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Organic liquid mobility induced by smoldering remediation

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HIGHLIGHTS

- Organic liquid waste (in sand) can be destroyed vis self-sustaining smoldering.
- Under certain conditions the organic liquid migrates downwards into the reaction.
- Liquid migration depends on air flow rate, system height, and liquid viscosity.
- The migrating liquid influences, but does not penetrate, the smoldering reaction.
- Organic liquid migration can be prevented by applying a predictable air flow rate.

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ABSTRACT

Laboratory column experiments plus analytical and numerical modeling together suggest that, under certain conditions, downward organic liquid mobilization can occur and impact smoldering behavior. This applies for organic liquids mixed with inert sand subjected to smoldering as thermal treatment. The observed effects include increased peak temperatures (here by up to 35%) and increased treatment times (here by up to 30%). Downward organic liquid migration occurs when (i) injected Darcy air flux is less than a threshold value (here less than 3 cm/s), (ii) treatment systems are tall (here 90 cm, not 30 cm), and (iii) the organic liquid is temperature-sensitive (viscosity less than 0.01 Pa s at 150 °C). The developed analytical equation provides the applied air flux that can negate the downwards organic liquid gradient required for migration. Smoldering behavior is demonstrated to adjust to liquid migration and thereby still destroy all the organic waste in the system. Smoldering is a relatively new, energy-efficient thermal treatment for organic liquid waste and these results are important for designing field applications of smoldering treatment.

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1. Introduction

Lagoons containing hazardous, organic liquids are ubiquitous and present a significant environmental challenge [1,2]. Oil sludge associated with the petroleum industry, for example, is generated at a rate of 60 million tons per year and more than 1 billion tons have accumulated worldwide [2]. Coal tar, a byproduct of coal carbonization, is another example; in 2006 China alone produced 9×10^9 kg of coal tar, representing 3.8% of the coal feed stock [3]. These are both examples of organic liquid wastes from industrial activities that are stockpiled in large quantities in lagoons and for which there are no solutions that are cost effective and technically efficient [1].

Organic liquids, when embedded in a porous medium, can be destroyed via smoldering combustion. This concept was recently introduced as a promising remediation approach for contaminated soil [4,5]. Smoldering for the destruction of organic liquid wastes is possible but requires the liquids to be mixed intentionally with an inert porous matrix, such as sand [6]. While not participating directly in the reaction, the sand provides important physical characteristics, such as air permeability, as well as thermal properties, such as heat retention [7,8]. This facilitates the smoldering of the liquids embedded in the pores of the matrix, destroying the liquids and leaving clean sand behind [4].

Smoldering is a flameless, exothermic oxidation reaction occurring on the surface of a condensed-phase fuel [9]. Combustion requires the combined presence of heat, organic compounds, and oxygen and converts the latter two to primarily energy, water, and CO_2. The majority of smoldering research focused on solid fuels (e.g., polyurethane foam, coal, peat) in the context of fire safety

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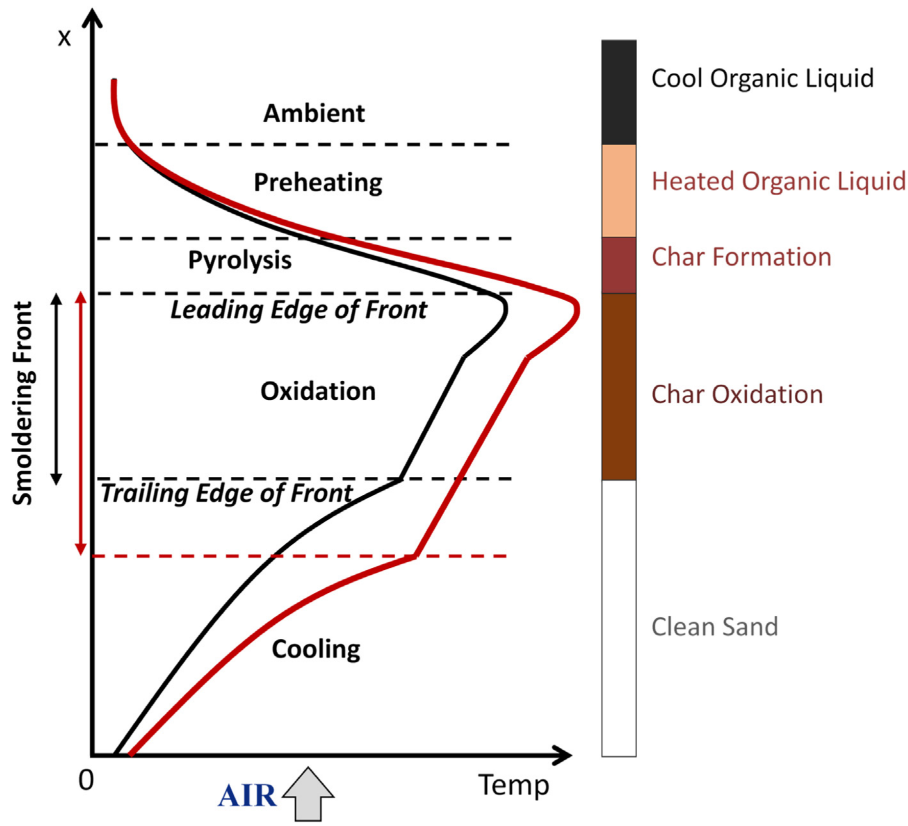

Fig. 1. Conceptual model of the distribution of temperature in a column experiencing upward, forward propagation of a smoldering reaction. The black line represents the standard model, while the red line represents how this changes in response to organic liquid migration (see Results and discussion). The key regions dividing the front and the dividing lines that bound the front thickness are named, and the form of the organic liquid associated with each region is identified on the right hand side. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

engineering [10,11]. Intentional smoldering of organic liquid/sand mixtures as a treatment method is a relatively new concept [6]. To ignite a smoldering reaction in these mixtures, a short and localized heat source is applied, followed by air injection [12]. Then the heat source is removed, since the organic liquid's combustion generates sufficient heat to make the reaction self-sustaining. The self-sustaining nature of the reaction underpins its potential for minimizing energy consumption and cost.

Smoldering remediation of contaminated subsurface soils has been demonstrated at the laboratory scale [4] and scaled-up as an in situ technology applied at contaminated sites [13]. Smoldering of organic liquid waste intentionally mixed with sand has been demonstrated at the scales of a laboratory column, an oil drum, and a pilot ex situ reactor [6]. The majority of this research has focused on engineering controls and process optimization, with less known about the underpinning processes. The potential for thermal mobilization of organic liquids has been extensively studied in the context of oil recovery [14] but it has never been studied for smoldering remediation. The time scales and structure of the oil recovery and smoldering thermal waves are different resulting in a very different balance of heat transfer modes [8]. This will affect thermal mobilization and motivate its study for the specific application of smoldering.

Smoldering is a complex phenomenon that depends on the balance between the rates and dominant modes of heat transfer and the rates and stoichiometry of suites of reactions [15,16]. The net effect is reflected in the temperature distribution, which is the main (and often only) metric available to monitor the smoldering reaction. Fig. 1 (black curve) illustrates the distribution of temperature for a typical smoldering reaction travelling upwards in a one-dimensional column. In this context, the column contains a high-viscosity organic liquid (e.g., coal tar, oil sludge) mixed intentionally with quartz sand to a specified saturation (e.g., 50%) with the remaining porosity occupied by air. Peak smoldering temperatures for these liquids are typically 700–1000 °C [6].

Of key interest in this work is the preheating region, which has temperatures in the range of 50–200 °C, where the sand/organic mixture is absorbing energy transmitted from the smoldering reaction below. Here exists the potential for organic liquid mobilization. Organic liquid is expected to migrate at a certain velocity (v_O) that, following Darcy's Law, depends on its viscosity and hydraulic gradient. At ambient temperatures, the high viscosity of long chain hydrocarbons means migration is slow even in the presence of significant hydraulic gradients [17]. However, liquid viscosity decreases rapidly with increasing temperatures [18]. Fig. 2 presents the sensitivity of liquid viscosity to temperature for a range of organic liquids used in this work and related studies. It reveals that viscosity may decrease by a factor of 10–1,000,000 over this temperature range, i.e., 50–200 °C, and the sensitivity is dependent on organic composition. Therefore, v_O may be significant in the preheating region.

Reduction of organic liquid viscosity leading to mobilization is intentional for applications such as in situ combustion for enhanced oil recovery [14] and steam injection for remediation [19]. In the context of smoldering destruction of organic liquids, any mobilization is unintentional. It is potentially undesirable and its effects are unknown. Moreover, it is impossible to monitor directly: there is no known technique for tracking organic liquid saturation in such hot systems. Any liquid mobilization is expected to be expressed in the temperature profile. A secondary impact could be the presence of residual organic remaining behind the front if mobilized liquid is able to penetrate the reaction into the normally clean sand below.

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