Frank-Kamenetskii theory is usually employed in the literature to investigate spontaneous ignition [4,24]. The theory allows to calculate ignition conditions from reactive properties like the activation energy and other physical parameters of the material such as the conductivity and the heat of reaction by finding the critical ambient temperature for a given sample size. Total heat production from reactions inside a material sample is proportional to its volume, but heat loss is proportional to its area. This means that as the size of the sample becomes larger, becuase volume increases with size faster than area, then the critical ambient temperature required for ignition decreases. The theory can therefore be used to predict spontaneous ignition for larger sizes at lower temperatures, provided that the mechanism of heat production is unchanged [4,24]. The heat transfer problem in this study corresponds to the transient heat conduction equation, shown in Eq. (1),

$$\nabla^2 T + \frac{QF(t)e^{-\frac{E}{RT}}}{k} = \frac{1}{\alpha}\frac{\partial T}{\partial t} \tag{1}$$

where T is the temperature of the fuel sample, E is the activation energy of the reaction, k is the conductivity of the fuel, R is the universal gas constant, Q is the heat of reaction per fuel mass, α is the thermal diffusivity of the fuel, t is time and $F(t)$ is the mass action law based on concentration of fuel and oxygen at any given time. There is no need to specify the dependency on concentration but an often invoked representation of this law is $[fuel]^a\,[O_2]^b$ [4].

Frank-Kamenetskii theory of ignition assumes that the material has a high reactivity and high activation energy so that a steady-state condition is reached [4,24]. To solve Eq. (1) at steady-state, Frank-Kamenetskii theory defines a dimensionless parameter δ,

$$\delta = \frac{QEfL^2e^{-\frac{E}{RT_a}}}{kRT_a^2} \tag{2}$$

where T_a is the ambient temperature and L is the characteristic length, half the smallest dimension of the fuel (for a cubic basket L is the side length, and for an infinite slab L is the thickness), f is the value of F(t) at initial time, so based on initial concentrations of fuel and oxygen [24]. Expressing the reaction rate as the Arrhenius law for dependence on temperature, Eq. (1) is solved at steady-state, and the following dependence of critical size and temperature is obtained, as shown in Eq. (3):

$$ln\left(\frac{\delta_c T_{a,c}^2}{L^2}\right) = \ln\left(\frac{QEf}{Rk}\right) - \frac{E}{R\,T_{a,c}} \tag{3}$$

where δ_c is the critical value of the dimensionless parameter in Eq. (2), and is used to relate the geometrical shape of the sample to the critical ambient temperature $T_{a,c}$ which corresponds to the minimum ambient temperature for which ignition of a given sample will occur. δ is a non-dimensional representation of the ratio of characteristic heating time to characteristic reaction time, so δ can be seen as a type of Damköhler number [23]. A solution to Eq. (3) satisfying the boundary condition $T=T_a$ on the wall(s) only exists when the condition $\delta \leq \delta_c$ is satisfied. Since δ_c is a function of geometry, this is found by looking up its value for the experiment geometry of interest in the literature [6,23,24]. In our experimental work we used cubic baskets, so δ_c=2.52 [24], and for geological formations and heaps we assume slab geometry which has δ_c=0.878 [24]. By plotting the experimental data of $ln(\frac{\delta_c T_{a,c}^2}{L^2})$ against $\frac{1}{T_{a,c}}$ (Eq. (3)), we obtain a correlation. If the correlation is a straight line, this validates the Frank-Kamenetskii theory. The